Non-linear wave phenomena in a gas-vapour mixture with phase transition

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NON-LINEAR WAVE PHENOMENA IN A GAS-VAPOUR MIXTURE WITH PHASE TRANSITION

PROEFSCHRIFT

ter verkrijging van de graad van doctor aan de Technische Universiteit Eindhoven, op gezag van de Rector Magnificus, prof.dr. J.H. van Lint, voor een commissie aangewezen door het College van Dekanen in het openbaar te verdedigen op dinsdag 30 juni 1992 om 16.00 uur

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1.1 Introduction

Compressible two-phase flow

Compressible flow is characterized by the central role that changes in the fluid density play in the dynamics of the fluid. However, the types of fluid motion thus classified are still diverse. One class is that of compressible flow for which an acceleration of the fluid is accompanied by marked adiabatic changes in the thermodynamic state of the fluid. Disturbances, i.e. infinitesimal wave elements changing the flow, travel with the local speed of sound relative to the flow. Since the velocity of the flow and the sound speed, depending on temperature, may vary greatly, the mutual distance of disturbances, being part of a wave of finite amplitude, may either increase or decrease, resulting in the flattening or steepening of wave fronts. The formation and existence of shock waves is one consequence of this non-linear behaviour of compressible flow.

When the compressible medium consists of a vapour or a gas-vapour mixture, then the flow may be heavily affected by the presence of the condensable component, due to the possible occurrence of a partial phase transition. For example, if a fluid element is cooled down in an adiabatic expansion, the vapour may easily become saturated. Equilibrium condensation would then occur, if nucleation sites were available and this state was to be remained for sufficiently long time. However, due to the rapidity of expansion, the vapour is cooled further into a metastable state. Eventually, at sufficient supercooling the breakdown of the metastable state will result in non-equilibrium condensation. The instant and mechanism of collapse depend on physical circumstances. Dealing with ultra-pure gas mixtures, condensation can only be initiated by spontaneous clustering of vapour molecules, forming stable liquid nuclei. This process, called homogeneous nucleation, is only effective at high supersaturation. More often, a gas flow contains impurities, which may be ions, salt crystals, aerosols, small particles originating from walls, or remnants of droplets remaining from former stages of expansion and compression of the flow. These impurities then serve as catalysing centres for condensation. This so called heterogeneous nucleation process becomes active already at supersaturations close to the saturated state.
After the occurrence of nucleation and condensation of vapour, a finely dispersed phase of liquid droplets has formed in the gaseous carrier. Mostly, the droplets are so small that they can be considered stagnant with respect to the carrier gas. Further, every change of state of the gas-vapour mixture will induce a partial change of phase in order to preserve equilibrium. The effect of phase change on the flow is twofold. Generally, the least significant effect is that the thermal properties of the fluid change due to the change in the composition of the fog. More importantly, latent heat is added to or subtracted from the flow, depending on the direction of phase transition. Alternatively stated, both features affect the compressibility of the fluid and therefore influence the flow. The direction of influence can easily be argued. Consider an isentropic compression of a gas-vapour mixture with droplets. Without phase change, this would lead to a temperature increase. However, to maintain phase equilibrium, liquid will evaporate, thereby subtracting heat from the system. Consequently, phase transition gives a reduction in the rise in the temperature. Therefore, the rise in the pressure is less, and effectively, a two-phase mixture is more compressible.

To understand a final feature of two-phase flow, we have to look at the microscale of a droplet. The actual phase transition takes place at the surface of the droplet. This means that vapour and heat have to be transferred to or from the droplet surface, which can only occur at a finite rate, depending on the degree of non-equilibrium. Indeed, compressible two-phase flow is characterized by non-equilibrium processes and thus shows relaxation behaviour. A typical example is condensation relaxation of an expanding flow after nucleation. The high supersaturation built up by the nucleation threshold is reduced by condensation at a rate depending on the degree of non-equilibrium. Another example is a shock wave in a fog. Suspended droplets pass the shock wave undisturbed, whereas the carrier gas is compressed and heated. The result is an unsaturated vapour, and again, the created non-equilibrium state relaxes by evaporation.

Non-linear wave phenomena in fluids with internal relaxation due to non-equilibrium condensation and evaporation processes are encountered in many technological and scientific environments. Examples are the unsteady periodic wave patterns that occur in steam turbines and combustion engines, or vapour explosions due to gas-tanker or gas-transport pipe-line accidents. In these cases, the wave phenomenon itself is the object of interest. However, steady and unsteady gasdynamic flow can also be used as an experimental tool to create rapid changes in the thermodynamic state of a vapour or a gas-vapour mixture in order to study nucleation phenomena or to investigate droplet growth and evaporation.
Outline of the thesis

This dissertation reports on an experimental and theoretical investigation of two types of gasdynamic flow with phase transition. The flow phenomena studied are: 1. the unsteady non-linear rarefaction wave in a gas-vapour mixture with stimulated condensation on foreign particles added to the mixture, and: 2. the shock wave in a fog, which causes the evaporation of droplets that are present. The intention of the study was to investigate the structure of these types of waves, influenced by the phase-transition process, and to establish the validity of models to describe droplet formation and growth.

The remaining sections of Chapter 1 are devoted to a general introduction to the subjects of expansion flows with condensation and to shock waves in gas-vapour-droplet mixtures. In Chapter 2 attention is paid to the thermodynamics and internal dynamics of a gas-vapour mixture with or without droplets. A simple model for catalysed condensation on insoluble particles is proposed and the wet-bulb model for droplet growth is explained. Chapter 3 deals with the modelling and the theoretical aspects of an unsteady rarefaction wave in a gas-vapour mixture. A simple analysis is performed to illustrate some basic features of unsteady relaxing expansion flow and a study is reported on the asymptotic self-similar solution, that develops when time exceeds the characteristic time of relaxation by orders of magnitude. Chapter 4 goes into the matter of shock waves in gas-vapour-droplet mixtures. The relaxation processes that occur behind a shock wave are identified and analysed, and based on that analysis, a simplified model is proposed to describe shock-wave induced evaporation. The purpose of Chapter 5 is to introduce a version of the numerical Random-Choice method, adapted for the computation of unsteady wave phenomena in gas-vapour mixtures with condensation and evaporation. In Chapter 6 a description is given of the experimental methods. The principle of the wave experiment is explained, the experimental set-up is described and the measuring techniques are considered in some detail. The experimental results of this investigation are presented in Chapter 7. Observations on the unsteady expansion of a gas-vapour mixture and the structure of the dispersed shock wave are reported, and measured droplet evaporation rates are compared to theoretical predictions by the wet-bulb model. The numerical results are described in Chapter 8. A numerical simulation of the wave experiment, compared with actual measurements, is presented and the asymptotic behaviour of the unsteady rarefaction wave in a gas-vapour mixture is studied. Finally, Chapter 9 contains a concluding discussion.
1.2 Expansion flows with condensation

Adiabatic expansion

The fast adiabatic expansion of a vapour in a carrier gas results in an isentropic change of state, which may produce a supersaturated state, depending on the density reduction and the initial state of the vapour. If a state of sufficiently large supersaturation is reached, a significant number of small liquid droplets, mostly referred to as nuclei, will form due to a nucleation process. The growth of these nuclei by condensation results in the depletion of vapour and the release of latent heat. The condensation process can be considered as a non-equilibrium relaxation process, heading towards the establishment of a new equilibrium state of droplets in a saturated vapour environment.

![Diagram illustrating adiabatic expansion and condensation](image)

**Fig. 1.2.1.**

Adiabatic expansion of a vapour, depicted in the diagram of vapour pressure versus temperature.

Symbols: L-V: liquid-vapour coexistence line, I: isentrope, II: path of relaxation, 0: initial state, S: saturated state, C: condensation onset, E: equilibrium state.

The complete process is shown in the vapour-pressure versus temperature diagram of Fig. (1.2.1). Starting at the initial point 0, the vapour pressure and the temperature decrease along the line of isentropic expansion until the vapour gets saturated at the point of intersection S with the line of coexistence of vapour and liquid. However, since we are dealing with a fast expansion, condensation will not set in at this point. On the contrary, the expansion continues until the collapse of the metastable vapour state is observed at the condensation onset point C, where a nucleation process has resulted in the formation of a significant amount of stable nuclei. The growth of the nuclei by condensation then introduces a non-equilibrium relaxation process towards the saturation curve.

Nucleation may either occur by catalysed condensation on impurities that are
present in the mixture and act as condensation centres, i.e. heterogeneous nucleation, or by spontaneous clustering of vapour molecules, i.e. homogeneous nucleation. The conditions required to activate these nucleation processes are quite different. Heterogeneous nucleation already occurs at small supersaturations close to saturation and therefore results in a rather gradual relaxation process. Opposed to this, homogeneous nucleation becomes manifest only at high supersaturation and results then in the formation of a very large number of nuclei, inducing rapid return to the equilibrium state.

The exact route of relaxation and the final state cannot be specified without additional knowledge on the expansion process. For example, the adiabatic expansion may be due to the gasdynamic flow of a vapour or a gas-vapour mixture. In such a flow, the acceleration of the fluid leads to a considerable adiabatic variation in the state of the compressible medium. As a result, in the case of expansion flows the vapour may condense into droplets as explained above. Depending on the amount of condensation, the flow is appreciably influenced due to the addition of latent heat to the flow and the subtraction of vapour from the flow. In the next two subsections, a short presentation follows of the features of some gasdynamic flows with condensation. Reviews on the subject are written by Wegener (1969, 1975), Wegener and Wu (1977), and Kotake and Glass (1981).

Nozzle flows

As can be read in the historical notes presented in a review by Wegener (1975), the interest in gasdynamic flow with condensation started with Prandtl in 1935, reporting a then unidentified shocklike phenomenon observed in the diverging part of a Laval nozzle, as sketched in Fig. (1.2.2). Soon, it became apparent that the phenomenon, referred to as an X-shock because of its two-dimensional shape, was due to the condensation of water vapour present in the undried air supplied to the nozzle. Hermann (1942) investigated the influence of different operating conditions on the structure and the position of the X-shock. He found that both increasing humidity and increasing cooling rate, which is determined by the geometry of the nozzle, affected the X-shock by shifting it towards the nozzle throat and by flattening its X-shape towards a normal shock structure. Oswatitsch (1942) was the first to give a full physical explanation of the phenomenon. He explained the sudden disturbances in the flow field behind the nozzle throat by the release of latent heat due to the growth of a large number of small droplets after breakdown of the supersaturated vapour state as a result of homogeneous nucleation. A simple
estimate performed by Oswatitsch excluded the possibility of marked condensation on foreign particles, undoubtedly present in the supplied air. Although heterogeneous condensation starts almost immediately after the saturation condition is reached, already in the subsonic section of the nozzle, its effect on the saturation ratio is completely insufficient to prevent spontaneous condensation from occurring due to the high rate of expansion. Indeed, Buckle and Pouring (1965) confirmed by experiment that heterogeneous nucleation is a minor effect in regular nozzle flows, unless particles are added to the flow in a very high concentration (above $10^{14}$ m$^{-3}$). But even in that case, the phenomenon is determined by spontaneous condensation. The virtual effect of the presence of foreign particles is only to apparently lower the humidity of the supplied air.

From a gasdynamic viewpoint, the flow can be interpreted as a diabatic flow, i.e. a flow with heat addition. First, the observation is made that from the point of onset the condensation process very rapidly decreases the existing high supersaturation and induces a situation of near-equilibrium. Therefore, the non-equilibrium condensation zone is fairly narrow and can be modelled as a Rankine-
Hugoniot transition of a perfect gas with external heat addition (Belenky, 1945; Heybey, 1947; Stever, 1958). Then, the pressure and temperature jumps and the change in the Mach number across the transition zone can easily be calculated. As discussed by Heybey (1947), the condensation transition can be categorized as a weak compression discontinuity with supersonic flow in front and behind. He also found a maximum value for the heat that can be added to the flow, given a fixed value of the pre-transition Mach number. This critical case corresponds to a Chapman-Jouguet transition with sonic outflow.

Some remarks must be made. Firstly, note that this model does not provide a decision on the amount of heat added to the flow by the condensation process. And secondly, although discontinuity theory is applied to the condensation zone and this zone is often referred to as a condensation shock, it is important to realize that the relaxation really takes place as a continuous transition from a state of dry gas to a near-equilibrium state with condensed phase in a zone of finite extent (Wegener, 1954). Gasdynamic shocks may be part of the solution but are not inherent to it.

It is interesting to see what happens when the humidity of the supplied air is increased. Starting at low humidity, the condensation zone lies at a relatively large distance from the nozzle throat. The amount of heat released is moderate, so the change in the Mach number through the zone towards unity is only small. Increasing the humidity gives a shift of the condensation onset point towards the throat, thus decreasing the pre-transition Mach number, and an increase of the heat addition since more vapour has to condense. The consequence is, as can be seen from diabatic flow theory (Stever, 1958), that the Mach number approaches unity behind the zone. The case where the post-zone Mach number equals unity corresponds to the Chapman-Jouguet transition and the heat addition is critical. Increasing the humidity of the supplied air still further, causes the flow to be thermally choked (Pouring, 1965; Barschdorff, 1970). Then, a gasdynamic shock wave develops inside the condensation zone, thus quenching the condensation heat release. Again increasing the humidity, results in an unsteady flow, since every newly generated shock wave runs out of the condensation zone through the nozzle throat without finding a stable position (Barschdorff, 1970). A detailed investigation on the influence of humidity, taking into account two-dimensional structures, is presented by Matsuo et al. (1985) and Schnerr (1989).

Finally, combined with light scattering techniques the nozzle has proven to be an outstanding instrument to study homogeneous nucleation and to determine condensation onset points (Wegener, 1977; Kotake & Glass, 1981; Ehrler, 1988).
Unsteady expansion flows

Whereas the gasdynamic flow in a Laval nozzle is steady, an unsteady expansion flow can be created by using a shock tube or a Ludwieg tube. For example, consider a configuration as sketched in Fig. (1.2.3). A tube, filled with a pure vapour or a gas-vapour mixture, is connected to a large evacuated vessel by a closed diaphragm. Opening the diaphragm results in an unsteady rarefaction wave, originating at the diaphragm and travelling into the mixture. Inside the wave, the mixture, initially at rest, is expanded adiabatically and is accelerated towards the vacuum vessel. Due to the choking condition, the flow is sonic at the connecting orifice, thereby controlling the depth of expansion. As the mixture is expanded, it cools down adiabatically, and if the critical condition is reached, vapour condenses into droplets. Condensation may influence the flow considerably by heat addition and vapour depletion.

Assume for a moment that condensation is prevented from occurring, i.e. the flow is frozen. Then first of all, the changes of state are isentropic throughout the wave. Further, the rarefaction wave is represented in the space-time diagram by a centred fan of characteristics, which correspond to lines of invariance of state. An illustration is given in Fig. (1.2.3). At both sides of the fan, a region exists of constant state. This particular type of solution is denoted as a self-similar solution. It inhibits a linear scaling of the extent of the rarefaction wave with time. This implies that gradients inside the wave weaken with increasing time.

Now that some basic properties of a rarefaction wave are discussed, we return to the case of induced condensation. Assume that the vapour is unsaturated initially. Then, a flow element that is accelerated along a particle path experiences a change of state as indicated in Fig. (1.2.1). This means that at some instant of time at some position inside the wave the saturation condition is attained inside this particular flow element. From that point on supersaturation will increase until it becomes sufficiently high to initiate significant condensation. This point at which condensation can actually be registered, either by a direct observation of the droplets or by a measurement of the deviation of the mixture state from the isentropic value, is called the onset point. Although condensation may certainly exist from the saturation point up to the onset point, it is of minor significance, so that the flow up to the onset may be treated effectively as frozen and isentropic. Therefore, it can easily be understood, that the saturation line S in the wave diagram is a straight line through the origin. The shape of the onset line C is determined by nucleation and droplet-growth processes and their dependence on the state of the mixture. The trend of decreasing gradients with increasing time will
Fig. 1.2.3. Unsteady rarefaction wave with condensation. The pressure history at the window is indicated.


change the time interval between saturation and onset towards larger values. However, its increase appears to be less than proportional to time, resulting in a curvature of the onset trajectory towards the front of the wave.

Behind the onset curve the zone of non-equilibrium relaxation extends, wherein droplets grow until an equilibrium is attained. In this zone condensation acts upon the flow by heat addition and vapour depletion, inducing disturbances that travel up- and downstream along the characteristic trajectories. First of all, this implies that the condensation processes taking place in different fluid particles influence each other. Or alternatively stated, the evolution of condensation inside a particular fluid particle is heavily depending on the history of the relaxing flow. A further consequence is that the characteristics are no longer straight lines but have the tendency to curve. In gas-vapour mixtures, the addition of heat to the flow is the dominant effect, resulting in the emission of compression disturbances that lead to the merging of characteristics. As a consequence, shock waves may appear in the condensation zone.
The first study of condensation in an unsteady rarefaction wave in a shock tube dates from 1951 and was performed by Wegener and Lundquist. They managed to delay water vapour condensation in air down to temperatures below 150 K. Then, condensation occurred by homogeneous nucleation. The structure of shock tube flow with saturated air as driver gas was investigated later by Glass and Patterson (1955) by means of schlieren photography. Beside shock wave, contact surface and rarefaction-wave head, the pictures clearly showed the path of condensation onset and an opaque region due to light attenuation by existing droplets.

A quantitative approach to study condensation onset in an unsteady rarefaction wave was made by Kawada and Mori (1973), using several refrigerant vapours of which some were mixed with an inert gas. The onset was detected by measuring light transmission and a comparison was made with the classical homogeneous-nucleation theory. By adjusting the theoretical predictions to their observations, it turned out for the majority of condensable substances studied, that the estimated values for the surface tension were smaller than the macroscopic values.

Barschdorff (1975) and Lee (1977) used an identical technique to investigate condensation kinetics in a shock tube. The experimental condition, i.e. the depth of the expansion, was chosen in such a way that the condensation onset coincided with the tail of the rarefaction wave exactly at the observation station. Thereupon, the onset was registered by means of a measurement of the pressure and of light scattering. To calculate the state at onset, both authors used the method of Wu (1977), performing an integration of the rate equations along a particle path assuming an undisturbed centred rarefaction wave. Barschdorff (1975) focused on the effect of the carrier gas on homogeneous nucleation of water vapour. He found no dependence on the type of carrier gas. However, an increasing deviation from the classical theory was observed with a decreasing vapour mass fraction. Lee (1977) studied the condensation onset of H₂O and D₂O vapour at slightly lower cooling rates. His results were in agreement with those of Barschdorff.

Experiments on the unsteady expansion of a water vapour-carrier gas mixture in a shock tube, concerning the relation between the position of the onset point inside the wave and the expansion rate were performed by Kalra (1975). He assumed a power law dependence of supercooling on cooling rate and obtained the coefficients by monitoring the onset event at two different observation stations. The forward curvature of the onset trajectory as shown in Fig. (1.2.3) was confirmed. Different approaches to provide a physical foundation to Kalra’s experimental findings were made by Sisilian (1975) and Kotake and Glass (1976). Sisilian used the classical model for homogeneous nucleation and a free-molecular-
flow model for droplet growth, omitting droplet surface-tension effects, to calculate the non-equilibrium condensation inside the flow field. The features of the expansion flow he predicted are in accord with those discussed in the beginning of this section. An important parameter to quantitatively modify the calculated flow was the surface tension of the condensable substance. However, although his calculations approached the observations of Kalra when reducing the value of the surface tension from its macroscopic value, satisfactory quantitative agreement was not obtained. Kotake and Glass presumed that, despite of filtration, the carrier gas in the experiments of Kalra contained a large number of foreign particles and replaced the model of homogeneous nucleation in the calculations of Sislian by a heterogeneous-nucleation model. Again the flow field had similar features. However, the contact angle of the cluster and the substrate was now used as a parameter to match the experiments of Kalra.

Recently, Spiegel (1987) operated the shock tube as an experimental tool to investigate binary nucleation, and Peters and Paikert (1989) used the unsteady gasdynamic flow inside a shock tube in a refined experimental procedure to actually measure nucleation rates as a function of the onset conditions.

1.3 Shock waves in gas-vapour-droplet mixtures

The shock wave and its relaxation zone

The propagation of a shock wave in a gas-vapour-droplet mixture is strongly affected by the presence of the droplets. Behind the shock wave a zone extends in which the mixture, brought into non-equilibrium by the shock wave, relaxes to a new equilibrium state. In the case that the shock wave propagates stationary in a uniform fog, the flow phenomenon is steady in the reference frame fixed to the shock wave.

Consider the shock wave and its steady relaxation zone in the shock-fixed reference frame of Fig. (1.3.1). The mixture of carrier gas and droplets, assumed to be at equilibrium, enters the shock front at supersonic speed. An abrupt deceleration of the gas phase to a subsonic speed occurs simultaneously with an increase of gas temperature, pressure and density. The droplets pass the shock wave undisturbed, retaining their initial velocity, temperature and size. The shock transition in the vapour-pressure versus temperature diagram is shown in Fig. (1.3.2). The pre-shock state is situated on the liquid-vapour equilibrium line. Across the shock, the vapour pressure increases proportionally to the pressure. The rise in
the temperature is accompanied by a rise in the saturation pressure of the vapour, which exceeds the change in the vapour pressure. As a result, the post-shock vapour state is unsaturated. Thus, a situation of non-equilibrium is induced by the shock wave.

By exchange of momentum, heat and mass between the droplets and the gas, the system tends towards a new equilibrium, as shown in Fig. (1.3.1). To describe the mechanisms involved, we shall consider the case that the droplet mass fraction is low, so that the state of the gas phase is unaffected by the relaxation processes. Then, the droplets decelerate to the constant gas velocity by the influence of drag. The restoration of equilibrium by heat and mass transfer is a subtle process, which may be explained with the help of Fig. (1.3.2). Initially, the droplets have the undisturbed temperature $T_1$. So, the droplets are cold compared to their heated environment ($T_1 < T_2$). Moreover, the compressed vapour is supersaturated with respect to the temperature of the droplets ($p_{v2} > p_{v1}$). Therefore, heat will be transported towards the droplets and vapour will condense on the droplets, thereby releasing heat. Both processes will result in a rapid increase of the droplet temperature. After having travelled a certain distance through the relaxation zone, the droplets will reach the saturation temperature of the vapour ($T_d = T_b$). At that point the condensation process turns into an evaporation process. Then, part of the heat transferred to the droplets will be absorbed by the evaporating liquid and the rise in the droplet temperature slows down. At a particular temperature, all the supplied heat will be used for evaporation and the droplet temperature becomes steady. This temperature is called the wet-bulb temperature. The remaining part of
the evaporation process then takes place at wet-bulb conditions.

In the description above the state of the gas phase was unaffected by the relaxation processes, since it was assumed that the droplet mass fraction is low. In the case that the mass fraction of liquid is high, the gas state variables will change in the relaxation zone due to a significant transfer of momentum, heat and mass. Especially, the temperature decrease and the vapour pressure increase may be such that finally a new phase equilibrium will develop without complete evaporation of the droplets.

\[ \text{Fig. 1.3.2.}
\]

\textit{Shock transition in the vapour-pressure versus temperature diagram.}

\textit{Symbols: L-V: liquid-vapour coexistence line, 1: pre-shock state, 2: post-shock state, }\textsubscript{pvs} \textit{saturation vapour pressure for post-shock state, }T_s \textit{saturation temperature for post-shock state.}

\textit{Review of the literature}

The phenomenon of a shock wave propagating in a suspension of droplets in a gas is closely related to that of a shock wave in a dusty gas. In the latter case, however, only momentum and heat is transferred between the phases. A first theoretical analysis on this subject was presented by Carrier (1958). He combined the steady conservation equations and the rate equations for momentum and heat transfer in a suitable form for calculation and presented some solutions. Marble (1962) introduced characteristic lengths for the relaxation of velocity and temperature, and investigated the manner in which these characteristic lengths and the particle mass fraction each influence the structure of the solution. An extensive discussion of the behaviour of the flow variables in the relaxation zone was presented by Rudinger (1964).

In addition to momentum and heat transfer, mass transfer between the phases was first taken into account by Nayfey (1966) and Lu and Chiu (1966). The
problem of ablating teflon particles in a gas behind a shock wave was considered by Nayfey. The set of equations was formulated using the transfer properties for free molecular flow. Numerical examples were presented to show the effect of relevant parameters of the problem. Lu and Chiu computed the relaxation zone of a mixture composed of air, water vapour and water droplets. Moreover, they distinguished between the case of complete evaporation and the case of evaporation towards a final two-phase equilibrium state.

Attention to the characteristic times of the phenomena that control the relaxing flow behind a shock wave was given by Marble (1969). However, the times he derived belonged to the isolated transfer processes of momentum, heat and mass, thus ignoring the effect of combined action. More appropriate characteristic times were introduced by Goossens et al. (1988) for the flow of gas-vapour-droplet mixtures and by Guha and Young (1989) for wet-steam flow. It was argued that the relaxation behind a shock wave can be considered to take place in two stages. The first stage consists of a rapid change of droplet temperature to the quasi-steady wet-bulb temperature, following from a balance of the heat supplied to the droplet and the latent heat absorbed by the evaporating liquid. In the case of a pure vapour, the evaporation rate is determined mainly by the heat flow resistance, so the wet-bulb temperature then equals the saturation temperature of the vapour. During the second stage, the evaporation process continues while the droplet temperature remains close to the dynamic equilibrium value.

Experimental work on the behaviour of wet steam after disruption by a shock wave has been reported by Yousif and Bakhtar (1974). After the supercooling and nucleating stages of a steam flow in a Laval nozzle, the two-phase mixture was led through a standing shock wave in the diverging part of the nozzle. A comparison of measured and theoretical pressure profiles gave reasonable agreement. Hastings and Hodgson (1979) made some preliminary observations of a shock wave in an aqueous fog in a shock tube. They observed a considerable dispersion of the shock wave.

A study of weak shock waves in a fog was made by Goossens et al. (1986). They used a combined expansion-shock tube. A mixture of nitrogen gas and water vapour, provided with condensation nuclei, was first expanded by unsteady expansion waves, resulting in the formation of a stagnant fog. Then, the mixture was recompressed and heated again due to the passage of a shock wave. It was shown by means of laser Mach-Zehnder interferometry that the shock front in a fog consists of a frozen part in which no evaporation takes place followed by an evaporation zone. Later, Goossens et al. (1988) presented experimental data on the characteristic evaporation time of droplets as a function of the shock strength.
That a shock wave can be used to determine diffusion coefficients and correction factors for the transition regime of intermediate Knudsen numbers was convincingly demonstrated by Roth and co-workers. The conditions were chosen such that the characteristic time of evaporation was much larger than the characteristic times of momentum and droplet-temperature relaxation. Moreover, the aerosol mass fraction was taken low, so the gas-phase variables remained constant during relaxation. Then, evaporation rates were measured for the flow behind the shock wave using light-scattering techniques. For the substances DOP and DIDP in argon, Roth and Fischer (1985) obtained in this way mass-flux correction factors for the transition regime that could be compared with theoretical models. Also, Timmier and Roth (1989) determined the temperature dependence of the diffusion coefficients for DOP, KBr and silicon oil PD5 in argon. The results for silicon oil indicated the presence of two compounds in the droplets.

Paikert (1990) experimentally studied the evaporation of water and hydrocarbon droplets in air and argon due to a reflected shock wave at the end wall of a shock tube. The micron-sized droplets were generated by homogeneous nucleation in an adiabatic unsteady expansion wave. Since the experimental observations took place at the end wall of the tube, droplets and gas were stagnant during evaporation. Consequently, the evaporation of the droplets was not affected by a velocity difference between the droplets and the gas. The experimental results could very well be explained by the droplet-growth model of Gyarmathy (1982).

A review of the literature up to 1977 on shock-wave chemistry in dusty gases and fogs has been given by Nettleton (1977).
Chapter 2. Thermodynamics and internal dynamics of a gas-vapour mixture with or without droplets

2.1 Introduction

As explained in the preceding chapter, compressible flow of a gas-vapour mixture is strongly affected by internal processes that occur in a fluid sample. Therefore, we shall proceed with a discussion of the thermodynamics and internal dynamics of a gas-vapour mixture with or without droplets. To begin with, we shall catalogue the equations that describe the state of the mixture. This will be followed by a consideration on the formation of droplets, resulting in a simple model for condensation on insoluble particles. Then, a basic description of the problem of a droplet in a gaseous carrier stream will be given, adapted to the needs of the present investigation. Finally, the last section shall contain a comparison of several droplet-growth models known in literature. In our discussion of the matter, notation and methodology will be adopted from Gyarmathy (1982).

2.2 A description of the mixture

The constituents

Consider a gaseous mixture of vapour and inert gas, existing at a temperature $T$ and partial densities $\rho_g$ and $\rho_v$, respectively. Then, by assuming that the gas and the vapour are thermally perfect gases, the partial pressures are given by:

\[ p_g = \rho_g R_g T, \quad (2.2.1) \]
\[ p_v = \rho_v R_v T, \quad (2.2.2) \]

where $R_g$ and $R_v$ are the specific gas constants of the gas and the vapour, respectively. Moreover, assuming that both constituents are calorically perfect, the specific internal energy $e$ and the specific enthalpy $h$ of the gas and the vapour have a linear dependence on the temperature:

\[ e_g = c_{vg} T, \quad h_g = c_{pg} T, \quad c_{pg} = c_{vg} + R_g, \quad (2.2.3) \]
\[ e_v = c_{vv} T + L_0, \quad h_v = c_{pv} T + L_0, \quad c_{pv} = c_{vv} + R_v, \quad (2.2.4) \]
where \( c_{vi} \) and \( c_{pi} \) are the specific heats of the gaseous constituent \( i \) at constant volume and pressure, respectively, and \( L_0 \) is the latent heat of evaporation at zero temperature.

Further, if droplets are present in the mixture of vapour and inert gas, then the droplets are assumed to be composed of a single, incompressible liquid with a density \( \rho_L \) and a constant specific heat \( c_L \). At room temperature and atmospheric pressure, it is allowed to make the simplifying assumption \( p/\rho_L < c_L T \) for liquid water, where \( p \) is the pressure and \( T \) is the temperature of the liquid. Then, the specific internal energy \( e_L \) and the specific enthalpy \( h_L \) of the liquid are identical in good approximation and are given by:

\[
e_L = h_L = c_L T. \tag{2.2.5}
\]

**The liquid-vapour interface**

By definition, we have for the latent heat of evaporation at the temperature \( T \):

\[
L \equiv h_v - h_L = (c_{pv} - c_L) T + L_0. \tag{2.2.6}
\]

For water under atmospheric conditions it is allowed to use \( \rho_v < \rho_L \), so that the equilibrium of liquid and vapour is described by the equation of Clausius-Clapeyron in differential form:

\[
\frac{dp_{VS}}{dT} = \frac{L_{p_{VS}}}{R_v T^2}, \tag{2.2.7}
\]

where \( p_{VS} \) is the saturation vapour pressure. Its temperature dependence can be obtained by integration of the last equation, after substitution of Eq. (2.2.6). The resulting equation is called the Rankine-Kirchhoff equation:

\[
p_{VS}(T) = p_{VS}(T_{ref}) \left( \frac{T}{T_{ref}} \right)^{\frac{c_{pv} - c_L}{R_v}} \exp \left( \frac{L_0}{R_v} \left( \frac{1}{T_{ref}} - \frac{1}{T} \right) \right), \tag{2.2.8}
\]

where the subscript \( ref \) pertains to a reference state. If the liquid phase exists in the form of a spherical droplet with radius \( r_d \), then the equilibrium vapour pressure will be different, due to the curvature of the interface:

\[\text{We shall refer to the influence of the surface tension on the equilibrium of liquid and vapour as the Kelvin effect.}\]
\[ p_{ve}(T, r_d) = p_{vs}(T) \exp \text{Ke}(T, r_d), \]  
(2.2.9)

with the Kelvin number defined as:
\[ \text{Ke}(T, r_d) = \frac{2\sigma}{\rho T_v r_d}. \]  
(2.2.10)

Here \( \sigma \) is the surface tension of the liquid. It may be noted that subscript \( s \) refers to saturation, i.e. a phase equilibrium at a flat liquid-vapour interface, while subscript \( e \) refers to an equilibrium at a curved interface.

**Characterization of the mixture**

The equations characterizing the state of the mixture will now be catalogued. To cover the most general case, we shall presume that droplets are present. The state of the gas phase is completely specified by four parameters, say the velocity \( u \), the temperature \( T \), the pressure \( p \) and the partial vapour pressure \( p_v \). An important parameter, describing the degree of non-equilibrium, is the saturation ratio \( \chi \), defined as the ratio of the vapour pressure and the saturation vapour pressure at the temperature of the vapour:
\[ \chi = \frac{p_v}{p_{vs}(T)}. \]  
(2.2.11)

The droplets are assumed to be spherical with a radius \( r_d \) and a mass \( m_d \):
\[ m_d = \rho \frac{4}{3} \pi r_d^3. \]  
(2.2.12)

Their sizes will in general be different, as will be characterized by the size distribution function \( F(r_d) \). Average droplet quantities are defined as:
\[ \langle Z \rangle = \int_0^\infty Z(r_d) F(r_d) \, dr_d, \]  
(2.2.13)

where \( Z \) is an arbitrary quantity. The velocity \( u_d \) and the temperature \( T_d \) of a droplet are depending on the droplet radius, so appropriate average quantities can be calculated with Eq. (2.2.13). Let the number of droplets per unit volume of the fog be denoted by \( n_d \), then the accumulated mass density of the droplets is defined as:
\[ \rho_d = \frac{n_d \rho}{\frac{4}{3} \pi \langle r_d^3 \rangle}. \]  
(2.2.14)
It is now assumed that $\rho_d \ll \rho_L$, so that the total volume occupied by the droplets can be neglected. With the partial densities $\rho_g$ and $\rho_v$ of the gas and vapour, respectively, the total density of the fog can be written as:

$$\rho = \rho_g + \rho_v + \rho_d = \rho_c + \rho_d,$$  \hspace{1cm} (2.2.15)

where we have simultaneously introduced the density of the carrier $\rho_c$. Moreover, mass fractions of constituent $i$ can be defined as:

$$f_i \equiv \frac{\rho_i}{\rho},$$  \hspace{1cm} (2.2.16)

$$g_i \equiv \frac{\rho_i}{\rho_c}.$$  \hspace{1cm} (2.2.17)

We introduce the following mass-weighted quantities:

$$R' \equiv g_g R_g + g_v R_v,$$  

$$c'_p \equiv g_g c_{pg} + g_v c_{pv},$$  \hspace{1cm} (2.2.18)

$$c'_v \equiv c'_p - R',$$  

$$\gamma' \equiv \frac{c'_p}{c'_v},$$

and:

$$\bar{R} \equiv f_g R_g + f_v R_v,$$  

$$\bar{c}_p \equiv f_g c_{pg} + f_v c_{pv} + f_d c_d,$$  \hspace{1cm} (2.2.19)

$$\bar{c}_v \equiv \bar{c}_p - \bar{R},$$  

$$\bar{\gamma} \equiv \frac{\bar{c}_p}{\bar{c}_v}.$$

By assuming that the droplets exert no pressure forces, the total pressure can be written according to Dalton's law as:

$$p = p_g + p_v = \rho_c R'T = \rho \bar{R}T.$$  \hspace{1cm} (2.2.20)

The pressure $p_d$ inside the droplets is higher than the pressure in the gas, due to the action of the surface tension at the curved interface of the droplets:

$$p_d = p + 2\sigma/r_d.$$  \hspace{1cm} (2.2.21)

The surface tension $\sigma$ is assumed to be independent of the droplet radius. Its dependence on the temperature is given for water in Appendix A. Finally, we can write for the specific internal energy $e$ and the specific enthalpy $h$ of the fog:

$$e = (f_g c_{vg} + f_v c_{vv}) T + c_v <f_d T_d> + f_v L_v,$$  \hspace{1cm} (2.2.22)

$$h = (f_g c_{pg} + f_v c_{pv}) T + c_v <f_d T_d> + f_v L_v.$$  \hspace{1cm} (2.2.23)

---

3 Typically, $\rho_d/\rho_L \approx 4 \times 10^{-3}$ for $r_d = 1 \mu m$ and $n_d = 10^{12} \text{m}^{-3}$. 

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2.3 The formation of droplets

A barrier to droplet formation

Whether droplets will be formed in a mixture of gas and vapour, depends on the saturation ratio $\chi$. Eventually, any degree of supersaturation ($\chi > 1$) leads to condensation. However, the formation of a small but stable droplet is hindered by the surface tension $\sigma$. This is illustrated by the equation for the free energy $\Delta F$ of formation of an unbound droplet with radius $r_d$ in an environment of temperature $T$ and saturation ratio $\chi$ (Abraham, 1974):

$$\Delta F = -\frac{4}{3} \pi r_d^3 \rho_v \xi_T T \ln \chi + 4 \pi r_d^2 \sigma,$$

from which it can be seen that $\Delta F$ has a maximum:

$$\Delta F^* = \frac{4}{3} \pi r^*^2 \sigma,$$

at a critical radius $r_d^* - r^*$ of:

$$r^* = \frac{2 \sigma}{\rho_v \xi_T T \ln \chi}.$$

By comparison with Eqs. (2.2.9) to (2.2.11), it follows that a droplet of critical size is in equilibrium with the vapour. However, this equilibrium is unstable. Droplets with a radius smaller than the critical radius will disappear, larger droplets will grow. Only due to a statistical-kinetical nucleation process, clusters of vapour molecules are formed with a radius larger than the critical radius. Then, subsequent
condensation can take place on these clusters.

The formation of a droplet is considered to occur by the impingement of a vapour molecule on a critical nucleus. Then, in first approximation, the rate $J$ of droplet formation per unit volume of mixture can be written as (Abraham, 1974):

$$J = c^* n^* ,$$

(2.3.4)

where $c^*$ is the impingement rate of vapour molecules on a critical nucleus and $n^*$ is the equilibrium number density of critical nuclei, related to the number density $n_1$ of vapour monomers by:

$$n^* = n_1 \exp(-\Delta F^*/kT) ,$$

(2.3.5)

with $k$ as the Boltzmann constant. This equation is in accordance with the classical nucleation theory (Abraham, 1974). The dependence of the nucleation rate on the saturation ratio is shown in Fig. (2.3.1) for water. It appears that the degree of supersaturation must be considerable before homogeneous nucleation leads to a significant rate of droplet formation.

\[ \text{Fig. 2.3.2.} \]

\textit{A liquid-phase embryo on a condensation particle.}

\textit{Heterogeneous nucleation}

In order to realize droplet formation at relatively low values of the saturation ratio, use can be made of insoluble particles. Such a particle acts as a catalysing surface on which condensation can take place. Consider a condensed phase embryo of radius $r_d$ on a spherical condensation nucleus of radius $r_n^*$ as shown in Fig. (2.3.2). The contact angle $\theta$ is determined by the affinity of the liquid to the particle material. Again, the radius $r^*$ of a critical embryo, i.e. an embryo in equilibrium with the vapour, is given by Eq. (2.3.3). However, the free energy of
formation of a critical embryo is reduced compared to that of an unbound droplet. According to Fletcher (1958):

\[
\frac{\Delta F^*_{\text{embryo}}}{\Delta F^*_{\text{unbound}}} = \mathcal{F}(\theta, x),
\]

where \(\Delta F^*_{\text{unbound}}\) follows from Eq. (2.3.2), and \(\theta = \cos \theta\) and \(x \equiv r_n/r^*\). The function \(\mathcal{F}(\theta, x)\) is given by:

\[
\mathcal{F}(\theta, x) \equiv \frac{1}{2} \left[ \frac{m}{g} \left( \frac{1}{2} \right)^3 - \frac{1}{2} x^3 (2 - 3h + h^3) + \frac{3}{2} mx^2 (h - 1) \right],
\]

where \(g = \sqrt{x^2 - 2mx + 1}\) and \(h = (z - m)/g\). Its dependence on the geometrical parameters is illustrated in Fig. (2.3.3). The reduction in \(\Delta F^*\) is most drastic for small contact angles, i.e. \(\theta \approx 1\). For \(r_n/r^* \geq 10\), the curvature of the nucleus surface has negligible effect on \(\Delta F^*\).

The mechanism of formation of stable embryos is very similar to the mechanism in the homogeneous case. Stable embryos form by cluster growth beyond the critical size due to the impingement of monomers. However, monomers are supplied by two means: either molecules condense on the embryo from the vapour phase, or monomers adsorbed on the substrate join the embryo by surface diffusion. The latter process is considered dominant (Sigsbee, 1969). In analogy with Eqs. (2.3.4) and (2.3.5), the rate equation is given by (Pound et al., 1954):

\[
J = c^* n_1 \exp( - \frac{\Delta F^*}{kT} ).
\]

Here, \(J\) is the rate of heterogeneous nucleation per unit area of substrate, \(c^*\) is the
impingement rate of adsorbed monomers on an embryo and \( n_1 \) is the number of adsorbed monomers per unit area. The values of the parameters that determine \( c^* \) and \( n_1 \) are uncertain for water (Kotake and Glass, 1976). Moreover, as reviewed by Van der Hage (1983), condensation experiments show that condensation of water on insoluble substrates occurs at saturation ratios substantially below those derived from the heterogeneous-nucleation theory using the values for the contact angle applicable to macroscopic droplets.

**The instantaneous-wetting model**

To circumvent the ambiguities mentioned, a simple instantaneous-wetting model will be used to describe droplet formation on hydrophilic particles, based on suggestions by Van der Hage (1983). It will be assumed that for all supersaturated states of the vapour, a thin liquid layer exists adsorbed to the surface of a condensation particle. Then, this perfectly wetted particle can be considered as a small droplet of radius \( r_n \). As explained before, this implies that the adsorbed liquid layer will grow by condensation, when the state of the vapour is such that \( r^* < r_n \), with \( r^* \) according to Eq. (2.3.3). Thus, the critical state of condensation onset, which we shall indicate by subscript \( c \), is given by the condition \( r^* = r_n \):

\[
R_n \rho_c R_v T_c \ln \chi_c = 2\sigma. \tag{2.3.9}
\]

If the critical state is reached by isentropic expansion from an initial unsaturated state without droplets, referred to by subscript \( o \), we have additionally:

\[
\frac{\chi_c}{\chi_o} \frac{p_{vs}(T_c)}{p_{vs}(T_o)} = \left( \frac{T_c}{T_o} \right)^{\gamma/(\gamma-1)}, \tag{2.3.10}
\]

where \( \gamma \) is the ratio of the specific heats of the gas-vapour mixture. Elimination of \( T_c \) from Eqs. (2.3.9) and (2.3.10) gives a relation between the radius \( r_n \) of the condensation particle and the critical saturation ratio \( \chi_c^* \). The model implies that during the expansion condensation on a particle of radius \( r_n \) is delayed until the critical saturation ratio \( \chi_c^* \) is reached, which value is determined by the particle radius. At that point, condensation is initiated instantaneously. If the nuclei in the mixture have uniform size, then there is only one unique critical state of droplet formation. The number of droplets formed is equal to the number of nuclei present.

---

3 Please note that here \( J \) and \( n_1 \) are formulated in terms of unit area of substrate, whereas for homogeneous nucleation the equivalent quantities were defined per unit volume of mixture.
Fig. 2.3.4. Critical saturation ratio as a function of the radius of the condensation nucleus. The substance is water. The critical state is obtained by an isentropic expansion ($\gamma = 1.4$) from the initial state: $T_0 = 295$ K, $\chi_0 = 0.8$. Points: instantaneous-wetting model. Shaded areas: heterogeneous-nucleation model; Areas a to d correspond to $\theta = 0^\circ$, $30^\circ$, $60^\circ$ and $90^\circ$. An additional explanation is found in the text.

Finally, the simple model of instantaneous wetting will be compared with the heterogeneous-nucleation model. To that end, we consider an isentropic expansion of a mixture of nitrogen gas and water vapour provided with spherical nuclei of uniform size $r_n$. We choose as initial conditions: $T_0 = 295$ K and $\chi_0 = 0.8$. The expansion rate is such that $d\chi/dt = 1$ ms$^{-1}$, and corresponds to the rates encountered in our experiments. The state of condensation onset according to the instantaneous-wetting model is uniquely determined. It is the critical state given by Eqs. (2.3.9) and (2.3.10). The heterogeneous-nucleation model, however, provides nucleation rates, rather than that it determines a unique supersaturated state at which droplet formation takes place on the condensation nuclei. A suitable critical state may be defined as the state at which one nucleation event takes place on one particle during a time interval that is one order of magnitude smaller than the characteristic time of expansion: $4\pi r_n^2 J = 10$ ms$^{-1}$. Further, the pre-exponential factor of Eq. (2.3.8) must be specified. Fletcher (1958) estimates $c^*n_1 = 10^{29}$ s$^{-1}$ m$^{-2}$, whereas Kotake and Glass (1976) use values of the order of $10^{35}$ s$^{-1}$ m$^{-2}$. We shall consider the range $10^{25}$ s$^{-1}$ m$^{-2} \leq c^*n_1 \leq 10^{35}$ s$^{-1}$ m$^{-2}$. Moreover, four different values for the contact angle will be used: $\theta = 0^\circ$, $30^\circ$, $60^\circ$ and $90^\circ$. Fig. (2.3.4) shows the result of the calculation. The critical saturation ratio is plotted as a function of the particle radius. The chain of
points gives the relation according to the simple model. The shaded areas are due to the nucleation model; every area corresponds to a different value of the contact angle. The influence of the contact angle is appreciable. The critical supersaturation decreases with decreasing contact angle. In the limit of large particles, the critical saturation ratio approaches an asymptotic value. The variation in the value of the pre-exponential factor by ten decades, seems to have little effect on $\chi_c$ for the case $\theta = 0^\circ$. For $\theta = 90^\circ$, the variation is substantial and the shaded area extends beyond the range of $\chi_c$ shown. The critical value of the saturation ratio predicted by the simple model agrees very well with the value provided by the nucleation model when $\theta = 0^\circ$ is used. Only for particle radii smaller than approx. 5 nm both models deviate.

2.4 A droplet in a gaseous carrier stream

Introduction

A fog is a two-phase system of randomly distributed droplets of liquid in a gaseous carrier, in the present study consisting of a mixture of vapour and inert gas. If the carrier state changes due to external influences, a disturbance of the equilibrium between the droplets and the carrier gas can easily occur, resulting in the exchange of heat, mass and momentum. Macroscopically, the droplets act as sources or sinks of heat, mass and momentum, changing the mean temperature, vapour mass fraction and velocity of the carrier. On a micro-scale however there exists a complex pattern of temperature, vapour mass fraction and velocity profiles around the droplets. Analysis of the problem in its total form is an impossible task and often the analysis is performed on the basis of a quasi-steady cellular model. The fog is considered to be subdivided into spatial cells, each containing one droplet. If the dimension of the cell is much larger than the dimension of the droplet, the cellular problem reduces to that of an isolated droplet in an infinite environment. Changes in the environmental conditions are handled quasi-steady and the transfer processes can readily be evaluated.

The problem of an isolated droplet in non-equilibrium with its surroundings has been the subject of extensive study. As early as 1877, Maxwell considered the steady evaporation of a droplet in his paper on diffusion. In his description, he used the theory of diffusive transfer of heat and mass, applicable to large droplets. However, for droplets with sizes of the order of the mean free path of the gas, free-molecular-flow effects and surface kinetics become important. Appropriate
treatments are those of Schrage (1953), concerning mass transfer, and Schaaf and Chambré (1961), concerning heat and momentum transfer. Combining continuum-flow and free-molecular-flow theory, Fuchs (1934, 1959) formulated a semi-empirical growth equation for the transition-flow regime. Effects of relative flow on droplet growth were correlated by Frössling (1938), using boundary layer and similarity theory. The drag on a droplet or sphere is a classical problem and there exist numerous treatments and empirical correlations, see for example Clift, Grace and Weber (1978). Recent developments and detailed considerations on many aspects and features of the subject can be found in reviews of Gyarmathy (1982), Wagner (1982) and Davis (1983).

![Figure 2.4.1](image.png)

**Fig. 2.4.1. Schematic representation of droplet in cell.**

a. Indication of the characterizing variables. b. Definition of the fluxes.

**The microscopic fluid cell**

We think the fog to be subdivided into spatial cells, each cell containing one droplet. Focusing on one cell, the situation is schematically depicted in Fig. (2.4.1a). At the outer border of the cell with effective radius $r_0$, the far-field conditions, i.e. the velocity $u_0$, the temperature $T_0$, the pressure $p_0$ and the saturation ratio $X_0$ of the carrier, are prescribed functions of time. The droplet, at the centre of the cell, is assumed to be spherical with a radius $r_d$ and a mass $m_d$. We assume no internal flow and no droplet rotation, so that the motion of the droplet can be characterized by a unique velocity $u_d$. For now, a temperature $T_r$ of the droplet surface and a mean temperature $T_d$ of the droplet are distinguished. In general, the carrier fluid and the droplet will not be in equilibrium. This will result

---

The time dependence should follow from the total macroscopic fluid-dynamics problem and will not be discussed in this chapter.
in an exchange of heat, mass and momentum and will cause the droplet parameters to change with time.

The balance equations

The temporal change of the droplet parameters can be expressed in terms of the fluxes by considering the balances of mass, momentum and energy of the droplet. The heat fluxes $H$ and $H_{\text{int}}$, the mass flux $M$ and the drag force $F$ are defined in Fig. (2.4.1b). The rate of change of the droplet mass and the droplet radius follow from the mass conservation equation:

$$\frac{dm}{dt} = \rho \frac{4\pi r^2 dr}{dt} = M.$$  \hfill (2.4.1)

The macroscopic fluid-dynamics problem will be considered to be a strictly one-dimensional problem, so the momentum equation for the droplet is a scalar equation:

$$m_d \frac{du_d}{dt} = F - m_d \frac{dp_d}{\rho \frac{dx}{dt}},$$  \hfill (2.4.2)

where $F$ is the drag force exerted on the droplet by the surrounding fluid. The last term in this equation represents the force on the droplet due to a pressure gradient associated with an acceleration of the carrier. The effects of gravity will be neglected. The energy balance of the droplet interior is written as:

$$m_d c_L \frac{dT_d}{dt} = H_{\text{int}} - M c_L (T_d - T_r).$$  \hfill (2.4.3)

Here, $H_{\text{int}}$ is the heat flux that enters the droplet, and the last term arises from the condensed mass to be heated from the surface temperature to the droplet mean temperature. A balance of the energy fluxes at the surface of the droplet results in:

$$H_{\text{int}} = H + M L(T_r).$$  \hfill (2.4.4)

A premise in writing down these equations is that the vapour mass arrives at the droplet or is given to the carrier with the temperature of the droplet surface and with the velocity of the droplet.

The time constant for temperature equilibration inside a droplet is given by:
where $a_\ell$ is the thermal diffusivity of the liquid. An analysis carried out by Gyarmathy (1982) resulted in the important conclusion that even in rapidly varying flows, like in steam turbines or supersonic nozzles, temperature differences inside the droplet are negligibly small for water droplets with a radius of the order of 1 $\mu$m. Only in time periods of the order of $\tau_{\text{int}}$ after an abrupt change in the environmental conditions, as occurs for example due to the passage of a shock wave, temperature differences have to be taken into account. For micron-sized water droplets, $\tau_{\text{int}}$ is of the order of microseconds. Therefore, we shall use Eqs. (2.4.1) to (2.4.4) with:

$$ T_r \approx T_d. \quad (2.4.6) $$

In using this approximation, it is not necessary to derive an equation for the internal heat flux $\mathbb{H}_{\text{int}}$, and we shall only be concerned with the flow of heat and mass inside the carrier and with the drag on the droplet.

**Assumptions regarding the transfer processes**

In this subsection, a number of simplifying assumptions will be discussed, that are necessary to make the analysis tractable that leads to the equations describing the fluxes $\mathbb{H}$ and $\mathbb{M}$, and the drag $F$.

The compositional dependence of the transport coefficients of the carrier, i.e. the thermal conductivity $\lambda$, the diffusion coefficient $D$ and the viscosity $\eta$, will be neglected, because we are exclusively dealing with carriers with low vapour content. Consequently, the transport coefficients of the carrier are approximated by those of the inert gas.

As mentioned before, the cell is assumed to be a concentric sphere with far-field conditions imposed on its outer boundary. The spatial variation of these far-field conditions will be considered negligible over distances of the dimension of the cell. This implies that the problem becomes spherically symmetric, or in the case of relative flow, will be cylindrically symmetric with the symmetry axis in the relative flow direction.

According to Hubbard et al. (1975), it is a good approximation to take the pressure uniform in the whole cell.

Based on the droplet number density according to Eq. (2.2.14), an effective
radius \( r_\infty \) of the cell can be defined:

\[
r_\infty^3 = \left( \frac{\rho_L}{\rho_d} \right) \langle r_d^3 \rangle.
\]

(2.4.7)

For \( r_d = 1 \mu m \) and \( n_d = 10^{12} m^{-3} \), it can be evaluated that the cell radius exceeds the droplet radius by a factor of 60. According to estimates carried out by Tishkoff (1979) and Gyarmathy (1982), proximity effects will alter the transfer rates of heat and mass, compared with the case of a droplet in an infinite environment, with about ten percent in the case of \( r_\infty/r_d \approx 20 \). Further, the drag experienced by a droplet in a fog may depend on the volume fraction of the droplets. For the flow at low values of the Reynolds number, the problem is analysed by Tam (1968). According to his drag formula, the drag on a droplet is affected less than one percent by the presence of surrounding droplets in the case that \( r_\infty/r_d \approx 60 \). Therefore, in this study the influences of neighbouring droplets will be neglected and the droplet will be considered to be embedded in an infinite carrier. Then, the spatial mean conditions of the carrier can be set equal to the far-field conditions.

A further simplification is the assumption of steady transfer processes. This has a number of implications. The time variation of the far-field conditions has to be sufficiently slow. The field quantities at the droplet surface have to be quasi-constant in time. And a change in the droplet radius due to condensation or evaporation has to have negligible influence on the transfer processes. Gyarmathy (1982) investigated these three conditions for the temperature field, by stating that the temperature field is negligibly disturbed, when changes in the excess thermal energy stored in the near field due to unsteadiness are ten percent or less of the heat transferred to the droplet. This resulted in a criterion for the constancy of the far-field conditions:

\[
\left| \frac{dT_\infty}{dt} \right| < \frac{a_\infty}{90} \frac{|T_\infty - T_r|}{r_d^2},
\]

(2.4.8)

with \( a_\infty \) as the thermal diffusivity of the carrier: \( a_\infty = \lambda/(\rho_c c_p) \), and a criterion for the constancy of \( r_d \):

\[
|T_\infty - T_r| < \frac{1}{260} \frac{\rho_L a_\infty L}{\lambda}.
\]

(2.4.9)

Referring to Appendix C, a typical value for the rate of change of the carrier temperature in this investigation is \( |dT_\infty/dt| = 10^4 K/s \). Using data from the tables in Appendix A, applied to a typical case of micron-sized water droplets in nitrogen.
gas at room temperature, the above criteria can be reformulated to a combined criterion for the temperature difference: $0.04 \, \text{K} < |T_\omega - T_I| < 8000 \, \text{K}$. As observed, this steady-state criterion is easily satisfied. Nix and Fukuta (1973) modelled the change of the macroscopic environmental conditions by means of source terms in the vapour diffusion and heat conduction equations, as is a correct representation under the assumption of a uniform pressure. From their results it can be deduced that for a stagnant growing droplet, non-steady effects can be neglected for micron-sized droplets, even in rapidly changing flows. Also Hubbard et al. (1975) concluded from numerical calculations of the full unsteady problem that transients in the carrier are of no importance. Deviations from the steady drag force on a moving sphere in a fluid due to unsteadiness can be considered to be caused by two effects (Rudinger, 1980; Clift et al., 1978). First, together with the acceleration of a sphere, an added fluid mass in the surroundings of the sphere has to be accelerated, too, resulting in a virtual mass for the sphere as: $m_s (1 + \rho_f / \rho_s)$, with $m_s$ as the sphere mass, $\rho_s$ as the sphere material density and $\rho_f$ as the fluid density. Second, the flow pattern around the sphere deviates from that for steady flow, due to the influence of the history of acceleration. The resulting extra force on the sphere is called the Basset force. The ratio of the Basset force and the steady drag force is of the order of $\rho_f / \rho_s$ (Rudinger, 1980). For water droplets in nitrogen gas, the density ratio $\rho_f / \rho_s$ is about $1/1000$. Hence, both unsteady effects may be neglected.

Free convection in the carrier will not be taken into account, since the size of the droplets of interest is of the order of a micron or less.

The thermal diffusion (Soret) effect and the diffusion thermal (Dufour) effect are neglected (Wagner, 1982).

**Introduction of the Nusselt numbers**

The rates of heat transfer $\dot{H}$ and mass transfer $\dot{M}$, and the drag force $F$ can be expressed conveniently with the help of dimensionless transfer numbers. Adopting the notation of Gyarmathy (1982), Nusselt numbers for heat, mass and momentum transfer are defined as:

\[
\text{Nu}_H = \frac{\dot{H}}{2\pi r_d \lambda (T_\omega - T_I)},
\]

\[
\text{Nu}_M = \frac{\dot{M}}{2\pi r_d D_m (p_{v\omega} - p_{vI})/p_{g\omega}},
\]

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A modified diffusion coefficient $D_m$, with dimension $\text{kg m}^{-1} \text{s}^{-1}$, has been introduced in the equation for the Nusselt number for mass transfer:

$$D_m = \frac{D_p}{R_v T_m}.$$  \hspace{1cm} (2.4.13)

The thermal conductivity $\lambda$, the viscosity $\eta$, the diffusion coefficient $D$ and the modified diffusion coefficient $D_m$ have to be evaluated at an appropriate temperature $T_m$, intermediate between $T_r$ and $T_w$. According to Hubbard et al. (1975), a one-third rule: $T_m = (2T_r + T_w)/3$, performs satisfactorily.

The vapour at the droplet surface is in equilibrium with the liquid, therefore $p_{vr}$ follows from Eq. (2.2.9):

$$p_{vr} = p_{ve}(T_r, r_d).$$  \hspace{1cm} (2.4.14)

Expressions for the Nusselt numbers will depend on the circumstances of droplet growth. In the next few subsections, we will explore several cases.

**The continuum-flow regime**

For large droplets, transfer of heat, mass and momentum is diffusion controlled. In the case that the droplet is stagnant with respect to its environment, considering momentum transfer is redundant, and the rates of heat and mass transfer can be derived from the differential equations for the temperature and the vapour-concentration fields:

$$\rho_v \frac{dT}{dr} = \left(\frac{\lambda}{r^2}\right) \frac{d}{dr} \left( r^2 \frac{dT}{dr} \right),$$  \hspace{1cm} (2.4.15)

$$\rho_v (v_v - v) = - (\rho_v + \rho_g) \frac{dg_v}{dr},$$  \hspace{1cm} (2.4.16)

supplemented with the correct boundary conditions. Here, $g_v$ is defined by Eq. (2.2.17), and $v_v$ and $v$ are the radial velocity of the vapour flow and the carrier counter flow, also called Stefan flow, respectively. The derivation is carried out in Appendix B and results in the following relations for the Nusselt numbers:
\[ \text{Nu}_{ct}^{o} = 2 \frac{\ln(1 + x)}{x}, \quad (2.4.17) \]

\[ \text{Nu}_{ct}^{ct} = 2 \frac{\ln(1 + y)}{y}. \quad (2.4.18) \]

The extra subscript \( o \) stands for zero relative flow and the superscript \( ct \) refers to continuum flow. In the derivation of Eq. (2.4.17), use has been made of the wet-bulb approximation, discussed further down in this section. The parameter \( x \) is the superheat parameter, often used in literature (Yuen and Chen, 1978; Lebedev et al., 1981) to correlate severe evaporation:

\[ x = \frac{c p v (T_\infty - T_R)}{L}. \quad (2.4.19) \]

The parameter \( y \) becomes important in the cases of high vapour mass fraction and strong mass flow:

\[ y = \frac{p_{v\infty} - p_{vR}}{p_{g\infty}}. \quad (2.4.20) \]

When the differences in the temperature and the vapour pressure are moderate, then the correction terms can be ignored and the Nusselt numbers attain a value of two. Only during rapid evaporation behind strong shock waves, the dependence on \( x \) and \( y \) has to be taken into account.\(^5\)

**Relative flow**

Relative flow in the continuum regime is characterized by the relative Reynolds number \( \text{Re}_d \) and the relative Mach number \( \text{Ma}_d \) of the droplet:

\[ \text{Re}_d = \frac{2r_d |u_\infty - u_d|}{\nu_\infty}, \quad \text{Ma}_d = \frac{|u_\infty - u_d|}{a_\infty}. \quad (2.4.21) \]

with \( \nu_\infty \) as the kinematic viscosity: \( \nu_\infty = \eta/\rho_\infty \) and \( a_\infty \) as the sound speed: \( a_\infty = \sqrt{\gamma \rho_\infty R T_\infty} \). Convective transport of heat and mass becomes already important for

\(^5\) Typically, for the range \( 0.1 \text{ bar} < p_\infty < 2 \text{ bar}, \ 250 \text{ K} < T_\infty < 350 \text{ K} \) and \( 0 < x_\infty < 4 \), the effect on the mass transfer rate is smaller than two percent.
low values of the Reynolds number. The general form of the empirical correlations is based on the similarity principle of boundary layer theory, and was first proposed by Frössling (1938). Correlations, due to Rans and Marshall (1952) and applicable to a Reynolds-number range far beyond the range needed for small droplets, are:

\[ \text{Nu}_{ct}^* = \text{Nu}_{ct}^{*0} \left(1 + 0.30 \text{Re}_d^{1/2} \text{Pr}^{1/3}\right) \quad (2.4.22) \]

\[ \text{Nu}_{ct}^* = \text{Nu}_{ct}^{*0} \left(1 + 0.30 \text{Re}_d^{1/2} \text{Sc}^{1/2} \text{Pr}^{1/3}\right) \quad (2.4.23) \]

The factor \( \Pi = p_g / p_\infty \) is nearly equal to unity for carriers with a low vapour content. The Prandtl and Schmidt numbers read:

\[ \text{Pr} = \nu_\infty / a_\infty \quad \text{and} \quad \text{Sc} = \nu_\infty / D \quad (2.4.24) \]

and have to be evaluated at the intermediate temperature \( T_m' \).

The correlation for the drag that will be applied is that of Schiller and Naumann (1933) for the range \( \text{Re}_d < 800 \):  

\[ \text{Nu}_{ct}^* = 3 \left(1 + 0.15 \text{Re}_d^{0.687}\right) \quad (2.4.25) \]

For \( \text{Re}_d \to 0 \), this expression results in a Nusselt number of three, which corresponds, according to Eq. (2.4.12), to Stokes drag for a slowly moving sphere.

A non-volatile sphere, in thermal equilibrium with a gas flow, has a temperature that is different from the far-field temperature of the gas, due to a compression of the flowing gas at the sphere. The resulting temperature is called the recovery temperature \( T_{rec} \) and equals:

\[ T_{rec} = T_\infty \left[1 + \frac{15}{16} (\gamma - 1) \text{Ma}^2_d\right] \quad (2.4.26) \]

The same effect applies to a droplet, as well. Therefore, for sufficiently large Mach numbers, \( T_\infty \) has to be replaced with \( T_{rec} \) in the definition of the Nusselt number for heat transfer, i.e. Eq. (2.4.10).

As shown in Appendix C, \( \text{Re}_d \) and \( \text{Ma}_d \) have low values in the case of a one-dimensional unsteady centred rarefaction wave, so relative-flow effects on the transfer of heat and mass are of no importance, and the drag can readily be modelled by the Stokes drag. In the case of a shock wave, the extended transfer equations have to be applied in a short period after passage of the shock wave.
The free-molecular-flow regime

In free molecular flow, the dimension of the droplet is small compared to the molecular mean free path of the ambient gas. More specific, according to Schaaf and Chambré (1961), the regime starts at $\text{Kn} \approx 7$, where $\text{Kn}$ is the Knudsen number:

$$\text{Kn} = \frac{\ell}{2r_d},$$

and $\ell$ is the molecular mean free path, according to Gyarmathy (1982):

$$\ell = \frac{2\eta \sqrt{(R^\omega T^\omega)}}{p^\omega}.$$  

For nitrogen gas at room temperature and atmospheric pressure $\ell \approx 0.1 \mu m$.

In free molecular flow, molecules emitted by and reflected from a droplet surface travel a relatively large distance before they bounce with the free stream molecules. Therefore, boundary layers are absent, or very diffuse, and the ambient gas is not disturbed by the presence of the droplet. The transfer of heat, mass and momentum is then determined by molecular impingement rates and surface kinetics.

The interaction of gas and vapour molecules with the droplet may be described by a number of coefficients, which express the average response of an incoming molecule on its encounter with the liquid surface (Gyarmathy, 1982). Thermal accommodation is represented by the coefficient $a_H$. It is defined by the relation:

$$T_{\text{reb}} = T^\omega - a_H (T^\gamma - T^\omega),$$

where $T_{\text{reb}}$ is the average temperature of the rebounding molecules. In general, the thermal accommodation coefficient differs for the gas and the vapour component. The condensation coefficient $a_M$ applies only to the vapour component and equals the fraction of impinging vapour molecules that actually stick to the liquid surface. Finally, we introduce the diffuse reflection coefficient $a_F$.

It distinguishes between the fraction $a_F$ of molecules, that reflect from the droplet surface diffusely and with a temperature $T^\gamma$, and the remaining fraction $1 - a_F$ of molecules, that reflect specularly and with a temperature $T^\omega$. Again, the gas and the vapour have different coefficients.

The kinetic theory of gases gives for the mass of vapour and gas, that impinges on unit surface area of the droplet per unit time:

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\[ \beta_{\text{V}} = \frac{P_{\text{V}}}{\sqrt{2\pi R_\text{V} T_\text{V}}} \quad \text{and} \quad \beta_{\text{G}} = \frac{P_{\text{G}}}{\sqrt{2\pi R_\text{G} T_\text{G}}}. \quad (2.4.29) \]

Then, as explained by Schaal and Chambré (1961), the rate of transfer of heat to a non-absorbing sphere with a temperature \( T_\text{X} \) is given by:

\[ H = 4\pi r^2 \left( a_{\text{MV}} \beta_{\text{V}} \Gamma_\text{V} + a_{\text{MG}} \beta_{\text{G}} \Gamma_\text{G} \right) (T_\text{X} - T_\text{R}), \quad (2.4.30) \]

where \( \Gamma = 0.5 (\gamma+1)/(\gamma-1) \). The equation that describes the transfer of vapour to a droplet is (Schragé, 1953):

\[ M = 4\pi r^2 a_{\text{MV}} (\beta_{\text{V}} - \beta_{\text{V}}) . \quad (2.4.31) \]

Here, \( -a_{\text{MV}} \beta_{\text{V}} \) is equal to the mass of liquid that leaves the droplet per unit area per unit time. Since \( M \) has to be zero in an equilibrium situation, it follows that \( \beta_{\text{V}} \) is given by:

\[ \beta_{\text{V}} = \frac{P_{\text{V}} e(T_\text{X})}{\sqrt{2\pi R_\text{V} T_\text{R}}}. \quad (2.4.32) \]

As treated in detail by Schaal and Chambré (1961) and summarized by Gyarmathy (1982), the drag on a sphere in free molecular flow at low Mach numbers is described by:

\[ F = 4\pi r^2 \left( \beta_{\text{V}} \zeta_\text{V} + \beta_{\text{G}} \zeta_\text{G} \right) (u_\text{X} - u_\text{D}) , \quad (2.4.33) \]

where \( \zeta = \frac{4}{3} + \frac{1}{6} a_F \sqrt{(T_\text{R}/T_\text{X})} \).

In writing down the balance equation for the energy fluxes at the vapour-liquid interface, Eq. (2.4.4), the assumption was made that the vapour arrives at or leaves the droplet with a temperature equal to that of the interface. As correctly pointed out by Barrett and Clement (1988), this assumption is invalid in the case of free molecular flow. Starting from correct premises, the flux balance equation should be:

\[ H_{\text{int}} = H + M \left[ L(T_\Gamma) - \frac{1}{2} R_\text{V} T_\text{R} \right] , \quad (2.4.34) \]

with \( H_{\text{int}} \) according to Eq. (2.4.3), and \( H \) and \( M \) according to Eqs. (2.4.30) and (2.4.31). However, for water the second term between square brackets is only a few
percent of the first one, and may readily be neglected.

The transfer equations for free molecular flow as given above are only applicable for Mach numbers very close to zero. This, however, is not a severe restriction, since the smallness of the droplets in the Knudsen regime guarantees a negligible velocity difference between the droplets and the carrier.

Now, the equations describing the transfer rates in the case of free molecular flow have to be combined with the defining equations for the Nusselt numbers, i.e. Eqs. (2.4.10) to (2.4.12), to obtain the Nusselt numbers for the Knudsen regime. An outline of the procedure can be found in the book by Gyarmathy (1982). The resulting expressions are rather complex. Therefore, by using $g_v \ll 1$ and $T_r \approx T_w$, the equations simplify to:

$$\text{Nu}_H^{fm} = B_H/\text{Kn} , \quad \text{Nu}_M^{fm} = B_M/\text{Kn} , \quad \text{Nu}_F^{fm} = B_F/\text{Kn} , \quad (2.4.35)$$

where:

$$B_H = \sqrt{(2/\pi)} \frac{\gamma_p^{+1}} {2\gamma_p} Pr a_{HG} , \quad (2.4.36)$$

$$B_M = \sqrt{(2/\pi)} \sqrt{(B_v / B_g)} Sc a_{MV} , \quad (2.4.37)$$

$$B_F = \sqrt{(2/\pi)} \left( \frac{4}{3} + \frac{\gamma_p^{+1}} {6} a_{FG} \right) . \quad (2.4.38)$$

The Prandtl number $Pr$ and the Schmidt number $Sc$ have to be evaluated at the temperature $T_w$.

The parameters $B_M$, $B_H$ and $B_F$ depend on the condensation coefficient $a_{MV}$ of the vapour and on the thermal accommodation coefficient $a_{HG}$ and the diffuse reflection coefficient $a_{FG}$ of the gas, respectively. Uncertainty exists in the literature about the value of $a_M$ for water. Reported values range from 0.04 to 1 (Wagner, 1982). In a recent review of the matter, Mozurkewich (1986) concludes that its value is close to unity. This viewpoint will be adopted. Schaal and Chambré (1961) have tabulated values of $a_H$ and $a_F$ for air on various metals and oil. All values are close to unity. Therefore, we shall assume for the coefficients $a_{HG}$ and $a_{FG}$ of nitrogen gas and liquid water, that these are equal to unity.

Using numerical data from the tables in Appendix A, and evaluating $Pr$ and $Sc$ at $T_w = 295$ K, typical values for these coefficients are obtained as:

$$B_H = 0.49 , \quad B_M = 0.62 \quad \text{and} \quad B_F = 1.48 . \quad (2.4.39)$$

---

* See also Appendix C.
The transition-flow regime

Actually, the largest part of most droplet-growth processes takes place in the regime of intermediate Knudsen numbers. For this reason Nusselt-number expressions for the whole range of Knudsen numbers are necessary. Proposed expressions should show the correct asymptotic behaviour for small and large Knudsen numbers. In 1934, Fuchs introduced the flux-matching method to describe mass transfer at arbitrary Knudsen numbers. Fukuta and Walter (1970) applied the method both to heat and mass transfer to obtain a consistent growth model. In the case of the flux-matching method, the carrier field is divided into two zones. The inner zone lies concentric around the droplet and has a thickness \( \Delta \). Within the inner zone, the transfer processes are described by free-molecular-flow equations, whereas the continuum-flow theory is applied in the outer zone. Then, the fluxes are matched at the common border, resulting in a multirange expression of the form:

$$\text{Nu} = \frac{\text{Nu}_{ct}}{1 + \frac{\text{Nu}_{ct}}{\text{Nu}_{ct}} \frac{\text{m}}{\text{m}} - \frac{\Delta}{(r_d + \Delta)}}. \quad (2.4.40)$$

Of course, in reality there are no distinct zones of continuum and free molecular flow, and \( \Delta \) has to be looked upon as a matching parameter of the order of the molecular mean free path, that is a function of the Knudsen number. The functional dependence has to follow from an empirical correlation or from a rigorous solution of the Boltzmann equation. Various expressions proposed in literature are discussed by Wagner (1982). Gyarmathy (1982) argues on the basis of experimental data, that it is allowed to omit the last term of the denominator, so that one obtains, for each of the Nusselt numbers of heat, mass and momentum transfer, an equation as:

$$\text{Nu} = \frac{\text{Nu}_{ct}}{1 + \left(\frac{\text{Nu}_{ct}}{\text{B}}\right) \text{Kn}}. \quad (2.4.41)$$

where \( \text{Nu} \), \( \text{Nu}_{ct} \) and \( \text{B} \) may be subscribed by either \( H \), \( M \) or \( F \).

Davis (1983) compared several theoretical and semi-theoretical expressions for mass transfer in the transition regime. For mixtures with the vapour lighter than the inert gas, there is no reason to rule out the simple equation (2.4.41). Heat

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7 We shall refer to the decrease in the Nusselt numbers with an increase in the Knudsen number as the Knudsen effect.
transfer data of Takao (1963) and Kavanau (1955), covering the whole Knudsen range, were correlated by Sherman (1963), using a correlation of the type of Eq. (2.4.41). The agreement he obtained was within the limit of the experimental accuracy. For the drag on a sphere, Gyarmathy (1982) compared the model equation (2.4.41), with the accurate correlation of Henderson (1976). He found good agreement for low Reynolds and Knudsen numbers, whereas deviations of more than ten percent were found for $Ma_d = (Kn Re_d)/(2\sqrt{\gamma}) > 0.1$.

Reconsideration of the momentum equation

By introducing the relative velocity $\Delta u = u_d - u_\infty$, and using Eq. (2.4.12) for the drag force, the momentum equation, Eq. (2.4.2), can be rewritten as:

$$
\frac{d\Delta u}{m_d} = \frac{2\pi r_d \eta Nu_F \Delta u}{m_d} \left( \frac{\partial p}{\partial x} + \frac{du}{\rho_l \partial x} \right).
$$

(2.4.42)

The velocity difference, for which the left-hand side of this equation becomes zero, is called the drift velocity $\Delta u_{drf}$, which, as can be seen at once, obeys the relation:

$$
\frac{\Delta u_{drf}}{\tau_{drf}} = \frac{1}{\rho_l \partial x} \left( \frac{\partial p}{\partial x} + \frac{du}{\partial x} \right),
$$

(2.4.43)

with $\tau_{drf}$ defined as:

$$
\tau_{drf} = \frac{2 \rho_l r_d^2}{3 \eta Nu_F}.
$$

(2.4.44)

The characteristic time $\tau_{drf}$ can be considered to be a measure for the time required to attain the drift velocity after disturbance. In the case that the characteristic time for the change of the environmental conditions is much larger than $\tau_{drf}$, then the droplets will travel with a velocity $u_\infty + \Delta u_{drf}$ all the time. Taking $Nu_F = 3$ and $r_d = 1 \mu m$, and using numerical data from Appendix A, we obtain $\tau_{drf} \approx 10 \mu s$, whereas in our investigation the time constant of environmental change is well above $1 \text{ms}^8$. Of course, Eq. (2.4.43) does not apply to time periods of the order of $\tau_{drf}$ after passage of a shock wave. A comparison of the pressure-gradient term and the acceleration term in Eq. (2.4.43), for the case of a frictionless flow with a negligible droplet mass fraction, results in:

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8 See Appendix C.
from which it can be concluded that the pressure-gradient term is negligible for liquid droplets in gaseous media.

**The wet-bulb approximation**

Consider for a moment a droplet with an arbitrary temperature, suddenly exposed to an essentially steady environment. We assume the droplet to be sufficiently large, in order to neglect Kelvin and Knudsen effects. The equation, describing the behaviour of the droplet, is obtained from Eqs. (2.4.3) and (2.4.4), combined with the assumption of a uniform droplet temperature, i.e. Eq. (2.4.6):

\[
\frac{d}{dt} \left( m_d c_L \frac{dT_d}{dt} \right) = H + M L(T_d) .
\]  

(2.4.46)

Due to the special premises, the temperature of the droplet will change in a short transient period, but will soon become steady. This steady situation then obeys:

\[
H = -ML ,
\]  

(2.4.47)

or after substitution of Eqs. (2.4.10) and (2.4.11):

\[
\frac{\nu_k \lambda}{\kappa_d} \left[ T_w - T_d \right] = -\frac{D_m}{\kappa_d} L(T_d) \left[ \frac{p_v - p_f}{p_g} \right] .
\]  

(2.4.48)

All parameters in this equation are functions of the environmental conditions, of the droplet radius and of the droplet temperature. Therefore, the droplet temperature can be calculated from this algebraic equation for given droplet radius and environmental conditions. Equation (2.4.48) is called the wet-bulb equation; the droplet temperature resulting from it is called the wet-bulb temperature.

Because of its algebraic form, it is advantageous to apply the wet-bulb equation to non-steady situations and to droplets of arbitrary sizes, as well. However, application is only valid if the magnitude of the interior heat flux is much smaller than the magnitude of the flux of latent heat to the droplet, say:

\[
|H_{int}| < \xi |ML| ,
\]  

(2.4.49)

where \( \xi \) is a small parameter. This criterion can be rewritten to obtain:
Evaluation of the criterion is possible if an additional relation for the relative change of the droplet temperature is obtained. For that, we assume a monodisperse fog, and use the wet-bulb equation as a starting point, written in the form:

\[
\frac{(T_\omega - T_d)}{\theta} = \frac{(p_{v\omega} - p_{vT})}{p_{g\omega}}.
\]

Here \( \theta \) has the dimension of a temperature and equals:

\[
\theta \equiv \frac{\frac{D m L N_u_M}{\lambda N_u_H}}{m}. \tag{2.4.51}
\]

This parameter is considered constant, since \( N_u_M \) and \( N_u_H \) have a nearly identical dependence on the droplet radius and on the relative velocity, and \( \frac{B_m}{\lambda} \) and \( L \) are only weakly dependent on \( T_m \) and \( T_d \), respectively. Differentiating Eq. (2.4.51) with respect to time, eliminating all time derivatives except those of \( T_d, m_d, T_\omega \) and \( p_{\omega} \), making the valid assumption: \( L/(R_v T_d) \gg K_e \), and combining the result with Eq. (2.4.50) yields:

\[
\frac{T_\omega}{\theta} + \frac{p_{vT}}{p_{g\omega}} \frac{r_m}{\tau_p} + \frac{p_{\omega}}{p_{g\omega}} \frac{R_v}{\overline{R_v}} f_d \left( \frac{p_{vT}}{p_{\omega}} \right) - \frac{p_{vT}}{p_{g\omega}} - \frac{p_{vT}}{p_{g\omega}} \frac{Ke}{3} < \frac{\xi L}{c_{vH}} \left( 1 + \frac{p_{vT}}{p_{g\omega}} \frac{L}{R_v T_d} \right). \tag{2.4.52}
\]

Some characteristic times have been introduced as:

\[
\tau_m = \frac{1}{\frac{dm_d}{m_d dt}}, \quad \tau_T = \frac{1}{\frac{dT_d}{T_d dt}}, \quad \tau_p = \frac{1}{\frac{dp_{\omega}}{p_{\omega} dt}}, \tag{2.4.53}
\]

which are the characteristic times for droplet growth, and for the change of the environmental temperature and pressure. The time constant \( \tau_m \) can be written as:

\[
\tau_m = \frac{m_d L}{|H| |R|} = \frac{2}{3 \frac{r_d}{L} \frac{T_\omega - T_d}{T_m}}. \tag{2.4.54}
\]

For the cases dealt with in this investigation, some approximations may be made in this context: \( p_{g\omega} \approx p_{\omega}, \ p_{vT} \approx p_{\omega} \) and \( \overline{R_v} \approx \overline{R} \), so that the final criterion becomes:
\[
\frac{T_m \tau_m}{\theta} + \frac{P_{VR} \tau_m}{\rho_\infty \tau_p} + \left| \frac{R_v f_d}{p_\infty} - \frac{P_{VR} \kappa e}{3 \rho_\infty} \right| < \frac{L}{c_L \theta} \left( 1 + \frac{P_{VR}}{\rho_\infty R_v T_d^2} \right). \tag{2.4.56}
\]

Distilling the independent parameters from this criterion, a total of seven is obtained, namely: \(\tau_T, \tau_p, T_m, \rho_\infty, \chi_\infty, f_d\) and \(r_d\). For this reason, an analysis of the criterion in its total form is too complicated, and only some special cases will be discussed.

First, consider \(\tau_T\) and \(\tau_p\) to be infinite, then the only terms that remain on the left are the terms between the modulus bars. Treating them separately results in:

\[
f_d < \frac{\xi L R_g}{c_L \theta R_v} \left( 1 + \frac{P_{VR} L \theta}{p_\infty R_v T_d^2} \right) \approx 0.02 \times 2 \frac{P_{VR}}{p_\infty} \frac{295}{T_d(K)}^2, \tag{2.4.57}
\]

\[
\kappa e < \frac{3 \xi L p_\infty}{c_L \theta p_{VR}} \left( \frac{L \theta}{R_v T_d^2} \right) \approx 0.1 \frac{p_\infty}{P_{VR}} + 10 \left[ \frac{295}{T_d(K)} \right]^2. \tag{2.4.58}
\]

The estimates were obtained with data for water and nitrogen from Appendix A, and the small parameter \(\xi\) was taken to be 0.1. Putting \(p_\infty = 100 P_{VR}\) and \(T_d = 295 K\), this leads to \(f_d < 0.04\), which is a noticeable result, and \(\kappa e < 20\), which will hardly ever be a problem.

Next, the case of a droplet in an adiabatically changing fog with a negligible droplet mass fraction is considered. The temperature and the pressure are related by the isentropic gas law, yielding a relation for the time constants of environmental change:

\[
\tau_p = \frac{\gamma_g - 1}{\gamma} \tau_T, \tag{2.4.59}
\]

where the ratio of the specific heats of the gas-vapour mixture is approximated by \(\gamma_g\). Substitution into Eq. (2.4.56) results in a minimum time constant for the change of the carrier temperature:

\[
\tau_T > \tau_T^{\min} = \tau_m \frac{c_L \theta}{\xi L} \left[ \frac{T_m}{\gamma_g} + \frac{P_{VR} \gamma_g}{\rho_\infty \gamma_g - 1} \right] \left( 1 + \frac{P_{VR} L \theta}{p_\infty R_v T_d^2} \right)^{-1}. \tag{2.4.60}
\]

This minimum time constant is plotted in Fig. (2.4.2) as a function of the saturation ratio \(\chi_\infty\) and the droplet radius \(r_d\). The curves shown belong to \(T_m = \)
295 K, \( p_\infty = 1 \text{ bar} \) and \( \zeta = 0.1 \), and have been calculated with data for water and nitrogen from Appendix A. As observed, the minimum allowable time constant increases rapidly with the droplet radius. Moreover, every curve has a singular point, where the validity criterion breaks down. This is because the time constant \( \tau_m \) for droplet growth is inversely proportional to \( |T_\infty - T_d| \) and becomes infinite in the equilibrium situation. For large droplets the singularity lies close to \( \chi_\infty = 1 \), while it shifts to higher values of the saturation ratio for decreasing droplet radius, which is due to the Kelvin effect.

According to Appendix C, the characteristic time \( \tau_T \) for the change of the environmental temperature in the case of an unsteady centred rarefaction wave is given by \( \tau_T \approx 3t \), where \( t \) is the time. Now, assume a droplet that starts to grow from zero radius at time zero and use the algebraic growth law described in the next section, i.e. Eq. (2.5.5). Then, by obtaining data for water and nitrogen from Appendix A and taking the conservative values \( \text{Nu}_H = 2 \) and \( T_d - T_\infty = 10 \text{ K} \), the following equation can be derived: \( \tau_T (\text{ms}) \approx 150 x_3^2 \left( \mu m^2 \right) \). Comparing this equation with Fig. (2.4.2), the conclusion is justified that the wet-bulb approximation may be applied under the circumstances of the present study.

2.5 Droplet-growth models

Recapitulation

In the preceding section, the processes that control the behaviour of a droplet in a gaseous carrier stream were discussed. It was argued that important simplifications can be made for the conditions dealt with in this investigation. Recapitulating, the temperature inside the droplet is uniform and the transfer processes in the carrier are quasi-steady. Moreover, the effects of relative flow on heat and mass transfer are negligible, the wet-bulb approximation is valid and, finally, the logarithmic corrections to the continuum Nusselt numbers are irrelevant. These assumptions, except the last one, do not hold in a short period after shock wave passage and an extended model has to be applied. We shall return to this point in Chapter 4. The last assumption is invalid in the case of rapid evaporation behind strong shock waves. In this section, the droplet-growth model based on the premises above and used in this investigation is summarized. Moreover, several growth models, known in the literature and based on additional simplifications, are discussed and compared with the present one.
Fig. 2.4.2. Minimum time constant for the change of the carrier temperature in order to satisfy the wet-bulb approximation. Plotted as a function of the saturation ratio $\chi_\infty$ and the droplet radius $r_d$. Calculated with data for water and nitrogen of Appendix A. Conditions: $p_w = 1$ bar, $T_w = 295$ K. Criterion parameter according to Eq. (2.4.49): $\xi = 0.1$.

The present growth model

Basic in the droplet-growth model is the wet-bulb approximation. This means that there is an instantaneous balance of the flows of heat and mass between a droplet and its gaseous environment as described by Eq. (2.4.47). Using this relation together with Eqs. (2.4.1) and (2.4.10), the droplet-growth law can be formulated in terms of the rate of change of the droplet radius squared:

$$\frac{d r_d^2}{dt} = \frac{\text{Nu}_H \lambda}{\rho_L L} (T_d - T_w) = \phi_{r_d^2},$$

(2.5.1)

where we have simultaneously introduced the droplet-growth function $\phi_{r_d^2}$. It is important to realize that this relation is only valid under wet-bulb conditions. Thus, the droplet temperature $T_d$ is not a free parameter, but is governed by this restriction. A straightforward approach to obtain the droplet temperature is to use the wet-bulb equation, i.e. Eq. (2.4.48):

$$\text{Nu}_H \lambda [T_w - T_d] = -\text{Nu}_M D_m L(T_d) \left[ p_{v_w} - p_{v_f} \right] / p_{g_w},$$

(2.5.2)
where:

\[ p_{\text{vr}} = p_{\text{vs}}(T_d) \exp \text{Ke}(T_d, r_d), \quad (2.5.3) \]

and the Nusselt numbers are given by:

\[ \text{Nu}_H = \frac{2}{1 + 4.08 \text{ Kn}} , \quad \text{Nu}_M = \frac{2}{1 + 3.23 \text{ Kn}} . \quad (2.5.4) \]

Here, use has been made of Eqs. (2.4.41) and (2.4.39), and the continuum Nusselt numbers are put equal to two.

For a sufficiently large droplet in an infinite reservoir, both the droplet temperature and the environmental quantities are constant, so Eq. (2.5.1) results in a linear relationship between the droplet radius squared and the time:

\[ \frac{r_d^2 - r_{d_0}^2}{\rho \lambda L} = \frac{\text{Nu}_H}{1} (T_d - T_w) (t - t_0) . \quad (2.5.5) \]

When using Eq. (2.5.2), droplet growth can still be calculated with sufficient accuracy for values of the saturation ratio \( \chi_w \) deviating considerably from unity. However, because of its implicit form a solution can only be obtained numerically at substantial computational cost. Moreover, a qualitative understanding of droplet-growth phenomena is difficult, since no analytical solutions can be formulated.

**Explicit droplet-growth models**

By introducing some simplifying assumptions, the implicit wet-bulb equation can be replaced by an equation that explicitly relates the droplet temperature to the droplet size and the environmental conditions. Once the droplet temperature is known, the growth rate of the droplet can easily be calculated with Eq. (2.5.1). In the literature, different simplifications are proposed to obtain different explicit relations for calculating the growth rate of a droplet. Most authors use a linearization of the Clausius-Clapeyron equation to obtain an explicit growth equation (Mason, 1953; Wagner, 1982; Mozurkewich, 1986; Barrett & Clement, 1988), whereas Gyarmathy (1963) chooses a different approach. To explore the applicability of these explicit equations, we will discuss the equations of Barrett and Clement (1988), Mason (1953) and Gyarmathy (1963), and compare them with the implicit model.
The function \( f(\chi_\infty, Ke_\infty) \) and the parameters \( C_1 \) and \( C_2 \), as defined by Eq. (2.5.6), according to Barrett and Clement (1988), Mason (1959) and Gyarmathy (1963).

The parameters \( Ke, L \) and \( \theta \) have a subscript \( \infty \) to indicate that they have to be evaluated at the temperature \( T_\infty \). The parameter \( \theta \) is defined by Eq. (2.4.52), with the Nusselt numbers according to Eqs. (2.5.4).

The general form of the explicit equations giving the droplet temperature is:

\[
\frac{T_d}{T_\infty} - 1 = f(\chi_\infty, Ke_\infty) \left[ C_1 + C_2 \right]^{-1}. \tag{2.5.6}
\]

Here \( Ke_\infty = Ke(T_\infty, r_d) \), and \( C_1 \) and \( C_2 \) are both functions of \( \chi_\infty, T_\infty, p_\infty \) and \( r_d \). The function \( f(\chi_\infty, Ke_\infty) \) and the parameters \( C_1 \) and \( C_2 \) are listed in Table (2.5.1) for the equations of Barrett and Clement (1988), Mason (1953) and Gyarmathy (1963). The parameter \( C_2 \) is the same for all models and is independent of the saturation ratio. For water in the temperature range 250–350 K, it is a decreasing and approximately linear function of the temperature that varies between 22 and 14. Differences between the models occur in the function \( f(\chi_\infty, Ke_\infty) \) and in the parameter \( C_1 \). The variation with temperature of \( C_1 \) is shown in Fig. (2.5.1) for a

---

*The derivation is outlined in Appendix D.*
typical case of water and nitrogen.

The simplifying assumptions underlying the explicit equations are listed in Table (2.5.2). Barrett and Clement (1988) and Mason (1953) assume that differences in the saturation vapour pressure are small, whereas Gyarmathy (1963) assumes that differences in the vapour pressure are small. For a water droplet in humid nitrogen, the vapour pressure and the saturation vapour pressure are shown in Fig. (2.5.2) for a typical case as functions of the radial distance to the droplet. An extra assumption made by Mason (1953), not necessarily needed to obtain an explicit equation, is that the partial vapour pressure is small compared to the total pressure.

In order to compare the explicit models with the exact model, an error parameter is defined as:

$$\delta = \frac{(T_d / T_\infty - 1)_{\text{explicit}} - 1}{(T_d / T_\infty - 1)_{\text{exact}}} .$$  \hspace{1cm} (2.5.7)
This error parameter gives the deviation of the temperature difference. However, it is also a good approximation for the deviation of the growth rate, as can be seen from Eq. (2.5.1). The error parameter is shown in Fig. (2.5.3) for the three explicit models as a function of the temperature and the saturation ratio for the case of a water droplet in humid nitrogen at a pressure of 1 bar. The droplet is assumed to be sufficiently large in order to neglect the Kelvin effect. The droplet-growth models discussed are based on a diffusive transport of vapour. Therefore, these models may not be applied to situations where the vapour pressure approaches the total pressure. Just to give an indication, the arbitrary chosen curve $p_{\infty} = 0.5 p_\infty$ is drawn in the diagrams. The deviation parameter $\delta$ may be approximated according to the equations listed in Table (2.5.3), as explained in Appendix D. The error approximation for the explicit model of Barrett and Clement (1988) is also derived by Mozurkewich (1986). The trends shown in the diagrams of Fig. (2.5.3) are confirmed by the approximations of Table (2.5.3).

Consider first the models of Barrett and Clement (1988) and Mason (1953). The diagrams for $\delta$ show great similarity. Obviously, the extra assumption of Mason (1953), i.e. $p_{\infty} \ll p_\infty$, has no serious consequences. On the contrary, it has a positive effect on the accuracy of the explicit model. As can be seen from Table (2.5.3), the assumption introduces an additional term in the equation for $\delta$. Since this term is negative and proportional to $x_\infty$, it enlarges the error with an

<table>
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<td>$D_m, \lambda, L, Ke$ may be evaluated at $T_\infty$</td>
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<td>$T_d \approx T_\infty$</td>
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<tr>
<td>$p_{VS}(T_d) \approx p_{VS}(T_\infty)$</td>
<td>B &amp; C, Mason</td>
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<td>$p_v \approx p_{\infty}$</td>
<td>Gyarmathy</td>
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<td>$p_g \approx p_\infty$</td>
<td>Mason</td>
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Table 2.5.2. Assumptions regarding the explicit growth models of Barrett and Clement (1988), Mason (1953) and Gyarmathy (1963). See Appendix D for more details.
Fig. 2.5.3. Percentage deviation $100 \cdot \delta$ for water and nitrogen, with $\delta$ defined by Eq. (2.5.7), as a function of $T_\infty$ and $X_\infty$, for the case $p_\infty = 1$ bar and $K_e \ll 1$. Models: a. Barrett and Clement (1988), b. Mason (1953), c. Gyarmathy (1963), d. Modified Gyarmathy (1969), explicit growth model according to Eq. (2.5.8).

Dashed line: models of Barrett & Clement and Gyarmathy have deviations of equal magnitude. Dashed-dotted line: $p_{v_\infty} = 0.5 p_\infty$.

Insignificant value for an evaporating droplet, but it has a small positive effect on the deviation in the case of condensation. For both models, the behaviour of $\delta$ is dominated by $C_1$ for the lower temperature range. Since $C_1$ decreases with the temperature, the deviation of these models will increase with the temperature. For higher temperatures, an asymptotic value for $\delta$ is reached independent of the temperature. A crude estimate can be obtained by using $C_1 \ll C_2$ and $C_2 \gg 2$, which then results in $\delta \approx 0.5 f(x_\infty, K_e \infty)$.

The model of Gyarmathy (1963) has an error diagram totally different from the two other models. Due to the behaviour of $C_1$, as depicted in Fig. (2.5.1) for a typical case, the error diagram shows relatively large errors for lower temperatures and higher accuracy for the high temperature range. Also, for an evaporating droplet in a gas mixture with a low vapour content, the model is inaccurate. This is
The parameter $\delta$ is defined by Eq. (2.5.7), the parameters $C_1$ and $C_2$ are specified in Table (2.5.1).

confirmed by the error approximation of Table (2.5.3), since if $\chi_\infty \to 0$, then $\delta \to 0.5 (\ln \chi_\infty - Ke_\infty)$. Further, there exists another curve of zero error beside the curve $\chi_\infty = 1$. This curve results from an exact balance of the errors due to the two fundamental assumptions made by Gyarmathy (1963), i.e. $p_v = \rho_{v\infty}$ and $T_d \approx T_\infty$.

It can be obtained from Table (2.5.3) that an approximation for the curve is: $C_1 = \sqrt{2C_2}$, or equivalently, $T_\infty (K)^3 \approx 4250^3 \sqrt{V(J)}$ in the case of water and nitrogen.

An attempt to obtain a more accurate explicit equation is to incorporate the approximate error given by Table (2.5.3) into the equation:

$$
\frac{T_d}{T_\infty} - 1 = f(\chi_\infty, Ke_\infty) [C_1 + C_2]^{-1} (1 - \delta).
$$

(2.5.8)
The result of this first-order correction is shown in Fig. (2.5.3d) for the model of Gyarmathy (1963). It shows a remarkable improvement with respect to the uncorrected equation.
3.1 Introduction

As explained in Chapter 1, an unsteady rarefaction wave propagating into a gas-vapour mixture may produce a supercooling of the vapour. When foreign particles are present in the mixture, they will act as condensation centres to precipitate the collapse of the metastable vapour state. Then, dropwise condensation will occur. Droplet formation on insoluble particles and non-equilibrium droplet growth were discussed in Chapter 2. A first investigation of the effect of phase transition on the unsteady rarefaction wave in a gas-vapour mixture provided with condensation nuclei will be reported in this chapter. First of all, the complete set of equations describing wave propagation in continuously varying two-phase flows will be presented, consisting of a combination of the Euler equations and the droplet-growth equations. Then, a short intermezzo explains the influence of phase transition on the entropy and the sound speed of the flow. Next, a transformation of the wave equations into the characteristic form invites to making a simple analysis of the wave problem in order to clearly demonstrate the effect of condensation on the flow. And finally, the asymptotic rarefaction-wave solution is considered that develops when the relaxation time of condensation becomes negligibly small with respect to the time. A part of this study is devoted to the steady condensation front.

3.2 The wave equations

Set of equations

We now consider a mixture of gas and vapour. Spherical condensation nuclei of uniform size are suspended in the mixture. The nuclei have a radius $r_n$ and a number density $n_n$. A change in the state of the gas-vapour mixture may result in the formation of droplets on the nuclei. The instantaneous-wetting model, explained in Section 2.2, will be adopted to describe this process. In the case that droplets have formed on the nuclei, we assume that the droplets do not split up or coalesce. Consequently, the number density $n_d$ of the droplets equals the number density $n_n$ of the nuclei. Moreover, as a consequence of the droplet-formation model
adopted, all droplets are of the same size locally. Further, we assume that the droplets are sufficiently small in order to neglect the velocity difference between the droplets and the carrier gas as given by Eq. (2.4.43).

Neglecting heat conduction, viscosity and diffusion, one-dimensional wave propagation is described by the Euler equations (Thompson, 1984):

\[
\begin{align*}
[\rho]_t + [\rho u]_x &= 0, \quad (3.2.1) \\
[\rho u]_t + [\rho u^2 + p]_x &= 0, \quad (3.2.2) \\
[\rho \left( \frac{1}{2} u^2 + e \right)]_t + [\rho u \left( \frac{1}{2} u^2 + e + p/\rho \right)]_x &= 0, \quad (3.2.3)
\end{align*}
\]

where the subscripts \( t \) and \( x \) refer to partial derivatives with respect to the time and the position, respectively. Phase transition influences wave propagation dominantly via the internal energy \( e \) of the mixture and as a minor effect via the total pressure \( p \), conform to Eqs. (2.2.22), (2.2.19) and (2.2.20):

\[
e = f_e c_v T f_c c_v^T + f_d c_v^T d + f_v L_0, \quad (3.2.4)
\]

\[
p = \rho \bar{u} T = \rho (f_g R_g + f_v R_v) T. \quad (3.2.5)
\]

We assume that, if droplets are present, they are in a state of a wet-bulb equilibrium with the gas-vapour mixture. Then, the temperature \( T_d \) of the droplets is given by the implicit wet-bulb equation, Eq. (2.5.2), formally expressed here as:

\[
T_d = T_{wb}(T, p, e, r_d), \quad (3.2.6)
\]

where \( \rho_v/p = f_v R_v/\bar{u} \). According to Eq. (2.2.14) and Eq. (2.2.16), the droplet mass fraction \( f_d \) is interrelated to the droplet radius \( r_d \) by:

\[
f_d = \left( \frac{n_d}{\rho} \right) \rho \frac{4}{3} \pi r_d^3. \quad (3.2.7)
\]

Along particle paths, the quantities \( f_v + f_d (=1-f_g) \) and \( n_d/\rho \) are conserved:

\[
\frac{D}{Dt}(f_v + f_d) = 0, \quad \frac{D}{Dt}(n_d/\rho) = 0, \quad (3.2.8)
\]

where \( D/ Dt \) denotes the material derivative: \( D/ Dt = \partial/\partial t + u \partial/\partial x \). Finally, the set of equations is completed with the droplet-growth equation, Eq. (2.5.1), written here for obvious reasons with the material derivative:
Using the set of equations given above, supplemented with the correct initial and boundary conditions, wave propagation in a gas-vapour mixture with condensation or evaporation may be calculated as long as the flow field remains continuous. In the case of flow discontinuities, the differential conservation laws are not applicable and the integral conservation laws must be applied across every discontinuity. Some types of flow discontinuities are described in Section 3.4 and Chapter 4.

**Entropy production**

It is well known that an adiabatic flow of an inert gas (mixture) with negligible heat conduction and viscosity is isentropic. In the case of a flow with phase transition, entropy will in general be produced, since on the droplet micro-scale heat and vapour transport takes place towards or from the droplets by heat conduction and diffusion. Only when the gradients of temperature and vapour concentration around a droplet are sufficiently small, these transport processes may be considered as near-equilibrium processes and the production of entropy is negligible. An equation for the rate of entropy production in the adiabatic flow of a gas-vapour-droplet mixture with condensation or evaporation can easily be derived.

By manipulation of the conservation equations (3.2.1) to (3.2.3), and using $\dot{h} = \frac{1}{\rho} \frac{Dp}{Dt}$, the energy equation (3.2.3) can be transformed into:

\[
\frac{Dh}{Dt} = \frac{1}{\rho} \frac{Dp}{Dt} = 0 ,
\]

which only says that a change of the state of a material element of the flow is adiabatic. As performed in Appendix E, combining this equation and the fundamental thermodynamic relation for a unit of mass of the gas-vapour-droplet mixture, yields an approximate equation for the rate of entropy production along a particle path:

\[
\frac{Ds}{Dt} \approx \frac{K_v}{\rho} [\ln \chi - K_e(T, r_d)] \frac{Df}{Dt} d. 
\]
It is easily recognized that $\frac{D\xi}{Dt}$ is essentially positive, since for a free relaxation process the term between square brackets and $\frac{Df_d}{Dt}$ have the same sign. In the case of $\chi \approx 1$, equilibrium processes prevail and indeed, the production of entropy reduces to zero.

**The sound speed**

Generally, the sound speed is defined as the square root of the derivative $\frac{\partial p}{\partial \rho}$ at constant entropy. However, in the case of a gas-vapour mixture with droplets the result of this definition ceases to be unique. The reason is that the state of the mixture has to be described by an additional parameter, since the composition may change due to the rearrangement of phases. If we take the droplet mass fraction $f_d$ as the additional parameter, two uniquely determined sound speeds can be defined.

Firstly, the restriction $f_d = \text{constant}$ may be used to introduce the frozen speed of sound:

\[
\tilde{a}_f^2 = \left(\frac{\partial p}{\partial \rho}\right)_s f_d,
\]

which represents the speed at which a wave disturbance travels relative to the flow, when the internal relaxation process is too slow to interact with the disturbance. The frozen sound speed is appropriate to describe wave propagation in a relaxing flow or in a flow without droplets.

However, demanding that $f_d$ equals at all time the equilibrium mass fraction $f_{de}$, the equilibrium speed of sound is obtained:

\[
\tilde{a}_e^2 = \left(\frac{\partial p}{\partial \rho}\right)_s f_d = f_{de}.
\]

Disturbances, inducing a slow variation of the state so that the equilibrium can be preserved, propagate relative to the flow at the equilibrium sound speed. The usage of this sound speed is only meaningful in the case of an equilibrium two-phase flow. The equilibrium sound speed is always smaller than the frozen sound speed.

Straightforward thermodynamics yields for a gas-vapour mixture with or without droplets:

\[
\tilde{a}_f^2 = \frac{\gamma}{\gamma - 1} T,
\]

\[
\tilde{a}_e^2 = \tilde{a}_f^2 \frac{1 + [(\gamma - 1)/\gamma] \left(\frac{\xi}{1 - 1}\right)^2 \left(\frac{p_v}{p}\right)}{1 + (\gamma - 1) \left(\frac{\xi}{1 - 1}\right)^2 \left(\frac{p_v}{p}\right)},
\]

where the mixture parameters $\gamma$ and $\tilde{\gamma}$ are conform Eqs. (2.2.19). The local
parameter \( \zeta = \frac{L}{\mathbb{R}_v} T \) is introduced only to facilitate reading. For water, it may be approximated by \( \zeta \approx 18 \left( \frac{295}{T(K)} \right) \), with typical values in the range 15 to 20. Of course, in accordance with the equilibrium requirement, the vapour pressure in Eq. (3.2.15) is determined by the temperature: \( p_v = p_{vs}(T) \). In the case of a major fraction of inert gas, i.e. \( f_g \approx 1 \), the ratio of the specific heats and the specific gas constant of the mixture may be approximated by those of the inert component. Then, the sound-speed ratio \( \frac{a_e}{a_f} \) is only depending on the temperature and the pressure and varies approximately in the range:

\[
\frac{1}{\gamma_g} \approx 0.7 < \frac{a_e}{a_f} < 1.
\] (3.2.16)

The equation for the equilibrium sound speed presented by Sislian (1975) corresponds with Eq. (3.2.15), and for the limiting case of a pure vapour, Eq. (3.2.15) reduces to Wegener's equation (Wegener, 1975), taking into account that both authors use the premise that \( L \) is constant.

**Characteristic equations**

The Euler equations (3.2.1) to (3.2.3) can be rewritten into the characteristic equations. The procedure is outlined by Becker (1969) for a flow with relaxation. The result is:

\[
dp + \rho a_f du = S dt \quad \text{along } \mathbb{C}^+; \quad dx/dt = u + a_f,
\]

\[
dp - \rho a_f du = S dt \quad \text{along } \mathbb{C}^-; \quad dx/dt = u - a_f,
\] (3.2.17)

\[
dp - a_f^2 d\rho = S dt \quad \text{along } \mathbb{C}^u; \quad dx/dt = u.
\]

Here \( a_f \) is the frozen sound speed according to Eq. (3.2.14). The source term \( S \) is due to phase transition and is positive for condensation and negative for evaporation. It is given by:

\[
S \approx \frac{a_f^2}{\rho} \left( \frac{\partial h}{\partial f_d} \right) \frac{\partial \rho}{\partial h} \phi_{f_d}(p, \rho, f_d).
\] (3.2.18)

The relaxation function \( \phi_{f_d}(p, \rho, f_d) \) is the rate of change of the droplet mass fraction \( f_d \) inside a material element. From Eqs. (3.2.7), (3.2.8b) and (3.2.9) it follows that:

\[
\phi_{f_d}(p, \rho, f_d) = \frac{n_d}{\rho} \frac{2 \tau_d N_{ud} a^l}{L} (T_d - T).
\] (3.2.19)
3.3 A simple analysis

In order to enlighten on some aspects of the phenomenon of an unsteady rarefaction wave propagating in a gas-vapour mixture and inducing condensation, we will use the characteristic equations (3.2.17) in linearized form. Therefore, we introduce a reference state denoted by subscript $o$, and express the state variables as disturbances relative to the reference state: $p' = p - p_o$ and $u' = u - u_o = u$, since we shall put $u_o = 0$. Then, the characteristic equations become:

\[
\begin{align*}
    p_2' + \rho_o a_{f0} u_2' &= p_1' + \rho_o a_{f0} u_1' + \int_{t_1}^{t_2} S \, dt \quad \text{along } C^+ : \frac{dx}{dt} = a_{f0}, \\
    p_2' - \rho_o a_{f0} u_2' &= p_1' - \rho_o a_{f0} u_1' + \int_{t_1}^{t_2} S \, dt \quad \text{along } C^- : \frac{dx}{dt} = -a_{f0}.
\end{align*}
\]

(3.3.1)

The problem we shall now consider is that of a rarefaction wave travelling to the left into a region of uniform state, which will be the reference state $o$. At the boundary $x = 0$, a constant positive velocity of the flow is prescribed: $u'(x=0) = u'_b$. The solution in a point $(x, t)$ of the flow field can be derived by constructing the characteristic lines originating from the uniform region and leading to the point of interest. An illustration is presented in Fig. (3.3.1). If one of the paths runs via the boundary, then, by applying Eqs. (3.3.1) to the situation, we find:

\[
p' = -\rho_o a_{f0} u'_b + \frac{1}{2} \left[ \int_{0}^{b} S \, dt + \int_{t_b}^{t} S \, dt + \int_{0}^{t} S \, dt \right],
\]

(3.3.2)
Here the integration paths and the time $t_b$ are defined in Fig. (3.3.1).

First, in the case that the medium consists of an inert gas (mixture), the source term $S$ equals zero and the solution is very simple:

$$
 u' = 0, \quad p' = 0 \quad \text{when} \quad x < a_{f_0} t, \quad \text{when} \quad x \geq a_{f_0} t.
$$

Here we have introduced $p_f'$ as the pressure behind the rarefaction wave in the case of frozen relaxation $(S=0)$. It may be observed that, because of the linear model, dispersion is absent and the rarefaction wave remains a discontinuous jump, travelling with speed $a_{f_0}$.

Second, in the case that we are dealing with a rarefaction wave in a gas-vapour mixture, dropwise condensation of vapour may occur. Whether it actually occurs depends on the depth of the expansion. According to the model for droplet formation explained in Section 2.3, condensation on foreign nuclei will be initiated when the pressure is lowered below the pressure $p_c$ of the critical state of droplet formation, even if it is just for a short instant. Thereupon, the mixture will relax to a new equilibrium with droplets. In the zone of relaxation, the source term $S$ will be non-zero. In the simple treatment of this section, we shall propose a simple but adequate relaxation model. We shall assume, that once the condensation is initiated in a material element of the flow, it will last for exactly a time $T$. Further, during relaxation the source term is considered to have a constant value.

The solution of this wave problem is depicted in the $(x, t)$ diagram of Fig. (3.3.2). The front of the wave travels along the $C^-$ characteristic $x = -a_{f_0} t$. As a result of the linearization, the particle paths coincide with the lines $x = constant$. We assume that initially the pressure after expansion is sufficiently low to start the relaxation process in the accelerated flow elements. Subsequently, relaxation lasts for a time $t$. The relaxation zone, where $S$ is constant and non-zero, is shown in Fig. (3.3.2) as the shaded area. By using Eq. (3.3.2), we can deduce for the pressure in the relaxation zone:

$$
p' = p_f' + \frac{1}{2} S t \quad \text{when} \quad t \leq t_c, \quad (3.3.5)
$$

meaning that the pressure is uniform in the relaxation zone and increases linearly.
with time due to relaxation. Here $t_c$ is the time:

$$t_c = (2/S) \left( p_c' - p_f' \right),$$  \hspace{1cm} (3.3.6)

when the pressure in the relaxation zone reaches the critical value. At that instant of time, the initiation of condensation is stopped on the leading characteristic of the relaxation zone, since the nucleation condition is no longer fulfilled. A contact discontinuity develops, travelling with the flow, to the right of which condensation is proceeding, while to the left the pressure is above critical, so no droplets are formed. It is interesting to deduce the time history of the pressure along this particle path, i.e. $x = -a_{f0} t_c$, by using Eq. (3.3.2). The result is depicted in Fig. (3.3.3). The time is divided into four intervals. The first interval corresponds to the time before passage of the wave. The pressure is undisturbed. Then, at $t = t_c$, the pressure drops to the critical value, or slightly above, which is insufficient to initiate condensation. So time interval II is a relaxation-free interval in which the pressure is above critical. The duration of the interval is equal to $\tau$. Entering the third interval, the pressure drops to a low value, since hardly any non-zero sources are found on the characteristics leading to the corresponding point in the $(x, t)$ diagram. The pressure is now below the critical value and relaxation starts lasting for a time $\tau$ until a new equilibrium is established in interval IV. Here, we have the remarkable fact that the relaxation zone has shifted away from the front of the wave, as can also be observed in Fig. (3.3.2). From $t = t_c$ onwards, the pressure starts to rise along the line $x = -a_{f0} (t - \tau)$ due to relaxation until again the critical value is reached. Again the condensation is stopped and the relaxation zone is shifted with respect to the front of the wave. This process repeats itself periodically.

Due to the linear wave model adopted in this section, some aspects of unsteady expansion flow with condensation are left out of consideration. As a result of the linearization, dispersion of the rarefaction wave does not occur. Neither does heat addition inside the relaxation zone lead to a curving of the characteristics or the formation of a shock wave. Nevertheless, the interaction of condensation with the flow is clearly illustrated. On passage of the rarefaction wave, the state of the mixture changes to supercritical, initiating a relaxation process. Subsequently, heat addition acts on the flow as described by Eqs. (3.3.2) and (3.3.3). As shown, a positive value of the source term $S$, corresponding to condensation, results in an increasing pressure inside the relaxation zone. Moreover, at some time $t = t_c$, which is obviously beyond the time range considered in Fig. (1.2.3) of Section 1.2, an interesting instability develops in the flow. Due to the rise of the pressure in the relaxation zone, the initial state after expansion ceases to be supercritical, the
Fig. 3.3.2. Solution of the linearized wave problem depicted in the \((x, t)\) diagram. Shaded areas: relaxation region, solid lines: characteristics, dashed lines: particle paths. The roman numbers indicate distinguishable intervals for the particle path \(x = -a_f t_c\) (explained in text).

Fig. 3.3.3. Pressure history along the particle path \(x = -a_f t_c\).

Roman numbers: I: before passage of the wave, II: pressure is above critical: no condensation, III: relaxation, IV: equilibrium.

Initiation of condensation stops and then restarts at some distance from the front. This phenomenon is a direct consequence of the simplifying assumption of this section that the source term \(S\) attains a constant non-zero value as soon as the critical state is passed by isentropic expansion from an unsaturated state. Let us see what happens along the leading characteristic of the expansion zone when we apply the droplet formation and growth model of Chapter 2. According to this model, the source term \(S\) does not have a constant value but is proportional to a driving force of condensation depending on the droplet radius and the saturation ratio.
Approximately, the driving force is: \( \ln x - \text{Ke}(r_d) \). Fluid elements, arriving at the front characteristic, will contain droplets that have a size equal to the size of the condensation particles, since condensation has not yet occurred. Further, the states on this characteristic are attained by isentropic expansion. Therefore, the saturation ratio is dependent on the pressure only. In a linear model, the dependence is one of negative proportionality. Now, at time zero, the pressure will have its frozen value on the first characteristic. We will assume that the corresponding saturation ratio is sufficiently high to give a positive value of the source term \( S \). As a result, the pressure increases along the characteristic. However, the saturation ratio decreases simultaneously, thus diminishing the source term \( S \). The rise of the pressure will proceed at a continually slower rate and the critical state will never be reached, since at that state the driving force of condensation, and thereby the source term \( S \), is zero by a balance of the effects of the saturation ratio and the droplet radius. Therefore, we have to conclude that the instability does not occur when we apply a linear wave model combined with a more realistic relaxation model. However, as will be shown in Section 8.3, non-linear effects will induce the flow instability already described in this section.

### 3.4 The asymptotic rarefaction wave

#### Qualitative description of the asymptotic rarefaction wave

As illustrated thus far, the adiabatic expansion of a gas-vapour mixture by an unsteady rarefaction wave is accompanied by non-equilibrium condensation. It appeared that condensation has its influence on the structure of the wave. Hence, it is interesting to investigate the solution to the wave problem in the asymptotic limit.

First, consider a fluid sample of gas and vapour that is being accelerated and expanded adiabatically by a rarefaction wave. The sample is assumed to contain spherical condensation nuclei of uniform size. Then, according to the concepts presented in Section 2.3, as soon as the critical state is attained, all condensation nuclei become active instantaneously and condensation is initiated. The subsequent non-equilibrium condensation process is a typical relaxation process, since the state of the mixture is changed by heat addition and vapour depletion until equilibrium

\(^1\)Part of this analysis has already been published (Smolders, Niessen and Van Dongen, 1989).
is restored in the fluid sample. The total elapsed time between reaching the critical state and a state of near-equilibrium is determined by the combined action of the phase-transition and expansion processes. At first instant, the total time of non-equilibrium condensation is relatively short, due to large gradients inside the wave. As gradients weaken with time, the condensation time increases moderately and finally reaches a constant value determined by the degree of non-equilibrium at the critical state. Anyhow, for the purpose of explanation, we introduce a characteristic time of relaxation $\tau$, chosen to have a constant value of the order of the total time of non-equilibrium condensation.

Next, we turn our attention to the rarefaction wave. At some point in the wave the state of the mixture is critical. Beyond that point the relaxation zone extends up to a point of near-equilibrium. The width of the relaxation zone is of the order of a characteristic length $L$, for example defined as $L \equiv a_0\tau$, with $a_0$ as the sound speed in the initial state. Therefore, the width of the non-equilibrium zone is approximately constant. Since the width of the rarefaction wave increases in proportion to time, the non-equilibrium zone will reduce to a virtual discontinuity in the asymptotic limit $t/\tau \rightarrow \infty$. Moreover, if the expansion process is continued behind the non-equilibrium zone, condensation in that part of the wave will essentially take place at equilibrium conditions, since the wave-induced change of state occurs at a very low rate in the asymptotic limit.

Another characteristic of the asymptotic wave solution is easily derived. In general, the solution depends on the independent variables $x$ en $t$. Or, in a rearranged dimensionless fashion: $x/a_0t$ and $\tau/t$. Taking $t/\tau \rightarrow \infty$, we find that the asymptotic solution only depends on the similarity variable $x/a_0t$, since the variable $\tau/t$ reduces to zero. So the asymptotic solution is self-similar. Again, as in the case of the frozen solution, the lines of invariance of state in the wave diagram form a centred fan of straight lines.

Collecting the conclusions reached so far, we are now able to give a qualitative description of the asymptotic rarefaction wave. In the asymptotic limit, the unsteady rarefaction wave in a gas-vapour mixture containing foreign particles is self-similar. Up to the point of the critical state, vapour condensation is absent in the rarefaction wave. The transition from the critical supersaturated state to a state of gas-vapour-droplet equilibrium occurs in a very sharp front, which may be approximated by a discontinuity in the flow field. Equilibrium condensation occurs in the remaining part of the rarefaction wave behind the condensation discontinuity. Inside the transition zone, non-equilibrium processes prevail. Thus, while changes of state are isentropic in the continuous regions of the wave, entropy is produced in the condensation front. The strength of the discontinuity depends on
the value $\chi_c$ of the saturation ratio in the critical state. A special case exists when $\chi_c = 1$. Then, the critical state is identical to the saturated state and a non-equilibrium condensation zone does not exist. In this case, the full rarefaction wave is continuous and isentropic.

**The steady condensation zone**

In the preceding subsection, it was argued that in the asymptotic limit the non-equilibrium condensation zone can be considered as a virtual discontinuity embedded in a self-similar continuous wave. Thus, the states in front and behind the transition zone are essentially constant. Moreover, we know that condensation rates inside the zone are finite, since the saturation ratio has an upstream critical value differing from unity by a fixed and finite amount. A combination of both facts then leads to the conclusion that the condensation process inside the non-equilibrium zone may be treated steady in the reference frame fixed to the zone.

The questions to be answered with respect to the steady condensation zone are twofold. In order to estimate the distance an unsteady rarefaction wave has to travel before it approaches its asymptotic form, the characteristic thickness of the steady condensation zone has to be determined. This can be done by considering the internal structure of the zone. On the other hand, modelling the transition as a discontinuous jump in the state of the mixture allows the determination of possible state transitions and front propagation speeds. This information can thereupon be used in a detailed description of the asymptotic rarefaction wave.

Let the velocity of the condensation front and the velocity of the mixture in the laboratory-fixed reference frame be denoted by $U$ and $u$, respectively. Then, the velocity $v$ of the mixture in the reference frame fixed to the condensation zone follows from:

$$v = u - U.$$  \tag{3.4.1}

Further, let the supersaturated vapour state in front of the transition zone and the final state of liquid-vapour equilibrium be referred to as state 1 and state 2, respectively. It is clear, there exists no liquid phase in state 1, i.e. $f_{d_1} = 0$, thus the vapour mass fraction is equal to the total mass fraction of the condensable component of the mixture: $f_{v_1} = f_{v_0}$. Moreover, the saturation ratio in state 1 has the critical value: $\chi_1 = \chi_c$. State 2 is an equilibrium state, so the vapour pressure $p_{v_2}$ has to satisfy the Rankine-Kirchhoff equation (2.2.8).

An arbitrary state inside the condensation front can be obtained by choosing a
value: $0 \leq f_d \leq f_{d2}$, and calculating the remaining variables by means of the steady integral laws of conservation of mass, momentum and energy:

$$
\rho v = \rho_1 v_1 = \Psi ,
$$

$$
p - p_1 = -\Psi^2 \left( \frac{1}{\rho} - \frac{1}{\rho_1} \right) ,
$$

$$
h - h_1 = \frac{1}{2} (p - p_1) \left( \frac{1}{\rho} + \frac{1}{\rho_1} \right) .
$$

(3.4.2)

Here $\Psi$ is a constant representing the mass flux through the transition zone. The specific enthalpy $h$ of the mixture can be written as:

$$
h = (1 - f_{v0}) c^v_s T + f_{v0} (c_{ps} T + L_0) - f_d L(T) + f_d c^v_s (T_d - T) .
$$

(3.4.3)

The term $f_d L(T)$ represents the latent heat released due to condensation of vapour. The last term is due to a temperature difference between the droplets and the gas mixture. Under wet-bulb conditions this term is negligible mostly. The position $\Delta x$ inside the zone corresponding to the calculated state can be found by transforming:

$$
\frac{df_d}{dx} = \frac{1}{v} \frac{df_d}{dt} = \frac{f_d}{v} ,
$$

(3.4.4)

into an integral equation:

$$
\Delta x = \int_0^1 \left( \frac{v}{\phi f_d} \right) df_d .
$$

(3.4.5)

The rate of change $\phi f_d$ of the droplet mass fraction inside a material element is given by Eq. (3.2.19). It depends on the droplet radius $r_d$ and the droplet number density $n_d$. These parameters can be obtained with:

$$
\frac{n_d}{\rho} = \frac{n_{d0}}{\rho_0} \quad \text{and} \quad r_d^3 = \frac{3 f_d \rho}{4 \pi n_d \rho \phi} ,
$$

(3.4.6)

with $n_n$ as the number density of the condensation nuclei and subscript $0$ referring to some initial state. Finally, Eq. (3.4.5) can be used to determine the characteristic width $L$ of the condensation front, that we choose to define as:

$$
L \equiv \Delta x (\Lambda = 0.1) \quad \text{with} \quad \Lambda \equiv \frac{\delta}{\chi c - 1} .
$$

(3.4.7)
Fig. 3.4.1. The structure of the steady condensation zone for a mixture of water vapour and nitrogen gas in the case of a Chapman-Jouguet expansion transition. a. Pressure, b. Temperature, c. Saturation ratio, d. droplet mass fraction.

Curves 1 to 5 correspond to $\chi_c = 1.05, 1.5, 2, 5$ and 10. State 1 is isentropically related to a reference state $0$: $p_0 = 1$ bar, $T_0 = 295$ K, $f_{v0} = 0.0136$, $n_{n0} = 10^{12}$ m$^{-3}$.

The internal structure of the condensation front has been calculated for a mixture of nitrogen gas and water vapour at typical conditions. The result is depicted in Fig. (3.4.1). The profiles of the pressure $p$, the temperature $T$, the saturation ratio $\chi$ and the droplet mass fraction $f_d$ are given for five different values of $\chi_c$ ranging from 1.05 to 10. The final downstream values of the variables shown are indicated by bars. State 1 in front of the zone differs for all cases, but is isentropically related to a common initial state: $p_0 = 1$ bar, $T_0 = 295$ K, $f_{v0} = 0.136$, $n_{n0} = 10^{12}$ m$^{-3}$. All calculated transitions correspond to Chapman-Jouguet expansion transitions, which means that the upstream flow is subsonic and the flow downstream of the front is exactly sonic. First of all, it may be observed that the condensation process inside the zone is a typical relaxation process. The non-equilibrium parameter, i.e. the supersaturation $\chi - 1$, tends to zero by the formation of condensate mass. At the position $\Delta x = L$, the supersaturation has reduced to one
tenth of its original value. The changes in the pressure and the temperature are moderate. The change in the density can be obtained from the pressure profile by means of Eq. (3.4.2). It can easily be seen that the density decreases monotonically in accord with the behaviour of the pressure. The profile of the temperature is more interesting. It results from the combined action of heat addition and expansion. For high values of $\chi_c$, condensation is dominant in the first part of the condensation zone, resulting in a rise in the temperature. In the last part, equilibrium is almost attained and a small decrease in the temperature is observed due to the final expansion process. For very low values of $\chi_c$, the heat addition is moderate in the whole zone, so the temperature shows a monotonic decrease. Whether the net effect is that the mixture is heated or cooled in the transition depends on the amount of latent heat released compared to the work of adiabatic expansion.

The characteristic width of the condensation zone is strongly related to the rate of condensation. Therefore, a dependence on the critical supersaturation $\chi_c^{-1}$ and the initial nuclei number density $n_{n_0}$ is expected. Fig. (3.4.2) shows the variation of $L$ with $\chi_c^{-1}$ and $n_{n_0}$. The characteristic width appears to decrease with increasing critical supersaturation. The theoretical curve may be approximated on the considered interval by an inverse square-root dependence: $L \sim 1/\sqrt{\chi_c^{-1}}$. The relation between $L$ and $n_{n_0}$ is quite well explainable. To that end, consider the condensation rate $\dot{f}_d$ given by Eq. (3.2.19). If we neglect the Kelvin and Knudsen effects, the droplet temperature is independent of the droplet radius. Then, using Eqs. (3.4.6), it can be shown that: $\dot{f}_d^3 = \left(\frac{n_{n_0}}{\rho_0}\right)^2$, so that: $L^3 = \left(\frac{\rho_0}{n_{n_0}}\right)^2$. Indeed, the theoretical curve shows such behaviour for small values of $n_{n_0}$. The deviation for higher values is due to droplet-size effects on the temperature of the droplets, which slows down the condensation process.

Thus far, we were concerned with the internal structure and width of the steady relaxation zone. Now, we shall consider the transition zone as a condensation discontinuity. Such an approach allows the systematic study of possible transition modes. Similar approaches were made by Belenky (1945) and Heybey (1947) to describe the non-equilibrium condensation zone in a Laval nozzle operated with moist air. Basically, the flow was considered to be the flow of a perfect gas (mixture) with constant thermal properties. The effect of condensation was merely to add heat to the flow. Thereupon, the amount of heat $Q$ added to the flow inside the jump was left as an undetermined parameter in the conservation equations of the condensation discontinuity. As a result, the model was very

---

2 Kelvin effect: the influence of the surface tension on the equilibrium of liquid and vapour. Knudsen effect: the decrease in the Nusselt numbers with an increase in the Knudsen number.
elegant and served pre-eminently to study qualitative features of condensation jumps. However, for our purposes a more detailed model is required. In our treatment, we shall put \( Q = f_{d_2} L_2 \) in accordance with Eq. (3.4.3) and let \( f_{d_2} \) be determined by the phase equilibrium in state 2.

The premises formulated in the beginning of this subsection will be maintained. That is, the state in front of the discontinuity is a supersaturated vapour state and the state behind is one of liquid-vapour equilibrium. To describe the jump, Eqs. (3.4.2) and (3.4.3) are applicable again. Only, the unsubscribed state variables have to be replaced by the variables of state 2. Moreover, it is advantageous to introduce Mach numbers for state 1 and 2. Since state 1 is a state without droplets and state 2 is a state of phase equilibrium, we shall use the corresponding sound speeds:

\[
Ma_1 = \frac{|u_1 - U|}{a_{f1}}, \quad Ma_2 = \frac{|u_2 - U|}{a_{e2}}. \tag{3.4.8}
\]

A numerical evaluation of the condensation discontinuity has been performed for several values of the critical saturation ratio \( \chi_c \). State 1 is related isentropically to a fixed reference state 0. In Fig. (3.4.3) the Rankine-Hugoniot curves and the \( Ma_2 - Ma_1 \) relations are shown for a mixture of water vapour and nitrogen gas\(^4\). Only those parts of the curves are shown that correspond to an entropy increase, to a real mass flux and to a positive droplet mass fraction downstream the discontinuity. The Chapman-Jouguet points, defined by \( Ma_2 = 1 \), separate the curves in four different regions:

- strong compression : \( Ma_1 > 1 \), \( Ma_2 < 1 \),
- weak compression : \( Ma_1 > 1 \), \( Ma_2 > 1 \),
- weak expansion : \( Ma_1 < 1 \), \( Ma_2 < 1 \),
- strong expansion : \( Ma_1 < 1 \), \( Ma_2 > 1 \).

Following the arguments of Landau and Lifshitz (1959), strong compression and strong expansion discontinuities are excluded as possible solutions. Consider first the strong compression discontinuity. The flow in front of this type of

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\(^3\)In the diabatic flow approach frozen Mach numbers are used as a natural consequence of the assumption of perfect gas flow. However, a frozen Mach number is inappropriate to describe equilibrium two-phase flow.

\(^4\)A Rankine-Hugoniot curve represents all states that can be reached across a condensation discontinuity starting from a specified initial state. The heat released by condensation depends on the transition and is not at all invariant when going along a Rankine-Hugoniot curve. In this the diabatic flow approach to describe condensation discontinuities is inaccurate.
Fig. 3.4.2. Characteristic width of the steady condensation zone for a mixture of water vapour and nitrogen gas. Curve I: as a function of the supersaturation, curve II: as a function of the initial nuclei number density. State 1 is isentropically related to a reference state 0: $p_0 = 1$ bar, $T_0 = 295$ K, $f_{v_0} = 0.0136$. Extra conditions: curve I: $n_{no} = 10^{12}$ m$^{-3}$. Curve II: $\chi_c = 2$.

discontinuity is supersonic. So, a precursor wave originating from the discontinuity cannot exist and the upstream flow conditions remain unaffected right up to the discontinuity. Since a condensation front requires a state in front of it that is critical, the consequence is that the position of the discontinuity, or equivalently its velocity, is prescribed by the upstream conditions. Using these premises, the state behind the discontinuity follows uniquely from the conservation laws. However, since the flow behind the discontinuity is subsonic, this will conflict in general with the downstream boundary conditions. Therefore, a strong compression discontinuity is overdetermined and will not exist in general. The reason to exclude the strong expansion discontinuity is different. Consider a small perturbation in the position of the discontinuity. As a result of the displacement, disturbances will be emitted by the discontinuity. Since the upstream flow is subsonic, one sound disturbance may be emitted upstream. The downstream flow is supersonic, so two disturbances may propagate downstream, as well. Moreover, a contact surface may float with the downstream flow. Every disturbance corresponds to a one-parameter
degree of freedom. Summing up the disturbances, including displacement and contact surface, results in a total number of five parameters to be determined. However, the number of restrictions imposed on the perturbation problem is four, viz. the three conservation laws and the onset condition. If we now prescribe the displacement of the discontinuity in a form proportional to $\exp(\omega t)$, it is clear that $\omega$ may take any positive and real value, since the number of parameters exceeds the number of equations. Therefore, a strong expansion discontinuity is unstable. Consequently, the only allowed condensation discontinuities in gasdynamic flow are weak compression and weak expansion discontinuities with the Chapman-Jouguet solutions as limiting cases.

**Quantitative description of the asymptotic rarefaction wave**

In analysing the self-similar solution of an unsteady rarefaction wave in a gas-vapour mixture with condensation, we will first discuss the case that condensation is initiated at a saturation ratio of unity. Then, the change in the state of the mixture is continuous. The solution is fully isentropic and follows from the characteristic form of the Euler equations, written for a left-facing simple wave as:

\[ u + \Gamma = 0 , \]
\[ p \Gamma = \int 1/(\rho a) \, dp , \]  \hspace{1cm} (3.4.9)
\[ p_0 \]
\[ x/t = u - a . \]  \hspace{1cm} (3.4.10)

This set of equations is completed with the isentropic condition, which relates all thermodynamic variables.

The characteristics of an isentropic expansion of a gas-vapour mixture into the two-phase regime are clearly shown in Fig. (3.4.4). In the first stage of the expansion process the mixture behaves as a perfect gas. As soon as the saturated state is attained, the single-phase gas-vapour mixture turns into an equilibrium two-phase mixture. Consequently, as explained in Section 3.2, the sound speed changes at this point discontinuously from the frozen into the equilibrium value. According to the definition of the sound speed, we may write: $(\partial p/\partial v)_S = -(\rho a)^2$, with $v = 1/\rho$, so the isentrope in the $(\rho, v)$ diagram shows a discontinuity in its slope at the saturation point.

The particular behaviour of the mixture during expansion has its influence on the shape of the unsteady rarefaction wave. According to Eqs. (3.4.9) and (3.4.10),
Fig. 3.4.3. The condensation discontinuity for a mixture of water vapour and nitrogen gas. a. Rankine-Hugoniot curve. b. Mach number relation. State 1 is isentropically related to a reference state 0: \( p_0 = 1 \) bar, \( T_0 = 295 \) K, \( f_{v0} = 0.0136 \). Symbols: \( \bullet \): Chapman-Jouguet point, \( \dashrightarrow \): zero droplet mass fraction downstream the discontinuity, \( \rightarrow \): zero entropy increase.

Fig. 3.4.4. Isentropic expansion of a gas-vapour mixture. The pressure and the sound speed are shown as functions of the specific volume. The saturated state is indicated. Symbols: \( F \): frozen expansion, \( E \): equilibrium expansion.

the thermodynamic variable \( \Gamma \) and the flow speed \( u \) are continuous functions of the pressure \( p \). However, the speed of sound shows a discontinuous change when passing the saturated state. The similarity equation (3.4.11) then results in a solution as depicted in Fig. (3.4.5a). Two rarefaction fans are separated by a region of constant state. In this region the vapour is saturated. The tail characteristic of the leading
Fig. 3.4.5.  
Self-similar rarefaction waves. 

a. Self-similar isentropic rarefaction wave, depicted in an \((x,t)\) diagram. 


Fan travels at \(x/t = u_s - a_{fs}\), while the first characteristic of the second fan travels at a speed \(u_s - a_{es}\).

The structure of an unsteady expansion wave in a gas-vapour mixture with equilibrium condensation as described here was not predicted by Sislian (1975). He assumed the initial state in front of the rarefaction wave to be saturated, thus only considering the second part of the wave that travels at the equilibrium speed of sound relative to the flow. The splitting of a rarefaction wave into two parts due to a discontinuity in the speed of sound is not unique to the case of vapour expansion. Zel'dovich and Raizer (1967) discuss the unsteady isentropic expansion of a metal, capable of undergoing a polymorphic transformation, and describe a similar behaviour.

If condensation starts at a fixed critical saturation ratio \(\chi_c > 1\), the change of the supersaturated vapour state to the state of liquid-vapour equilibrium can only be discontinuous. The position of the condensation discontinuity in the self-similar solution is completely determined by the value of \(\chi_c\). As stated in the first part of this section, the discontinuity is either a weak compression, a weak expansion or a Chapman-Jouguet discontinuity of either two types. A weak compression discontinuity and a Chapman-Jouguet compression discontinuity are not allowed, because \(Ma_1 > 1\), which means that the discontinuity should travel at supersonic speed. This cannot be matched with the prescribed value of \(\chi_c\). The Chapman-
Jouguet expansion discontinuity is a possible solution. In this case, $Ma_1 < 1$ and $Ma_2 = 1$. This means that the discontinuity lags behind the leading rarefaction fan, thus creating a uniform region. The second rarefaction fan remains connected to the discontinuity. Finally, we consider the weak expansion discontinuity. Again $Ma_1 < 1$, but now $Ma_2 < 1$, which implies that every left-running wave will overtake and change the discontinuity. A self-similar solution only then exists when the state behind the discontinuity is uniform. Which type of discontinuity occurs depends on the depth of the expansion. Some typical examples of possible self-similar solutions with condensation discontinuities are shown in Fig. (3.4.4b).
Chapter 4. Shock-wave compression of a gas-vapour mixture with droplets

4.1 Introduction

A shock wave, propagating in a gas-vapour-droplet mixture, abruptly induces a situation of non-equilibrium, followed by processes of exchange of momentum, heat and mass between the droplets and the gas-vapour mixture. A sketch of the phenomenon was given in Section 1.3 of Chapter 1. This chapter will be devoted to a more detailed investigation. The first section reveals the mechanisms of relaxation. By attributing characteristic times to the various processes, different stages of the relaxation process will be identified. The analysis will be based on some simplifying assumptions. The droplet mass fraction is taken sufficiently low, so that the gas-phase variables do not alter during relaxation. The droplet size is assumed not to affect the phase equilibrium. And finally, continuum expressions are used to describe the transfer processes. The second section of this chapter concerns a model, based on the conclusions of the first section, describing the evaporation of droplets behind a shock wave. The dispersed shock wave will be modelled as a discontinuity followed by an evaporation zone. The discontinuity represents the change in the state due to the gas shock and the relaxation of momentum and droplet temperature at quasi-constant droplet size. The evaporation zone is the region in which the droplets evaporate under conditions of wet-bulb and momentum equilibrium. The model will also be applicable to small droplets for which size effects come into play and will deal with a change in the gas state due to a significant transfer of heat and mass between the phases.

4.2 The mechanisms and characteristic times of relaxation

Shock compression

The analysis can most conveniently be performed in the reference frame fixed to the shock wave, in which the flow will be considered steady. A sketch was already presented in Fig. (1.3.1) of Chapter 1. It will be assumed that the mixture

¹Part of the analysis presented in this section has already appeared in a slightly different form (Goossens, Cleijne, Smolders and Van Dongen, 1988).
flowing towards the shock wave is in equilibrium, and that the suspended droplets are spherical and equally sized. On passage of the shock wave, the velocity and the state of the gas show a discontinuous change, whereas the droplets pass undisturbed, retaining their original velocity, temperature and size. Especially, the droplets will remain spherical and no deformation or break-up of the droplets will take place. The gas state immediately behind the shock wave is directly related to the initial state by the shock conservation laws. For a perfect gas mixture, these relations are (Thompson, 1984):

\[ \frac{P_2}{P_1} = 1 + \frac{2\gamma'}{\gamma' + 1} (Ma_{ff}^2 - 1) \]
\[ \frac{P_{v2}}{P_{v1}} = \frac{P_2}{P_1} \]
\[ \frac{\rho_{c1}}{\rho_{c2}} = \frac{v_2}{v_1} = 1 - \frac{2 \frac{Ma_{ff}^2}{\gamma' - 1}}{Ma_{ff}^2}, \]
\[ \frac{T_2}{T_1} = \frac{P_2 \rho_{c1}}{P_1 \rho_{c2}} \]

where subscripts 1 and 2 refer to pre- and post-shock state, the velocity of the mixture in the shock-fixed reference frame is denoted by v, and \( \gamma' = c_p^2 / c_v^2 \) is the ratio of the specific heats of the gas-vapour mixture. The shock Mach number \( Ma_{ff} \) is the fully-frozen Mach number based on the fully-frozen speed of sound:

\[ Ma_{ff} = \frac{v_1}{a_{ff1}}; \quad a_{ff} = \sqrt{\gamma' R_1 T} \]  

From the induced state of non-equilibrium, the mixture will tend to a new equilibrium by the exchange of momentum, heat and mass. The purpose of this chapter is to derive the characteristic times associated with the relaxation processes, and to investigate the interaction of these processes. In order to make the analysis tractable, a couple of simplifying premises will be introduced. Firstly, we shall consider the case that the droplet mass fraction is sufficiently low, so that the state of the gas-vapour mixture is not affected by the exchange of momentum, heat and mass. Then, with respect to the droplets the gas phase acts as a large reservoir of constant velocity \( v_2 \), pressure \( P_2 \), temperature \( T_2 \) and vapour pressure \( P_{v2} \). A consequence is, that the droplets will fully evaporate, since the vapour remains unsaturated. Further, the effect of the curvature of the liquid-vapour interface on the phase equilibrium will be neglected. Finally, to describe the transfer processes

---

Water droplets remain spherical if the Weber number, defined as \( We = \frac{2\pi d \rho_c}{(v-v_d)^2/\sigma} \), does not exceed the value of 5 (Gyarath, 1982). For impulsively started gas flows, some droplets start to break up at \( We = 10 \), while at \( We = 14 \) every droplet will disintegrate by bag-type break-up (Wierzb, 1990).
continuum expressions will be used, neglecting free-molecular-flow effects. Then, according to Chapter 2, the Nusselt-number correlations for the transfer of momentum, heat and mass from gas mixture to droplet are:

\[ \begin{align*}
    \text{Nu}_p &= 3 \left( 1 + 0.15 \text{Re}_d^n \right) \quad \text{with} \quad n = 0.687, \\
    \text{Nu}_h &= 2 \left( 1 + 0.30 \text{Re}_d^{1/2} \text{Pr}^{1/3} \right), \\
    \text{Nu}_m &= 2 \left( 1 + 0.30 \text{Re}_d^{1/2} \text{Sc}^{1/3} \right),
\end{align*} \]

with \( \text{Re}_d = 2r_d(v_d - v) / \nu \), \( \text{Pr} = \nu / \alpha \), \( \text{Sc} = \nu / \beta \), \( \nu = \eta / \rho_c \) and \( \alpha = \lambda / (\rho_c c_p) \).

**Exchange of momentum**

Attention is first directed towards the momentum exchange process. The droplet is decelerated from its original velocity \( v_1 \) to the velocity \( v_2 \) of the carrier gas. The momentum equation for a droplet follows by combining Eqs. (2.4.2) and (2.4.12). Neglecting any effect of mass transfer and considering a constant carrier state, the result is:

\[ \frac{Dv_d}{Dt} = -2\pi r_d^2 \eta_2 \text{Nu}_p (v_d - v_2). \]  

The time derivative in Eq. (4.2.6) refers to "particle time", and is related to the space derivative by: \( D / Dt = v_d d / dx \). Equations (4.2.6) and (4.2.3) can easily be solved. The result can be expressed as a time dependence of the Reynolds number \( \text{Re}_d = 2r_d(v_d - v_2) / \nu_2 \), starting from the initial value \( \text{Re}_d = \text{Re}_d(v_d = v_1) \):

\[ t = \frac{2 \rho c r_d^2}{9 \eta_2^2} f(\text{Re}_d; \text{Re}_d) \] ;

\[ f(\text{Re}_d; \text{Re}_d) = \ln \frac{\text{Re}_d^2}{\text{Re}_d} + \frac{1}{n} \ln \frac{1 + 0.15 \text{Re}_d^n}{1 + 0.15 \text{Re}_d^n}. \]

---

\(^3\)Here it is assumed that the parameters \( x \) and \( y \), defined by Eqs. (2.4.19) and (2.4.20), are negligibly small, which is valid when the strength of the shock wave is not too high. Moreover, the vapour content is assumed low enough to neglect the factor \( \Pi_g \) in Eq. (2.4.23).
A typical example of the change of the Reynolds number as a function of the time is shown in Fig. (4.2.1).

From Eqs. (4.2.4) and (4.2.5), it follows that the relative contribution of convection to heat and mass transfer has become less than 0.1 when $\tilde{Re}_d < 0.1$. Therefore, as a characteristic time for momentum relaxation, we take the time interval in which the Reynolds number decreases from its initial value $\tilde{Re}_{d_1}$ to the value 0.1:

$$\tau_{\text{mom}} \equiv \frac{2 \rho \xi_r r_{d_1}^2}{9 \eta_2} f(0.1; \tilde{Re}_{d_1}). \quad (4.2.9)$$

It may be noted that the time constant in front of the function $f(0.1; \tilde{Re}_{d_1})$ is equal to the characteristic time of momentum relaxation defined by Marble (1969). Here, the Reynolds-number function appears extra, because the momentum relaxation time is related to the instant of negligible convective heat and mass transfer.

**Exchange of heat and mass**

Next, we focus on the transfer of heat and mass. A characteristic time for the diffusion of heat inside a droplet of radius $r_{d_1}$ was introduced in Chapter 2 as:

$$\tau_{\text{int}} \equiv \frac{r_{d_1}^2}{3a_\xi}. \quad (4.2.10)$$

We shall anticipate the smallness of $\tau_{\text{int}}$ and take the droplet temperature $T_d$ uniform and equal to the surface temperature.

The droplet temperature changes due to the heat transferred to the interior of the droplet. The internal heat flux follows from a balance of energy at the interface of the droplet and the gas. As a result, the change of the droplet temperature is related to the heat flux $\dot{H}$ and the mass flux $\dot{M}$ from the gas to the droplet by:

$$m_d c_\xi \frac{dT_d}{dt} = \dot{H} + \dot{M}L. \quad (4.2.11)$$

Initially, the compressed vapour condenses from the heated gas phase on the still cold droplet. Therefore, $\dot{H}$ and $\dot{M}L$ have equal and positive sign, and the droplet temperature increases rapidly. After a short time, the droplet temperature rises above the saturation temperature of the vapour and the liquid starts to evaporate,
thereby absorbing heat. Finally, the droplet temperature approaches the quasi-steady wet-bulb value governed by \( H = -\frac{ML}{\dot{H}} \). The wet-bulb temperature \( T_{wb} \) is given by Eq. (2.4.48). For constant reservoir conditions, and if the evaporation is completely governed by heat conduction and vapour diffusion, the wet-bulb solution is exact.

The time history of the droplet temperature \( T_d(t) \), starting from the initial value \( T_d(0) = T_1 \), follows from Eq. (4.2.11), inserting Eqs. (2.2.12), (2.4.10), (2.4.11) and (2.4.14), and neglecting the influence of the droplet radius on the vapour pressure at the droplet:

\[
\frac{dT_d}{dt} = \frac{3}{\rho c_T r_d^2} \left[ \frac{\nu_H}{2} \left( T_2 - T_d \right) + \frac{D_{m2}}{\lambda_2} \left( \frac{\nu_M}{2} \left[ p_{v2} - p_{vS}(T_d) \right] \right) \right], \tag{4.2.12}
\]

where \( D_{m2} \) is given by Eq. (2.4.13). A good approximate solution of Eq. (4.2.12) can be obtained by direct numerical integration neglecting the variation of \( r_d \), which implies that only the thermal effect of mass exchange is taken into account. Besides, to this end, it must be realized that the Nusselt numbers, \( \nu_H \) and \( \nu_M \), depend on the Reynolds number, which is a known function of time, conform Eqs. (4.2.7) and (4.2.8). Once \( T_d \) has been calculated, first-order approximations of \( -\frac{ML}{\dot{H}} \) and \( r_d \) can be obtained, using Eqs. (2.4.10), (2.4.11) and (2.4.1).

An example of a numerical result is given in Fig. (4.2.1), where the time histories of the droplet velocity, the droplet temperature, the ratio \( -\frac{ML}{\dot{H}} \), and the droplet radius are shown. In the early phase condensation occurs, when the vapour pressure has been increased by shock compression, while the droplet temperature is still close to its initial pre-shock value. After some time, \( -\frac{ML}{\dot{H}} \) becomes positive, so that evaporation starts. As \( -\frac{ML}{\dot{H}} \) approaches unity, the internal heat flux vanishes, and the droplet temperature attains its wet-bulb value \( T_{wb} \). According to this example, the droplet radius changes only little during the stage of droplet velocity and temperature relaxation, so the assumption of invariant \( r_d \) seems to be valid here.

As a characteristic time for the approach to the wet-bulb state, we adopt the following definition:

\[
\tau_{wb} = t \left( -\frac{ML}{\dot{H}} = 0.9 \right). \tag{4.2.13}
\]

Formally, \( \tau_{wb} \) can be written as:

\[
\tau_{wb} = \frac{r_d^2 \rho c_T}{\frac{3}{\lambda_2}} g(M a_{eff}, R e_{d1}), \tag{4.2.14}
\]
Fig. 4.2. Calculated relative Reynolds number, droplet temperature, droplet radius squared, and the ratio $-ML/H$ as functions of the time for a water droplet in humid nitrogen gas. Characteristic times $\tau_{\text{mom}}$ and $\tau_{\text{wb}}$ are indicated. Conditions: $M_{\text{eff}} = 1.5$, $p_1 = 0.6$ bar, $T_1 = 270$ K, $\chi_1 = 1$, $r_{d1} = 1$ $\mu$m, $Re_{d1} = 31.9$, $T_2 = 356$ K.

where the function $g(M_{\text{eff}}, Re_{d1})$ has to follow from a numerical integration of Eq. (4.2.12) and from the definition of $\tau_{\text{wb}}$, i.e. Eq. (4.2.13).4

Once the wet-bulb state is attained, all heat transferred from the gas to the droplet is used for evaporation, and the droplet has the temperature $T_{\text{wb}}$. Moreover, after momentum relaxation, convection no longer contributes to heat and mass transfer, so that $Nu_H = Nu_M = 2$. Therefore, if the time exceeds $\tau_{\text{wb}}$ and $\tau_{\text{mom}}$, Eq. (2.5.1) can be rewritten to give an evaporation rate:

$$\frac{Dr_d^2}{Dt} = -\frac{r_d^2}{\tau_{\text{ev}}},$$

(4.2.15)

where $\tau_{\text{ev}}$ is a characteristic time for evaporation:

$$\tau_{\text{ev}} = \frac{r_{d1}^2 \rho L(T_{\text{wb}})}{2\lambda_2 (T_2 - T_{\text{wb}})}.$$  

(4.2.16)

4 Actually, the function $g$ depends also on the pre-shock conditions.
Since $\tau_{ev}$ is a constant, Eq. (4.2.15) predicts a linear decrease in the droplet radius squared with time. For a water droplet in humid nitrogen gas, the characteristic evaporation rate $r_{d1}^2/\tau_{ev}$ has been plotted in Fig. (4.2.2a) for a water droplet in humid nitrogen gas as a function of the Mach number for a given set of pre-shock conditions. As shown, the evaporation rate varies considerably over this limited range of Mach numbers.

**The characteristic times compared**

Comparing the characteristic times for internal heat diffusion $\tau_{int}$, momentum transfer $\tau_{mom}$, the arrival at the wet-bulb state $\tau_{wb}$, and evaporation $\tau_{ev}$, we obtain the following results:

\[
\frac{\tau_{int}}{\tau_{ev}} = \frac{2 c_p L(T_{wb})}{3 \rho_c^2 a_2 a_T}, \quad (4.2.17)
\]

\[
\frac{\tau_{wb}}{\tau_{ev}} = \frac{2 c_{\ell} L(T_{wb})}{3 L(T_{wb})} f(\alpha_{eff}, \kappa_{d1}), \quad (4.2.18)
\]

\[
\frac{\tau_{mom}}{\tau_{ev}} = \frac{4 c_p L(T_{wb})}{3 L(T_{wb})} f(0.1; \kappa_{d1}), \quad (4.2.19)
\]

For water droplets with initial radii of 0.5, 1 and 5 $\mu$m, nitrogen as inert gas, an initial temperature of 270 K and an initial pressure of 0.60 bar, the time constant ratios (4.2.17) to (4.2.19) are plotted in Fig. (4.2.2b) as functions of the shock strength.

First of all, we observe small values for the ratio $\tau_{int}/\tau_{ev}$, implying that droplet evaporation may be modelled with uniform droplet temperature. The increase in the ratio with increasing Mach number is due to a decrease in the evaporation time, rather than a change in the time constant of heat diffusion.

Also, the characteristic time of wet-bulb relaxation $\tau_{wb}$ is much smaller than the evaporation time $\tau_{ev}$. Even for the unfavourable case of the highest Mach number and the lowest initial droplet radius shown, we still have $\tau_{wb} < 0.05 \tau_{ev}$.

---

5 The results of Fig. (4.2.2b) have to be interpreted in conjunction with an analysis on the mechanical stability of droplets behind shock waves. A conservative value for the critical Weber number for a water droplet in impulsively started gas flow is 10 (Wierzba, 1990). This corresponds to: $(r_{d1}, \alpha_{eff}) = (0.5 \mu m, 2.08), (1 \mu m, 1.85)$ and $(5 \mu m, 1.46)$, where we have used: $\rho = 0.6$ bar, $T_1 = 270$ K and $\chi_1 = 1$. 

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Fig. 4.2.2. Characteristic times of relaxation as functions of the fully-frozen Mach number for a water droplet in humid nitrogen gas.

a. Characteristic evaporation rate $r_1^2/T_{ev}$.

b. Ratios of the characteristic time of internal heat diffusion $T_{int}$, of the formation of the wet-bulb state $T_{wb}$, and of momentum relaxation $T_{mom}$ to the characteristic time of evaporation $T_{ev}$. The arrows indicate the parametric change of the droplet radius from 0.5 μm via 1 μm to 5 μm.

Dashed curve: ratio $T_{wb}/T_{ev}$ when neglecting the effect of convective transfer of heat and mass.

Pre-shock conditions: $p_1 = 0.6$ bar, $T_1 = 270 K$, $\chi_1 = 1$.

Another observation is that the ratio $r_{wb}/r_{ev}$ depends on the initial droplet radius. The trend of a larger ratio with a smaller droplet size can simply be understood. It means that the approach of the wet-bulb state is accelerated by convective transfer of heat and mass to the droplet. For larger initial droplets the contribution of convection is more important. By way of illustration, the dashed curve in Fig. (4.2.2b) gives the time ratio $r_{wb}/r_{ev}$ in the case of purely diffusive transport of heat and mass.

The characteristic time of momentum exchange $T_{mom}$ is larger than the characteristic time of wet-bulb relaxation $T_{wb}$ for most of the Mach numbers shown. Only for low Mach numbers, $T_{mom}$ is exceeded by $T_{wb}$, but then their values are insignificantly small. Finally, we remark that typically $T_{mom} < 0.1 T_{ev}$ for $M_{aff} < 1.7$, and $T_{mom} < 0.2 T_{ev}$ for $M_{aff} < 2.0$.

We will now investigate the effect of momentum and wet-bulb relaxation on the evaporation process. To do so, we define a time $T_{max}$ as:
\( \tau_{\text{max}} = \max(\tau_{\text{mom}}, \tau_{\text{wb}}) \).

(4.2.20)

For \( t > \tau_{\text{max}} \), the droplets drift with a negligible relative velocity with respect to the gas, and the droplet temperature is close to the wet-bulb temperature. Then, the droplet radius squared decreases linearly with time. For \( t < \tau_{\text{max}} \), the evaporation rate is influenced by the additional relaxation processes. During the formation of the wet-bulb state, the temperature of the droplet is lower than the wet-bulb value. Therefore, evaporation is slowed down by this process, since the vapour pressure at the droplet is an increasing function of the droplet temperature. The process of momentum relaxation implies that the droplets and the gas have a velocity difference. Consequently, convection contributes to the heat and mass transfer, resulting in an enhancing effect on evaporation.

The combined effect of both relaxation processes on evaporation can be studied by defining an average evaporation rate \( |\phi_{r_d^{2}}|_{\max} \):

\[
|\phi_{r_d^{2}}|_{\max} = \frac{|r_d^{2}(\tau_{\text{max}}) - r_d^{2}|}{\tau_{\text{max}}},
\]

and by comparing this rate to the unaffected rate \( r_d^{2}/\tau_{\text{ev}} \). The rate \( |\phi_{r_d^{2}}|_{\max} \) can be estimated by the approximate method used earlier in this section. The ratio \( |\phi_{r_d^{2}}|_{\max} \tau_{\text{ev}}/r_d^{2} \) is plotted in Fig. (4.2.3) as a function of the Mach number for a water droplet in humid nitrogen gas. Due to the definition of \( \tau_{\text{max}} \), a discontinuity in the slope of the curves exists at the point where \( \tau_{\text{mom}} = \tau_{\text{wb}} \). Every curve shows a Mach number for which the rate ratio equals unity. In that case, the effects of momentum and wet-bulb relaxation exactly cancel one another. For higher Mach numbers, the enhancing effect of convection dominates over the decelerating effect of a lower droplet temperature. Typically, a ten percent higher rate in the first phase of relaxation is achieved when \( \mathcal{M}_{\text{ff}} = 1.95 \) for \( r_d^{1} = 1 \mu m \) and \( \mathcal{M}_{\text{ff}} = 1.5 \) for \( r_d^{1} = 5 \mu m \).

The results of Figs. (4.2.2b) and (4.2.3) can be combined to estimate the total-evaporation time \( \tau_{\text{tot}} \) using the relation:

\[
\frac{\tau_{\text{tot}}}{\tau_{\text{ev}}} - 1 = \frac{\tau_{\text{max}}}{\tau_{\text{ev}}} (|\phi_{r_d^{2}}|_{\max} \frac{\tau_{\text{ev}}}{r_d^{2}} - 1).
\]

(4.2.22)

It can be verified easily that the total-evaporation time is very well approximated by the characteristic time for evaporation \( \tau_{\text{ev}} \) for the ranges of the Mach number and the initial droplet radius considered here. For example, the most unfavourable case \( \mathcal{M}_{\text{ff}} = 2, r_d^{1} = 5 \mu m \) gives \( \tau_{\text{tot}} = 0.95 \tau_{\text{ev}} \).
Fig. 4.2.3. Ratio of the evaporation rate $|Q_{\text{d}, \text{max}}^{2}|$, defined by Eq. (4.2.21), and the uninfluenced rate $r_{\text{d1}}^{2}/r_{\text{ev}}$ as a function of the fully-frozen Mach number for a water droplet in humid nitrogen gas. Pre-shock conditions: $p_{1} = 0.6$ bar, $T_{1} = 270$ K, $\chi_{1} = 1$; $r_{\text{d1}} = 0.5$, 1 and 5 μm.

4.3 A model for the evaporation of droplets behind a shock wave

**Introduction**

In the previous section, it was shown that for moderate Mach numbers the major part of the evaporation process behind a shock wave takes place while the droplets float with the gas flow without lag and the droplet temperature is determined by a balance between heat conduction and vapour diffusion. Here we shall propose a model for shock-induced evaporation of droplets that is based on an extrapolation of this result. Moreover, droplet-size effects will be taken into account and the assumption of a negligible droplet mass fraction will be abandoned.

**The model**

The model to be discussed in this section is built on the premise that the characteristic times for the equilibration of momentum and the formation of the wet-bulb state are very much smaller than the characteristic time for evaporation of the droplets. Then, the relaxation zone behind a shock wave can be divided into
two regions. The relaxation to a momentum and wet-bulb equilibrium takes place in the first region. Since these processes are assumed to be very much faster than the evaporation process, droplet size does not change in this zone. Subsequently, the evaporation relaxation of the droplets occurs in the last subzone, while the droplets are stagnant with respect to the gas and the droplet temperature equals the quasi-steady wet-bulb value.

Since an appreciable droplet mass fraction is now permitted, the state of the gas phase may change due to the processes in the first subzone. However, this zone is considered very narrow. Therefore, we model the first relaxation zone as a discontinuity in the flow variables, that can be joined to the gas shock, thus forming one discontinuity. In modelling this new shock discontinuity, the small temperature difference between the droplets and the gas due to the wet-bulb equilibrium behind this discontinuity is not important. Then, the transition from the pre-shock state 1 to the state 2 of momentum and wet-bulb equilibrium can be described by:

\[
\begin{align*}
\frac{p_2}{p_1} &= 1 + \frac{2\hat{T}}{\gamma+1} (Ma_{pf}^2 - 1), \\
\frac{p_2}{p_1} &= \frac{P_{v2}}{P_{v1}} = \frac{P_2}{P_1}, \\
\frac{\rho_2}{\rho_1} &= \frac{v_2}{v_1} = 1 - \frac{2Ma_{pf}^2}{\gamma} \frac{1}{Ma_{pf}^2}, \\
\frac{T_2}{T_1} &= \frac{P_2 \rho_1}{P_1 \rho_2},
\end{align*}
\]

where \( \hat{T} = \tilde{c}_p / \tilde{c}_v \) is the ratio of the specific heats of the gas-vapour-droplet mixture and \( Ma_{pf} \) is the partly-frozen Mach number based on the partly-frozen speed of sound:

\[
Ma_{pf} = v_1 / a_{pf1}; \quad a_{pf} = \sqrt{\hat{T} \tilde{c}_v T}.
\]

For given pre-shock conditions and given Mach number, the structure of the evaporation zone, that follows behind the redefined discontinuity, can be calculated. The complete set of equations that is needed for this task will now be discussed. As pointed out already, during evaporation, the droplets and the gas have equal velocities, i.e. \( v_d = v \), and the droplet temperature is due to a dynamic wet-bulb equilibrium, described by Eq. (2.5.2) and formally expressed here as:

\[
T_d = T_{wb}(T, p, pv, r_d),
\]

where \( p_v / p = f_v R_v / \tilde{K} \). As indicated, the droplet temperature may depend on the size of the droplet due to the Kelvin and Knudsen effects, i.e. Eqs. (2.2.9) and (2.4.41). For an appreciable droplet mass fraction, the state of the gas-vapour mixture may
change as a result of intense momentum, heat and mass transfer between the phases. Therefore, balances of mass, momentum and energy have to be applied:

\[ \rho v = \rho_2 v_2, \]
\[ \rho v^2 + p = \rho_2 v_2^2 + p_2, \]
\[ \frac{1}{2} v^2 + h = \frac{1}{2} v_2^2 + h_2, \]

with the enthalpy and the pressure depending on the distribution of the phases:

\[ h = f_g c_p T + f_v c_p v T + f_d c_v T_d + f_v L_0, \]
\[ p = \rho \bar{v} T = \rho (f_g \bar{v}_g + f_v \bar{v}_v) T, \]

and \( f_v = 1 - f_g - f_d \). The droplet mass fraction is related to the droplet number density and to the droplet size:

\[ f_d = \frac{n_d}{3} \frac{4}{3} \pi \frac{r_d^3}{\rho} \rho_l / \rho. \]

Here \( n_d / \rho = n_d^2 / \rho_2 \), since the droplets do not break up or coalesce. Choosing a droplet size, the corresponding state of the mixture can be calculated using the set of equations above. However, the time after passage of the discontinuity when a droplet attains this size and the position inside the evaporation zone where this state occurs are still unknown. They are found by integrating the droplet-evaporation law:

\[ \frac{\text{Dr}_d^2}{\text{Dt}} = v \frac{\text{dr}_d^2}{\text{dx}} = \phi_{r_d^2}. \]

The rate of change \( \phi_{r_d^2} \) of the droplet radius squared inside a material element is given by Eq. (2.5.1). It depends on the state of the gas-vapour phase and on the droplet radius.

An example of shock-induced evaporation

The presently discussed model is used to calculate a typical case of shock-induced evaporation. An equilibrium mixture of water droplets in humid nitrogen gas is disturbed by a weak shock wave. The initial conditions of the mixture are: \( p_1 = 0.6 \text{ bar}, T_1 = 270 \text{ K}, f_{d1} = 0.005 \) and \( r_{d1} = 1 \mu m \). The strength of the shock wave is: \( M \alpha_p = 1.2 \). The change in the state of the gas-vapour phase and the change in the
temperature and size of the droplets are given in Fig. (4.3.1) as functions of the time, as they occur in a material element passing the shock wave at time zero. The time is non-dimensionalized by the characteristic time of evaporation \( \tau_{ev} \), defined by Eq. (4.2.16).

As shown, shock compression results in an increase in the pressure and the temperature, and a decrease in the saturation ratio to a value below unity. Thus, the droplets start to evaporate. The present model assumes that the evaporation takes place while a dynamic wet-bulb equilibrium exists. The time history of the droplet temperature, i.e. the wet-bulb temperature, is indicated in the figure.

The initial droplet mass fraction is sufficiently high to let phase transition affect the state of the gas phase. Heat subtraction and vapour addition lead to a decreasing temperature and an increasing saturation ratio. Therefore, the circumstances for evaporation deteriorate, as can be deduced from the approach of the droplet temperature to the gas-phase temperature. This is one reason that the rate of change of the droplet radius squared decreases during evaporation. Another factor that contributes to this result is that the transfer of heat and mass to or from a droplet becomes increasingly difficult when the droplet size decreases, which is due to a transition from the continuum-flow to the free-molecular-flow regime. Both effects result in a total-evaporation time that is approximately twice the characteristic time of evaporation. Neglecting these decelerating factors leads to a linear evaporation behaviour as represented by the dashed line. In that case, the rate of evaporation is constant and equals \( r_{d1}^2/\tau_{ev} \) as predicted by the simplified model of the preceding section. The initial slope of the curved evaporation profile is reflected by the dotted line. By comparing the dashed and the dotted line, it can be seen that the Knudsen effect is responsible for a decrease in the initial evaporation rate of about twenty percent.

Apart from affecting the transfer processes, the droplet size has another influence on the evaporation rate. For given droplet temperature, the vapour pressure at the droplet increases with an increasing curvature of the interface. This affects the wet-bulb equilibrium in such a way that the temperature difference between the droplets and the gas is enlarged. Consequently, it accelerates the evaporation process. However, this effect is only noticeable when the droplets are very small or when the conditions are close to the equilibrium conditions. For the present calculation of evaporation, the Kelvin effect is observed as a rapid decrease in the droplet temperature in the last stage of the process.

For the typical example discussed here, the initial amount of liquid mass is insufficient to force the saturation ratio to unity by the phase-transition process. Therefore, a complete evaporation of the droplets occurs. However, when increasing
Fig. 4.3.1. Evaporation of water droplets behind a shock wave in humid nitrogen gas. Plotted are the time histories of the pressure, the temperature, the droplet temperature, the saturation ratio and the droplet radius squared as observed in a material element passing the shock wave at time zero. Dotted line: initial slope of the evaporation curve, dashed line: evaporation curve neglecting droplet-size effects and the change in the gas-phase state. Conditions: $p_1 = 0.6$ bar, $T_1 = 270$ K, $\chi_1 = 1$, $f_{d1} = 0.005$, $r_{d1} = 1 \mu m$, $Ma_{pf} = 1.2$. Characteristic time: $\tau_{ev} = 3.6$ ms.

the initial droplet mass fraction or when decreasing the shock strength, the situation will be met that after shock-wave passage the gas-vapour-droplet mixture will relax to a new equilibrium with partially evaporated droplets.

**Time of total evaporation**

The preceding example showed that evaporation is slowed down by the change of the gas-vapour state due to heat subtraction and vapour addition, and by free-molecular-flow effects on the transfer of heat and mass. Their influence on the total-evaporation time will now be investigated. This will be done by comparing the total-evaporation time $\tau_{tot}$ to the characteristic time $\tau_{ev}$, defined by Eq. (4.2.16).

First, we shall consider the case that the mass fraction of water droplets in a
mixture of saturated water vapour and nitrogen gas is negligibly small. Then, for the evaporation of the droplets behind the shock wave, the gas phase can be considered a reservoir of constant conditions. Therefore, only droplet-size effects influence the reduced total-evaporation time. The ratio $\tau_{\text{tot}}/\tau_{\text{ev}}$ is depicted in Fig. (4.3.2a) as a function of the partly-frozen Mach number $\text{Ma}_{\text{pf}}$ for several initial droplet radii. For an initial droplet radius of 5 $\mu$m, the total-evaporation time $\tau_{\text{tot}}$ deviates only little from $\tau_{\text{ev}}$. Obviously, during the major part of the evaporation process the droplets are too big to experience a substantial decrease in the transfer coefficients. On the contrary, when the initial droplets have a small radius of 0.1 $\mu$m, the droplets are in the transition and free-molecular-flow regime all the time, and the evaporation occurs much slower than predicted by the characteristic time of evaporation. Finally, it may be noted that the time ratio $\tau_{\text{tot}}/\tau_{\text{ev}}$ shows the tendency to increase with decreasing Mach number. This is because at lower shock strength the post-shock gas state has a larger molecular mean free path. Therefore, the transition regime to free molecular flow starts at a higher droplet radius.

Second, we focus on the case that the droplets have an initial radius of 1 $\mu$m,
and study the influence of the initial mass fraction of liquid by varying it between zero and two percent. The results are shown in Fig. (4.3.2b). At high Mach numbers, the influence of the non-reservoir conditions is only moderate. Due to the high post-shock temperatures, the driving force for evaporation, i.e. \( \ln \chi \), varies only little during evaporation. However, with decreasing Mach number the total-evaporation time increases relative to the characteristic time of evaporation, which implies for weak shock waves a significant rise in the saturation ratio of the vapour during the continuing evaporation process. All curves, except the curve of negligible initial droplet mass fraction, show an exploding time ratio when approaching limiting Mach numbers, which are indicated in the figure. At these Mach numbers, the state of the gas-vapour mixture is affected to such an extent that a complete evaporation is no longer reached and the droplets remain to exist.
Chapter 5. Numerical method

5.1 Introduction

The development of numerical techniques capable of computing unsteady wave phenomena with internal relaxation is of great importance for apparent reasons. Theoretical models, usually based on simplifications because of the complexity of the problem, may be verified by comparison with experimental data. And once the validity of a theoretical model has been established, numerical simulation can be used as a supplement to or even a replacement of experimental work for engineering purposes or to extend the fundamental knowledge on the gas-dynamic behaviour of condensable gases.

The special character of wave propagation in gas-vapour mixtures with or without droplets lies in the induction of condensation or evaporation processes, that relax with some characteristic time that may differ greatly from the characteristic time of change associated with the wave phenomenon. Relaxation processes can have a marked influence on a wave. A shock wave running into a medium with droplets becomes partially or fully dispersed. A rarefaction wave, if of sufficient strength, initiates condensation when it runs into an unsaturated gas-vapour mixture. This then leads to the formation of several types of discontinuities embedded in the wave. Obviously, an appropriate numerical method has to treat stiff source terms resulting from relaxation effectively and correctly without using unnecessary small time steps in the computation of the propagation of the wave. Further, discontinuities embedded in continuously varying flow fields have to be represented with sufficient resolution.

According to Moretti (1987), the numerical methods for the computation of flows with discontinuities can be subdivided into two groups. One group can be described as the shock-fitting methods. With these methods, continuously varying flow fields are treated by some accurate integration of the differential equations, whereas discontinuities are handled separately by using their own algebraic jump equations. Shock-fitting methods are accurate and perform well in the case of a discontinuity as a boundary, but become cumbersome when embedded discontinuities are present or are formed. An example of this type of method is the \( \lambda \)-scheme applied by Walther and Algermissen (1985) to a chemically reacting supersonic jet flow. The other group of methods is that of the shock-capturing methods. With these methods, the whole flow field, including discontinuities, is computed using the same discretization of the equations in divergence form. There
is no need for tracking discontinuities since they are handled automatically. Some examples of shock-capturing methods are the Piecewise-Linear method, applied to dusty gas flows with shocks by Sommerfeld and Nishida (1986), the Random-Choice method (RCM), applied to rarefaction waves in dusty gases by Elperin, Igra and Ben-Dor (1988), and the Weighted-Average-Flux method and Roe's method, as discussed by Toro (1989) for the case of reactive two-phase flows.

Methods based on finite difference approximations to the derivatives of the conservation laws show a smearing of shock waves and contact discontinuities across several mesh intervals. This is due to the truncation error of the method and the artificial viscosity applied to suppress spurious oscillations. In contrast, the Random-Choice method produces shock waves and contact surfaces with infinite resolution, since the solution is found by random sampling of a sequence of Riemann problems. For the computation of the phenomena reported herein, this feature is advantageous. The gas-dynamic shock running into a fog is clearly distinguishable from its trailing relaxation zone. And further, condensation arising in the unsteady expansion wave in a gas-vapour mixture results in the formation of a compression wave in the relaxation zone. The transition of this wave into a shock wave that runs supersonically with respect to the flow is of great influence on the structure and the behaviour of the relaxing flow and gives rise to the appearance of contact surfaces. The high resolution of the RCM makes these discontinuities clearly identifiable. One disadvantage is that the RCM solution is rather noisy due to the procedure of random sampling. However, with the improvement of the sampling technique this problem is largely solved (Groth and Gottlieb, 1990).

The purpose of this chapter is to present a version of the Random-Choice method adapted for the computation of unsteady non-linear wave phenomena in gas-vapour mixtures with condensation and evaporation. The physical model that will be used to describe the gas-vapour-droplet mixture and its behaviour has been discussed in detail in previous chapters. Here, it will be summarized.

The mixture of gas and vapour is considered to be seeded with foreign particles of uniform size. A change in the state of the gas-vapour mixture may result in the formation of liquid condensate on the particles, which will be looked upon as a process of instantaneous wetting as discussed in Section 2.2. The implications of this view are that the droplet cloud will be monodisperse, and that the droplet number density will be equal to the number density of the condensation nuclei. Further, existing droplets are regarded not to break up or coalesce, and to be stagnant with respect to the gas. Moreover, droplet growth is considered to be influenced by droplet-size effects and to occur under wet-bulb conditions.

As long as the flow field remains continuous, wave propagation may be
described by the Euler equations given in Section 3.2. Regions of continuous flow are connected by flow discontinuities. Three types of discontinuities can be distinguished. The contact surface, that travels with the flow and separates fluid with different entropy, but equal pressure and velocity. Further, the shock wave, that travels supersonically with respect to the gas in front of it. The mixture, undergoing a shock transition, is compressed and heated, but essentially no phase transition occurs. Note that, if droplets are presented, they are in a wet-bulb state both in front and behind the shock wave. And finally, the condensation front, which only exists if the gas-vapour mixture in front of the discontinuity is in a supersaturated state without droplets. The state behind the discontinuity is a state of gas-vapour-droplet equilibrium.

5.2 The Random-Choice method

An outline of the method

The Random-Choice method (RCM) is a time-explicit numerical technique to solve non-linear hyperbolic partial differential equations. It was first presented in a practical form for numerical computation by Chorin (1976). Since then many changes have been introduced. An up-to-date overview of the method for one-dimensional unsteady flow is given by Toro (1987). The major advantage of the RCM is its representation of shock and contact discontinuities with infinite resolution, without any dispersion and without overshoots and undershoots. Shock waves and contact surfaces are handled automatically, so the RCM is able to deal with complex wave interactions.

The RCM uses the exact analytical solutions of a sequence of Riemann problems and a quasi-random sampling procedure to advance the solution of a given problem from one time level to the next. Some difficulties arise when the governing set of equations contains inhomogeneous terms due to relaxation processes. Firstly, an exact solution to the Riemann problem does not exist, and secondly, the relaxation processes have, in general, a characteristic time or length differing from the characteristic time or length of the wave. Therefore, a splitting technique has to be applied to deal with the inhomogeneous terms separately.

Let the solution \( U(x, t) \) be known in discrete points \( x_i = i \Delta x \) at time \( t_n = n \Delta t \), and be referred to as \( U_i^n \). And let the solution be approximated by piecewise constant functions, given for one computational cell by:

-90-
Fig. 5.2.1. Wave pattern of the shock-tube problem depicted with an \((x,t)\) diagram.

Symbols: HP: high-pressure section of the shock tube, LP: low-pressure section of the shock tube, R: rarefaction-wave fan, S: shock wave, C: contact discontinuity.

\[
U(x,t_n) = \begin{cases} 
  u^n_i & \text{if } x \leq (i+\frac{1}{2})\Delta x \\
  u^n_{i+1} & \text{if } x > (i+\frac{1}{2})\Delta x
\end{cases}, \quad (5.2.1)
\]

with a discontinuity at the centre of the cell. An initial-value problem with initial data as described by Eq. (5.2.1) is called a Riemann problem. The Riemann problem has an exact analytical solution in the absence of inhomogeneous terms in the governing equations. The solution is self-similar and consists of leftward and rightward running waves, originating at the initial discontinuity and separated by a contact surface. Depending on the initial data, the waves can either be shock waves or rarefaction waves, so that a total of four different combinations exists. In the case of the shock-tube problem, a special type of Riemann problem, the wave pattern is as depicted in Fig. (5.2.1). The complete problem can now be considered as a sequence of Riemann problems. At the next instance of time \(t_{n+1}\), their combined exact solutions form the solution to the full problem. The solution of a Riemann problem is multi-valued and quasi-random sampling is applied to choose a solution value at a random position, which then is assigned to a computational node at time \(t_{n+1}\).

It is noted that smooth parts of a flow, in principle described by the Euler equations, can also be described by a sequence of Riemann problems, where shock discontinuities are part of the solution. The reason is that solutions of the integral
conservation laws and solutions of the Euler equations are identical in the limit of vanishing discontinuities.

Our implementation of the RCM is basically similar to that described by Toro (1987). To calculate the Riemann problems, we applied an exact Riemann solver using Newton-Raphson iteration, where the iteration is initialized with an approximation proposed by Gottlieb and Groth (1988). Further, a non-staggered grid is used, with the sampling procedure illustrated in Fig. (5.2.2). Every time step only one sampling position is selected quasi-randomly according to the binary VDC sequence (Collela., 1982). Then, the solution of the Riemann problem at the sampling position is assigned to the closest computational node. On average, a correct wave propagation is assured, since the binary VDC sequence produces quasi-random numbers that lie alternately in the upper and lower half of the sampling range. The starting sequence number used is 100. The Courant-Friedrichs-Lewy (CFL) condition is based on the maximum wave speed present in the sequence of Riemann problems of the given time level.

As discussed by Groth and Gottlieb (1990), the numerical accuracy and convergence rate of the RCM depends heavily on the random-number sequence that is applied. These authors show that if the VDC sequence is used the RCM has a performance that closely approaches that of a first-order method.
Computation of wave phenomena with phase transition

The complete set of equations, describing wave propagation in continuous parts of the flow field, was given in the first subsection of Section 3.2. This set appeared to be inhomogeneous in Eq. (3.2.9), due to the possibility of phase transition. Then, application of the RCM demands a two-step procedure to advance a given solution from time \( t \) to \( t + \Delta t \). In the first step the inhomogeneous set of equations is made homogeneous by replacing Eq. (3.2.9) with \( \frac{Df_v}{Dt} = 0 \), implying that phase transition is not allowed in this step. The exact solutions of the related Riemann problems are calculated and random sampling provides a solution to the frozen problem at the new time level. This new solution is then modified to account for relaxation in the second step of the calculation. The modification consists of an integration of Eq. (3.2.9), which now is just an ordinary differential equation, over the time interval \( \Delta t \) at constant density \( \rho \) and internal energy \( e \) of the mixture:

\[
\Delta (r_d^2) = \int_t^{t+\Delta t} \phi_{r_d^2}(r_d^2) \, dt ,
\]

where \( \Delta (r_d^2) \) is the change in the droplet radius squared resulting from the correction. Knowing the change in the droplet size, the state of the mixture can be modified accordingly.

The function \( \phi_{r_d^2} \) is given by Eq. (2.5.1). Here it has a unique dependence on \( r_d^2 \) due to the constraints on \( \rho \) and \( e \). For a gas-vapour-droplet mixture of nitrogen and water, the behaviour of the relaxation function is illustrated in Figs. (5.2.3a,b). The curves shown are characterized by the state of the mixture without droplets. Then, a single curve covers all the states that can be attained by a change of state at constant density and internal energy of the mixture. A negative value of \( \phi_{r_d^2} \) corresponds to evaporation, a positive value to condensation. At a zero of \( \phi_{r_d^2} \), droplets do not grow or evaporate. A change in the droplet radius may affect the relaxation process in different ways. For sufficiently large droplets a change in \( r_d \) affects the state of the gaseous carrier. For example, in the case of condensation the carrier will be heated and vapour depletion will occur. This effect is a true relaxing effect, resulting in a stable equilibrium where the relaxation function is zero. However, for small droplets the Knudsen and Kelvin effects, described respectively by Eq. (2.4.41) and (2.2.9), act dominantly on \( \phi_{r_d^2} \). The Knudsen effect gives a decrease of the magnitude of \( \phi_{r_d^2} \), the Kelvin effect decreases the value of \( \phi_{r_d^2} \), i.e. it enhances evaporation and slows down condensation. Due to the Kelvin effect there may even exist an unstable equilibrium state at a small droplet size where the
relaxation function has a zero. For low droplet number densities, the effect of a change in the carrier state and the effects of the Kelvin and Knudsen numbers occur in distinct ranges of droplet size, as shown in Fig. (5.2.3b). Then, an intermediate range exists where the relaxation function is constant, implying a linear change in the droplet radius squared with time.

The overall time step $\Delta t$ is determined by the CFL condition for wave propagation and may differ substantially from the characteristic time with which the medium relaxes. Therefore, an appropriate and efficient numerical or analytical integration of Eq. (5.2.2) is performed after investigating the character of the relaxation function within the time interval of integration. A computational flow chart is provided in Appendix F.

Finally, we would like to point out that as a consequence of our splitting method the condensation discontinuity is not a priori included in the REM algorithm, unlike shock waves and contact discontinuities, which are. This implies that even in the case where the thickness of a condensation front is smaller than the width of a computational cell, the condensation front will not be represented as a discontinuity but will be smeared across several computational cells. However, implementation of the condensation discontinuity in the solution of a Riemann problem is not, in principle, impossible, and is similar to handling detonation waves in reacting flow, as done by Chorin (1977). This would, however, consume a considerable amount of computation time.

5.3 A special boundary condition

In this section, we will briefly explain the imposition of the boundary condition in the REM in the case of a nozzle connected to a tube. We shall assume that the flow in the nozzle is frozen with respect to phase transition. Consider the nozzle to be positioned to the right of the flow region and denote the state at the connecting cross section of the tube and the nozzle by subscript 2. At the throat of the nozzle, the flow is choked. Effectively, this results in a fixed value for the Mach number $Ma_2$ at the position just in front of the nozzle, which depends on the ratio of throat and tube diameter. By transforming this boundary condition prescribing the Mach number into a boundary condition prescribing the flow speed, the well-known piston boundary condition is obtained that is easy to implement. The flow speed is related to the Mach number by:

$$u_2 = Ma_2a_2,$$

(5.3.1)
so, for the transformation of the boundary condition, the sound speed in state 2 has to be calculated.

Let the state of the mixture at the computational node closest to the boundary be known and let it be denoted by subscript 1. In general, the Mach numbers in state 1 and 2 will not be equal, so both states have to be matched by means of a single left-facing wave. If $n_2 = \frac{n_1}{M_2}$, this wave will be a rarefaction wave, otherwise a shock wave.

In the case of a rarefaction wave, it can be shown (Thompson, 1984) that $u + 2a/\gamma - 1$ does not change across the wave. Then, using the invariance of this quantity, the sound speed $a_2$ can be obtained:

$$a_2 = \frac{M_1 + 2/(\gamma - 1)}{M_2 + 2/(\gamma - 1)} a_1. \quad (5.3.2)$$

In the case of a shock wave, the integral mass conservation equation can be written, using the shock relations (4.3.1), as:

$$a_2 = \frac{2(1 - M_2^2)}{(\gamma + 1) M_2 a_2} + \frac{M_1}{M_2}, \quad (5.3.3)$$

Fig. 5.2.3 Relaxation function $\phi_{rd}$ as a function of the droplet radius squared, when applying constraints of constant density $\rho$ and constant specific internal energy $e$. The gas-vapour-droplet mixture consists of nitrogen and water. The curves are characterized by the state of the mixture without droplets: $p = 0.85$ bar, $\rho_v = 0.0145$ kg/m$^3$, $\rho_d = 0$ kg/m$^3$.

a. $n_d = 10^{12}$ m$^{-3}$, curves 1 to 7 correspond to $T(K) = 270, 280, 288, 290, 292, 300$ and 310.

b. $T = 288$ K, curves 1 to 3 correspond to $n_d (m^{-3}) = 10^{12}, 10^8$ and 1.
where $Ma_s = (u_1 - U)/a_1$ is the shock Mach number, and $U$ is the shock speed. From this equation, the sound speed $a_2$ follows, if $Ma_s$ is known. An implicit relation to determine $Ma_s$ is yielded by combining Eq. (5.3.3) with $a_2/a_1 = \sqrt{T_2/T_1}$ and using the shock relation for the temperature jump:

$$\frac{2(1 - Ma_s^2)}{(\gamma+1) Ma_s a_2} + \frac{Ma_1}{Ma_2} = \sqrt{1 + \frac{2(\gamma-1)(\gamma Ma_s^2+1)(Ma_s^2-1)}{(\gamma+1)^2 Ma_s^2}}.$$  

(5.3.4)
6.1 Introduction

In this chapter, it is explained how the two wave phenomena of interest can be studied in one experiment. First of all, the basics of the experiment is described. Then, a detailed description is given of the experimental set-up and procedures. And finally, the employed techniques to observe the wave phenomena and to follow droplet growth and evaporation are discussed.

Fig. 6.2.1.
Wave experiment: schematic set-up and wave diagram.

6.2 An outline of the wave experiment

The wave-induced growth and evaporation of droplets in a gas-vapour mixture and the influence of phase transition on non-linear waves has been studied by means of a double-wave experiment performed in a combined expansion-shock tube. A schematic of the apparatus for creating the double-wave phenomenon and a schematic wave diagram are shown in Fig. (6.2.1). The set-up consists of three parts, separated by closed diaphragms. The high-pressure section (HPS) is filled with a driver gas and the vacuum vessel (VV) is evacuated. The test section (TS) contains a mixture of an inert gas and a vapour. Moreover, the mixture is seeded with foreign nuclei to stimulate heterogeneous condensation. The temperature of the tube and the vessel is uniform, and the high-pressure section is pressurized to
some higher pressure than the test section.

On opening the diaphragm at the vessel side, an unsteady rarefaction wave (R) runs into the test section. At the orifice (0) between the test section and the vacuum vessel the wave-induced flow is choked. In terms of boundary conditions, this means that a given contraction ratio corresponds to a fixed subsonic Mach number upstream of the orifice. And as a result, the depth of the expansion is controlled by the effective area of the orifice. The gas-vapour mixture in the test section, that is reached by the rarefaction wave, is accelerated towards the vacuum vessel and experiences an adiabatic expansion, which results in a rise in the saturation ratio of the vapour. When the critical saturation ratio is attained, condensation on the nuclei is initiated and droplets are formed. The rarefaction wave reflects at the still closed diaphragm at the side of the high-pressure section and gives a renewed expansion of the mixture and an additional growth of the droplets. After passage of the second rarefaction wave a stagnant fog has formed, which remains stationary until the next reflected wave arrives. Opening the second diaphragm at a pre-determined instant of time causes a shock wave (S) to travel into the fog. On passage of the shock wave, the gas-vapour-droplet mixture is suddenly accelerated, compressed and heated, and subsequently, the droplets evaporate when traversing a finite zone behind the shock wave. At the point of measurement (M) the shock wave and its relaxation zone may be interpreted steady, if three experimental conditions are fulfilled. Firstly, the shock wave should arrive at the observation station after the second rarefaction wave has passed and the fog has returned to an equilibrium state. Further, complete evaporation of the droplets should have taken place before the third rarefaction wave arrives. And finally, the origin of the shock wave, i.e. the position of the HPS-membrane, should be a few relaxation lengths away from the point of measurement in order for the dispersed shock wave to develop.

6.3 The experimental set-up and procedures

The experimental apparatus

A more detailed but still schematic view of the experimental set-up is given in Fig. (6.3.1). The test section (TS) and the high-pressure section (HPS) consist of Ni-coated steel tubes with an internal square cross-section of 10 x 10 cm². The length of the test section is 12.8 m. Measuring stations are located along the tube at distances
Fig. 6.3.1. Schematic of the experimental set-up.


of 5.90 m and 6.25 m from the HPS-end of the test section. Typically, at the observation points the test time for shock-induced evaporation, which is the time between the passage of the second rarefaction wave and the arrival of the third, is approx. 15 ms. Further, the orifice (O) of the test section at the side of the vacuum vessel (VV) has a square cross-section of 6.5 x 6.5 cm². The volume of the vacuum vessel is 0.4 m³. This size guarantees that the flow at the orifice remains choked during the time period of the experiment.

Polyester membranes (M) separate the test section from the other compartments. The thickness of a membrane is either 30 or 50 µm, depending on the pressure difference the membrane has to withstand. The installed membranes rest against cross-shaped supports with resistance wires mounted to them (Kanthal wire, φ 0.1 mm, ρ = 150 Ω/m) and can be opened independently by electrically heating the wires (I = 1 A, Δt heating = 20 ms). To obtain a correct wave pattern at the observation station, a time period between opening both membranes is used of 65 to 70 ms.

The test section is by-passed by a recirculation system, which is connected at three positions to the test tube by valves (V1, V2, V3) that close flush to the tube wall. A vapour supply (VS), a gas ejector (E) and rotary pumps (RP) are included in the recirculation system. The capacity of a single pump is about 50 ltr/min.
The relative humidity ($\text{RH}$) and temperature ($T$) of the recirculating mixture can be monitored with a single probe (Vaisala Humicap HMP 124B). A capacitive gauge, based on a water-absorbing polymer film, is implemented for the measurement of the relative humidity. The temperature is measured by means of a Pt-100 element.

Condensation nuclei can be produced by heating an oxidized wire of 80:20 NiCr alloy (NiCr) with a diameter of 0.5 mm. The released particles are composed of $\text{Cr}_2\text{O}_3$ and $\text{CrO}_3$ (Goldsmith et al., 1966). During heating, the temperature of the wire is approx. $1000^\circ\text{C}$ and a gas flow past the wire is maintained of about 50 ltr/min. As has been determined by electron microscopy, the particles that are emitted by the wire have a diameter of $30 \pm 10$ nm.

The filling procedure

Prior to an experiment, a careful procedure has to be followed to prepare the test tube and to obtain a well-known and uniform initial test mixture. First, polyester membranes are installed to separate the different sections and all valves (V1 to V4) are opened. Then, after the evacuation of the whole system, water vapour is supplied to the test section of the tube. Nitrogen gas is added by means of the sonic ejector mounted in the recirculation system. This causes a first circulation and mixing of the vapour and gas. When the final test gas pressure of about 1 bar is attained, valve V4 is closed and the recirculation of the mixture is continued by means of the rotary pumps. The mixture is sucked out of the test tube at valves V1 and V3, led through the device for producing the condensation nuclei, and, before re-entering the test tube at valve V2, it is thoroughly mixed by means of a turbulent flow in a 0.8 m long tube with an internal diameter of 10 mm. This process of recirculation is of 10 minutes duration. During the last 5 minutes, the oxidized NiCr wire is heated, thereby emitting the condensation nuclei. As a result of the filling procedure, a uniform mixture of gas, vapour and nuclei is obtained. Afterwards, the variations in the monitored relative humidity and temperature of the mixture due to inhomogeneities are damped completely. Separate tests, using two nuclei counters (Gardner CNC) to measure the nuclei concentration at different locations in the test tube, showed that the concentration of the nuclei is uniform within the uncertainty of the measurement, which is 10%. Simultaneous to filling the test section, the driver section is pressurized. The choice of the driver gas depends on the strength of the shock wave that has to be generated. For a shock Mach number smaller than 1.6, nitrogen gas is used, otherwise hydrogen gas.
**Measuring procedures and data recording**

Shortly before the actual experiment takes place, the temperature, the relative humidity and the total pressure of the test mixture are obtained. The initial concentration of nuclei is not measured. Then, during the experiment, the wave phenomena are observed at two locations along the test tube. At a distance of 6.25 m from the HPS membrane, the pressure (P) and light extinction (LE) at three wavelengths are recorded. In addition, the gas density (W21) and, again, the pressure (P) are measured at a position that is 0.35 m closer to the HPS membrane.

The measurement of the pressure occurs by means of carefully calibrated piezoelectric transducers (Kistler 603B). The sensitive membrane of the transducer is coated with epoxy resin to prevent erroneous reading due to temperature gradients. The thickness of the coating is approx. 0.2 mm. A laser Mach-Zehnder interferometer in phase-quadrature mode is used to measure the density of the gas mixture. An explanation of the interferometric method can be found in the next section. The measurement at three different wavelengths of the attenuation of laser beams due to the generated fog allows the determination of the modal droplet size, the variance in droplet size and the droplet number density. A detailed description of the method is given in the next section. An advantage of the present experimental method of producing a fog by expanding the initial mixture is that the vapour state at the tube walls remains unsaturated, so that no condensation occurs at the observation windows.

All data are recorded by means of a LeCroy 6810 transient recorder. From the measured quantities several other physical properties can be deduced. For that purpose, the obtained time profile of the density has to be corrected for its location of measurement that deviates from that of the pressure and the light extinction. This can be done by shifting in time the first and second rarefaction wave and the dispersed shock wave of the density signal in such a way that the fronts of the rarefaction waves and the shock discontinuity match those of the pressure signal obtained at \( x = 6.25 \) m. Then, the gas temperature can be calculated from the pressure and the matched gas density by assuming ideal-gas behaviour. In this calculation, the effect of condensation on the specific gas constant of the mixture is not taken into account, meaning that the temperature is underestimated. The relative error is approx. \(-0.5 (g_{v_0} - g_v)\). Further can be deduced: the droplet mass density, the droplet mass fraction, the vapour pressure and the saturation ratio, which is the ratio of the actual vapour pressure to the saturated vapour pressure. The shock Mach number is obtained from the pressure jump by using the shock
relations. The IZI object beam traverses the test tube perpendicular to the tube axis. Therefore, the density jump across the shock wave is not measured. This was done previously (Goossens et al., 1986) by aligning the IZI object beam at an oblique angle with respect to the shock front. It was confirmed experimentally, that the pressure and density jump are well described by the frozen Rankine-Hugoniot relations.

6.4 The measuring techniques

Introduction

In the preceding section, a brief description was presented of the measuring techniques used in this investigation. The pressure is measured by means of a piezoelectric transducer. This technique is standard and will not be discussed here. However, the interferometric technique, designed by Goossens and Van Dongen (1987), to measure gas density will be explained. It allows the accurate determination of large changes in the gas density of mixtures containing droplets. Thereupon, an extensive description of the three-wavelengths light-extinction method will be given. The well-known principle of light scattering and extinction by a small particle is used as a basis for an experimental method to characterize a polydisperse droplet cloud of which the state changes rapidly with time.

The laser Mach-Zehnder interferometer

The general principle of the laser interferometric measurement of the gas density is that a laser beam is split into two separate beams. The object beam is led through the enclosure with the gas of which the density has to be determined and is made to interfere with the reference beam at the surface of a light-sensitive detector. If the amplitude of the electric field of the reference beam is $A_R$ and the electric-field amplitude of the object beam after passing the object is $A_O$, than it can easily be derived that the interference signal $S$ at the detector is proportional to:

$$S = (A_O^2 + A_R^2) + 2A_O A_R \cos \varphi,$$

where $\varphi$ is the phase difference at the detector between the object beam and the reference beam. This phase factor has a linear dependence on the density $\rho$ of the medium:
\[ \varphi = \varphi_0 + 2\pi l \kappa \rho / \lambda. \quad (6.4.2) \]

Here, \( l \) is the geometric path length within the object, \( \lambda \) is the wavelength of the laser light and \( \kappa \) is the Gladstone-Dale constant of the medium of interest. As shown, the detector signal is a multi-valued function of the gas density, which may lead to an ambiguity in interpreting the measuring signal.

In order to avoid this difficulty, Goossens and Van Dongen (1987) proposed a procedure to operate the interferometer in phase-quadrature mode. A schematic diagram of the set-up is depicted in Fig. (6.4.1). The original laser beam is linearly polarized in the direction perpendicular to the interferometer plane. A quarter-wave plate is placed in the reference beam to transform the polarization state from linear into circular. Polarizers are positioned in front of the detectors, which select polarization components in one of the two, but different, directions, that are at an angle of 45 degrees with respect to the original polarization direction. It can be shown that the signals \( S_1 \) and \( S_2 \), measured by the photo detectors 1 and 2, are proportional to:

\[ S_1 = \frac{1}{4} (A_o^2 + A_r^2) + \frac{1}{2} A_o A_r \sin (\varphi - \frac{\pi}{4}), \]
\[ S_2 = \frac{1}{4} (A_o^2 + A_r^2) + \frac{1}{2} A_o A_r \cos (\varphi - \frac{\pi}{4}), \quad (6.4.3) \]

where, now, \( A_o \) and \( A_r \) are the amplitudes of the electric field in the points A and B of Fig. (6.4.1), respectively, and \( \varphi \) is a phase factor, that is again given by Eq. (6.4.2). The expressions \( S_1(\varphi) \) and \( S_2(\varphi) \) describe a circle in the \((S_1, S_2)\) plane. As a result, an arbitrary change in the density can be determined unambiguously by measuring the change in \( \varphi \) in a number of steps \( \Delta \varphi < \pi \). The centre and the radius of the circle depend on the electric-field amplitudes of the two laser beams. Therefore, the measurement is influenced by the presence of particulate matter in the gas, since this will result in an attenuation of the object beam by light scattering.

In implementing the method, a He-Ne laser (SpectraPhysics, Stabilite model 120, 5 mW) with a wavelength of 632.8 nm is used and photo diodes (Telefunken BPV34) are chosen as light-sensitive detectors. The measuring data are digitized and stored by a LeCroy 6810 transient recorder. The calculation, by means of a personal computer, of the change in the gas density occurs via the Lissajous diagram of the measured signals. An algorithm is used that accounts for the change in the position of the centre of the circle due to the change in the state of the scattering droplet cloud. Although the Gladstone-Dale constant depends on the composition of the gas mixture, the value for pure nitrogen gas was used in the calculations. For a mixture of nitrogen and water, this leads to an overestimation of the change in the mixture.
density with a relative amount approximately equal to $0.3 \, g_{vo}$, where $g_{vo}$ is the initial fraction of water vapour before condensation. This estimate is based on the values of the Gladstone-Dale constants for water and nitrogen as listed in the tables of Appendix A.

An introduction to the three-wavelengths light-extinction method

Obviously, in experimental studies of waves with condensation and evaporation, it is important to measure the parameters that characterize the state of the fog. Such parameters are the droplet number density and the parameters that describe the droplet size distribution. Methods of measurement using optical properties are preferable because of their non-interfering character and their ability for direct and fast recording. One of these methods is the light-extinction method, which is based on the principle that the attenuation of a plane light wave traversing a suspension of particles is a function of the wavelength.

Using the wavelength dependence of light attenuation was originally proposed by Teorell in 1931. More recently, an analysis of the light-extinction method was performed by Wittig et al. (1981). In their experimental set-up, they used two laser light sources with different wavelengths. In that case, the method only allows the determination of an average particle size, which is dominated by the larger particles. Wittig et al. confirmed the reliability of the light-extinction method by measurements of the light attenuation by suspensions of polystyrene particles of known size and with a negligible size variation.

By extending the method to three light sources with different wavelengths,
additional information on the particulate medium can be obtained. Wittig et al. (1990) illustrate that it is possible in principle to determine in this way the real part of the refractive index of scattering particles of uniform size. On the other hand, if the refractive index of the particulate matter is known, the three-wavelengths method can be applied to measure the size distribution of a poly-dispersion. Smolders, Willems, De Lange and Van Dongen (1989) chose this option to observe the wave-induced growth and evaporation of water droplets in a dense and rapidly changing cloud. The method, used in that investigation, will now be discussed. First, the principles of the method will be explained. Then, the experimental set-up will be described and analysed.

The principles of the three-wavelengths light-extinction method

Consider a cloud of spherical droplets, illuminated by a parallel monochromatic light beam. The droplets will attenuate the incident light by scattering and by absorption. Provided that multiple scattering of light may be neglected, the transmitted and incident intensities $I$ and $I_0$ are related to the thickness $l$ of the particulate medium by the Lambert-Beer law:

$$I = I_0 \exp (-\beta l) \ .$$

Here, $\beta$ is the extinction coefficient of the scattering medium. Let the droplet population have a certain size distribution $F(r_d)$ and a number density $n_d$. Then, the extinction coefficient can be written as:

$$\beta = n_d \langle r_d^2 Q \rangle \ ,$$

in which $\langle r_d^2 Q \rangle$ denotes the average extinction cross section, defined according to Eq. (2.2.13), and $Q$ denotes the extinction efficiency.

The extinction efficiency $Q$ is a function of the size parameter $\alpha = 2\pi r_d/\lambda$ and the refractive index $m$ of the droplet material. It can be calculated using the Mie theory (Bohren and Huffman, 1983). The dependence of the extinction efficiency on the size parameter is shown in Fig. (6.4.2) for $m = 1.33$. The extinction-efficiency curve has a number of regularly spaced maxima and minima, which bound a structure that oscillates approximately about the value 2. Superposed on this structure, an irregular ripple structure exists. For $\alpha < 1$, the extinction efficiency is a monotonically increasing function of the size parameter. In the Rayleigh limit, i.e.

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1 The refractive index of the medium embedding the droplets is assumed unity, i.e. the continuous medium is a gas.
Fig. 6.4.2.
The extinction efficiency as a function of the size parameter according to the Mie theory.
Dashed curves: R: Rayleigh theory, D: diffraction theory.
Refractive index \( m = 1.33 \).

\[ \alpha = \frac{2\pi r_d}{\lambda} \]

\[ \sigma = 2^{1/2} \]

\[ \alpha = 2^{1/2} \]

\[ \beta_{ij} = \frac{\beta_i}{\beta_j} = \frac{<r_d^2 \sigma_i>}{<r_d^2 \sigma_j>} \quad (6.4.6) \]

It follows from Eq. (6.4.6), that the dispersion quotient is only depending on the unknown size distribution of the droplets, on the wavelengths chosen and on the known refractive indices corresponding to the wavelengths.

\[ \frac{\sigma}{\sigma_m} = 0.1 \]

\[ \sigma = 0.1 \]

\[ \sigma = 0.1 \]

\[ \sigma = 0.1 \]

\[ \sigma = 0.1 \]
We assume that the droplet size distribution satisfies a zeroth-order lognormal distribution (ZOLD) with a modal droplet radius $r_m$ and a width parameter $\epsilon$ (Kerker, 1969):

$$f(r_d) = \frac{1}{\sqrt{2\pi} \epsilon r_m} \exp \left[ -\frac{\left( \ln \frac{r_d}{r_m} \right)^2}{\frac{\epsilon^2}{2}} \right]. \quad (6.4.7)$$

The specific form of the size-distribution function chosen is not very critical if the relative width is small (Dobbins and Jizmagian, 1966). The ZOLD has the advantage that it is defined on the domain $r_d > 0$, which agrees with the physical reality. Some examples of ZOLD functions are shown in Fig. (6.4.3). The distribution functions have an increasingly skewed shape with increasing $\epsilon$. For $\epsilon \to 0$, the ZOLD approaches the normal distribution.

As a result of the size-distribution function assumed and due to the special structure of the extinction-efficiency curve, the dispersion quotient $\beta_{ij}$ has become a function of the two distribution parameters $r_m$ and $\epsilon$. Therefore, if the attenuation of light due to a polydisperse cloud of droplets is measured at three different wavelengths, two independent dispersion quotients $\beta_{12}$ and $\beta_{32}$ can be obtained, from which $r_m$ and $\epsilon$ can be determined. Once these parameters are known, the droplet number density $n_d$ and the droplet mass density $\rho_d$ can be calculated according to:

$$n_d = \beta_1/\pi <r_d^2 q_1>, \quad (6.4.8)$$

$$\rho_d = n_d \rho \frac{4}{3} \pi r_d^3. \quad (6.4.9)$$

For a cloud of water droplets, the theoretical dependence of the dispersion quotients $\beta_{12}$ and $\beta_{32}$ on the modal droplet radius $r_m$ and the relative width $\epsilon$ is shown in Fig. (6.4.4) for the chosen wavelengths: $\lambda_1 = 632.8$ nm, $\lambda_2 = 807$ nm and $\lambda_3 = 1152$ nm. It may be noted that the special structure of the extinction-efficiency curve is directly responsible for the behaviour of the dispersion quotients. In the $(\beta_{12}, \beta_{32})$ plane, the area of possible solutions is indicated by lines of constant $r_m$ and $\epsilon$. For $0.8 \mu m \leq r_m \leq 1.8 \mu m$, these lines form an open web, so, for this range of the modal droplet radius, measured values of the dispersion quotients give full information on the droplet size distribution. For $r_m > 1.8 \mu m$ say, the method no longer provides a unique solution, since the solutions move spirally towards the point (1,1) with increasing modal radius. The reason for this is that the extinction efficiency is an oscillating function of the size parameter that damps to the value 2. At the other end, for $r_m < 0.8 \mu m$ say, the web forms a tail towards the Rayleigh
Fig. 6.4.4. Theoretical dependence of the dispersion quotients $\beta_{12}$ and $\beta_{32}$ on the modal droplet radius $r_m$ and the relative width $\varepsilon$ for a polydisperse cloud of water droplets.

a. The "web" of solutions. The dashed and the solid curves correspond to constant $r_m$ and $\varepsilon$, respectively.

b. The dispersion quotient $\beta_{12}$. Curves 1 to 5 correspond to $\varepsilon = 0.02, 0.07, 0.12, 0.17$ and 0.22.

Parameter values: $\lambda_1 = 632.8$ nm, $n_1 = 1.3318$, $\lambda_2 = 807$ nm, $n_2 = 1.3282$, $\lambda_3 = 1152$ nm, $n_3 = 1.3226$. 
limit. In this limit, the dispersion quotients no longer depend on the droplet size and the solution is stationary at the coordinates \((|\lambda_2/\lambda_1|^4, |\lambda_2/\lambda_3|^4) = (2.64, 0.24)\). Due to the low resolution of the method in the tail, the parameters of the distribution function cannot be obtained from the dispersion quotients in an unambiguous way. Therefore, a concession has to be made. Fig. (6.4.4b) shows that, if an estimate of \(\epsilon\) is available, \(r_m\) can still be obtained from \(\beta_{12}\) with little error for \(r_m < 0.8 \mu m\). For example, assuming \(\epsilon < 0.12\), results in an uncertainty for the modal droplet radius of \(\Delta r_m = 0.05 \mu m\), independent of the value of \(r_m\).

**The implementation of the three-wavelengths light-extinction method**

A schematic representation of the experimental set-up to measure light attenuation at three different wavelengths is shown in Fig. (6.4.5). In this set-up, two He-Ne lasers (SpectraPhysics, Stabilite model 120, 5 mW) with wavelengths of 632.8 nm and 1152 nm, and a diode laser (Philips N515CQL) with a wavelength of 807 nm are used as light sources. The Gaussian \(e^{-2}\)-beam diameter of the He-Ne lasers is 0.8 mm, while the beam diameter of the diode laser is reduced from originally 2.4 mm to 0.8 mm by a system of two positive lenses. Reference beams, obtained by means of beam splitters, are used to measure the variation in the laser output. This procedure is superfluous for the diode laser, which has a perfectly constant output. However, the light power of the He-Ne lasers shows a variation of
about 27% with a dominant frequency of 50 Hz.

After passage of the shock tube, the intensities of the object beams are measured with photo diodes (Telefunken BPV34, EG&G YAG-100). Otherwise as suggested by the drawing of Fig. (6.4.5), the light beams pass the shock tube at the same axial position, but are separated 7 mm in the other lateral direction. To avoid the interference of light reflected at the windows of the shock tube with the direct light, the laser beams are aligned with respect to the shock-tube windows at a somewhat oblique angle (but still perpendicular to the tube axis).

An advantage of the light-extinction method is that, for the kind of experiments that are presented, a calibration of the detection system can be performed simultaneous with the actual measurement. For that purpose, the set-up is equipped with shutters. During recording, the shutters are opened at an instant just before the formation of the droplets. Then, the recorded signals contain the 0% and 100% levels, which are necessary to calculate the extinction coefficients from the measuring signals.

If clouds are to be investigated that cause severe light extinction, care must be taken not to receive the light that is scattered by the droplets. Scattered light can be excluded by limiting the detection angle of the detector by means of a lens-pinhole system in front of the detector. Such a system consists of a positive lens with a focal length \( f \), followed by a pinhole with a diameter \( D \) positioned in the focal plane of the lens. Only that light passes the system that enters with an angle that is smaller than the detection angle:

\[
\theta_{\text{det}} = \frac{D}{2f} \quad (6.4.10)
\]

The lower limit for the detection angle is determined by the diameter of the laser beam at the pinhole, since the laser beam has to pass undisturbed. In the experimental set-up, lenses are used with a focal length of 25 cm and the pinholes have a diameter of 1 mm. As a result, the detection angle is 0.002 rad.

An estimate for the amount of scattered light detected can simply be obtained by using the theory of light scattering. As derived in Appendix G, the ratio of the detected power \( P_s \) due to single scattering and the power \( P_b \) of the attenuated light beam is:

\[
\frac{P_s}{P_b} = n_d l \pi r_d^2 \frac{i_o(a,m)}{a^2} \theta_{\text{det}}^2 \quad (6.4.11)
\]

In this equation, \( i_o(a,m) \) is the scattering function evaluated in the forward direction (Bohren and Huffman, 1983), which depends on the size parameter and
Modified scattering function $i_o(a,m)/a^2$ as a function of the size parameter $a$ for a refractive index $m$ of 1.33.

the refractive index. As illustrated in Fig. (6.4.6) for $m = 1.33$, the modified scattering function $i_o(a,m)/a^2$ is a rapidly increasing function of the size parameter. The data of this figure can be used to calculate the power ratio of Eq. (6.4.11) for all conditions. For the conditions encountered in this investigation, i.e. $\rho_d < 0.01 \text{kg/m}^3$, the increase of the measuring signal due to the detection of scattered light is less than 0.001.

It is clear that when the detected power due to scattering becomes a significant fraction of the total detected power, the parameters that characterize the fog can no longer be obtained from the measuring signals via the simple principle explained in the previous subsection without introducing a systematic error. Therefore, since $P_s/P_b$ is proportional to $n_d l \tau \rho_0^2 \approx \beta/2$, the limit for measuring dense fogs is set by the range of validity of the underlying principle, rather than by the accuracy of the measurement.

The accuracy of the measurement is important for obtaining correct results in the case of a fog that causes only little attenuation. Via the definition of the dispersion quotient and Eq. (6.4.4), it follows that the relative error in $\beta_{ij}$ can be written as:

$$\frac{\Delta \beta_{ij}}{\beta_{ij}} = \frac{\xi}{l} \left[ \frac{1}{\beta_i} + \frac{1}{\beta_j} \right], \quad (6.4.12)$$

where $\xi$ is the relative error resulting from the measurement:

$$\xi = \left| \frac{\Delta (I/I_0)}{(I/I_0)} \right|. \quad (6.4.13)$$

This error may be due to numerous factors. For instance, a drift or variation of the detector sensitivity, light that reaches the detectors, other than the direct light
from the lasers, or a displacement of the laser beams during and due to the experiment. Anyhow, fact is that $\xi$ has to be determined by an experimental test. If we assume that $\xi$ is more or less independent of the state of the fog, than Eq. (6.4.12) states that the relative error of the dispersion quotients decreases with increasing extinction. Or equivalently, the method is least accurate for optically dilute clouds. When $\Delta \beta_{12}/\beta_{12}$ and $\Delta \beta_{32}/\beta_{32}$ have been estimated, the errors in the parameters of the distribution function can be obtained graphically from Figs. (6.4.4a,b) and the errors in the droplet number density and the droplet mass density can be derived with the method described in Appendix H.

Finally, it must be noted that the results delivered by the light-extinction method are due to averaging along the paths of the light beams within the object. Consequently, if the fog is not uniform along these paths, then this will be reflected in the obtained size distribution and droplet number density.
Chapter 7. Experimental results

7.1 Introduction

In Chapter 6, the experiment was described that has been used in the investigation. Basically, an unsteady rarefaction wave is created inside a test tube filled with a gas-vapour mixture. Due to the wave, the mixture cools down and droplets are formed by condensation of the vapour. After reflection at the end wall of the tube, the rarefaction wave travels back through the gas-vapour-droplet mixture and induces a renewed condensation. Finally, a stagnant and steady fog is obtained. Then, a travelling shock wave is created in the fog. On passage of the shock wave, the mixture is suddenly compressed and heated, causing the droplets to evaporate. The complete phenomenon is observed at measurement positions that are approximately halfway the test tube.

The experimental findings of the study will be presented in this chapter. The first section deals with the expansion phase of the experiment. It describes the analysis of the experimental signals and explains the phenomena that are observed. In the second section, the results on the shock-wave induced evaporation are presented. The experimental structure of the dispersed shock wave is interpreted in terms of a steady-wave model. Moreover, measured droplet evaporation times are compared to theoretical predictions obtained with the wet-bulb model.

A comparison of the experimental observations with a numerical simulation of the wave phenomenon with the Random-Choice method will be postponed to the next chapter.

7.2 The unsteady expansion of a gas–vapour mixture

Some introductory remarks

A series of experimental runs was performed with a test mixture of water vapour and nitrogen gas, seeded with condensation nuclei. The initial conditions of the gas-vapour mixture were approximately the same. The modal values of and the variations in the pressure, temperature and saturation ratio are according to:

Part of the experimental results presented in this chapter has already been published (Smolders et al., 1991).
Fig. 7.2.1. Experimental signals for the unsteady expansion of a mixture of nitrogen gas and water vapour (experimental run 417). a. Pressure. Dashed curve: test gas was dry nitrogen. b. Extinction coefficients for the wavelengths $\lambda = 632.8$ nm, 807 nm and 1152 nm. c. and d. Signals of the Mach-Zehnder interferometer. The dashed lines indicate the centre signal values and the switch-over point, that were chosen in the analysis of the signals.

$$-0.01 < p_0 \text{ (bar)} - 1.00 < 0.05,$$
$$-1 < T_0 \text{ (K)} - 293 < 1,$$
$$-0.05 < \chi_0 - 0.82 < 0.03.$$

The depth of the expansion was controlled by the size of the orifice between the test tube and the vacuum vessel. The orifice was not changed during the investigation. The final droplet size after expansion was regulated by adjusting the temperature of the heated NiCr wire, with which the condensation particles are produced. For the set of experiments, the variation in the initial number density of the nuclei resulted in a range of final droplet radii from 1.08 $\mu$m to 1.64 $\mu$m.

The experimental results on the unsteady expansion of a gas-vapour mixture with condensation will now be discussed on the basis of a single experiment, which is representative of the other experiments.
Fig. 7.2.2. Analysis of the interferometric signals (experimental run 417).

a. Lissajous diagram. The centres, chosen in the analysis, are indicated.

b. Calculated time history of the density of the gas mixture. Dashed curve: before correction, solid curve: after correction. Initial density: $\rho_{co} = 1.15 \text{ kg/m}^3$.

The experimental signals

As mentioned in Chapter 6, the change in the gas density is observed by interferometry at an observation station at a distance of 6.9 m from the vacuum-vessel membrane, which is the point of origin of the unsteady rarefaction wave. The pressure and the attenuation of laser light at three different wavelengths are measured at a position that is 0.35 m closer to this point. For a typical experiment, the pressure signal, the interferometric signals and the measured extinction coefficients are depicted in Fig. (7.2.1). The first and the second rarefaction wave are clearly noticeable in the pressure and the interferometric signals. The adiabatic expansion of the gas-vapour mixture results in the formation and growth of droplets. This reveals itself as a rapid increase of the extinction signals at the tail of the first rarefaction wave. The sensitivity for attenuation at small droplet sizes increases with decreasing wavelength. Later in the droplet-growth process, a different response of the light-extinction signals, depending on the wavelength, is observed. The object beam of the Mach-Zehnder interferometer suffers from attenuation due to the growth of droplets, as well. This is seen as a sudden decrease in the amplitude of the oscillating signals, approx. 10 ms after arrival of the first wave. Finally, the obtained pressure signal is compared to a pressure signal originating from the unsteady expansion of pure nitrogen gas. Obviously, the effect of condensation is to generate a weak compression wave behind the rarefaction wave and to increase the level of the trailing pressure plateau.
To illustrate the procedure to calculate the density of the gas mixture, the signals of the Mach-Zehnder interferometer are plotted in a Lissajous diagram, shown in Fig. (7.2.2a). Due to the phase-quadrature mode of the interferometer, the signals form circles of which the centre and the radius depend on the electric-field amplitudes of the two laser beams. The change in the density can now be calculated unambiguously by determining the change in the phase angle, while the signals describe their circular trajectory. For that, the changing centre of the orbit must be calculated. Since, approximately, the signals move around two different circular paths only, it suffices to determine the centres of these circles and to use them in the calculation. The instant of switching over from one centre to the other coincides with the instant of droplet formation, since from that moment on the object beam of the interferometer is attenuated by light scattering. The switch-over point and the centre signal values are indicated in Figs. (7.2.1c,d).

Using the procedure described above, the interferometric signals can be transformed into a time profile of the gas density. The result is shown in Fig. (7.2.2b) indicated by the dashed curve. However, the measurement of the gas density occurs at a position along the tube, that differs from the position of the other measurements. Therefore, a correction must be applied to the calculated density profile as described in Section 6.3. The solid curve of Fig (7.2.2b) represents the corrected profile. The first rarefaction wave has been shifted forward in time, the last rarefaction wave has been shifted backward. The widths of the rarefaction waves have not been adapted. Finally, the difference between the Gladstone-Dale coefficients of nitrogen and water has not been taken into account. This leads to an overestimation of the change in the density of the gas mixture with a relative amount of $0.3 g_{v0} = 0.003$.

**The calculation of the droplet parameters**

The light-extinction signals of Fig. (7.2.1b) can be used to derive the time history of the droplet parameters. The procedure has been described in Section 6.4. The deduced dispersion quotients $\beta_{12}$ and $\beta_{32}$ are shown in the $(\beta_{12}, \beta_{32})$ plane of Fig. (7.2.3a). The time increases along the curve in the direction of larger modal droplet radii. In the final state after two expansions, the droplets have grown to a size distribution with a modal radius $r_m$ of approx. 1.55 $\mu$m. The relative width $\epsilon$ of the size distribution remains more or less the same during growth. Its value is approx. 0.07. This implies a narrow distribution.
Fig. 7.2.3. Analysis of the dispersion-quotient signals (experimental run 417).

a. Signals plotted in the \((\beta_{12}, \beta_{32})\) plane. The theoretical dependence of the dispersion quotients on the distribution parameters is indicated. The error bounds are based on Eq. (6.4.12) with \(\xi = 0.01\).

b. Method of analysis for small droplet sizes. On the right: experimental \(\beta_{12}\)-signal, on the left: theoretical dependence of \(\beta_{12}\) on \(r_m\) and \(\epsilon\).

A time history of the modal droplet radius and the relative width can be obtained by interpolating between the theoretical curves of constant \(r_m\) and \(\epsilon\). However, for modal radii smaller than 0.8 \(\mu m\) the resolution is lost, so that a different method must be used. Consider Fig. (7.2.3b). To the right, the time history of \(\beta_{12}\) is depicted for the initial stage of the growth process. To the left, the theoretical dependence of \(\beta_{12}\) on \(r_m\) and \(\epsilon\) is shown. Using the observation that \(\epsilon\) does not deviate very much from 0.07 during the last phase of droplet growth, the
measured $\beta_{12}$ values can be related to $r_m$ values. For instance, for $t = 10$ ms, we have: $r_m = 0.65 \mu m$, when using $\epsilon = 0.07$. Once $r_m$ and $\epsilon$ have been obtained, the droplet number density $n_d$ and the droplet mass density $\rho_d$ can be calculated by using Eqs. (6.4.8) and (6.4.9).

The error in the dispersion quotients $\beta_{1j}$ due to the measurement is given by Eq. (6.4.12). There, $\xi$ is the relative error in the measured transmission signals, given by Eq. (6.4.13). From experimental runs with dry nitrogen gas, an upper bound for $\xi$ has been estimated to be 0.01. The error bounds, drawn in Fig. (7.2.3a), are based on Eq. (6.4.12) using this value. Obviously, such an experimental error gives an uncertainty for the modal droplet radius of about 0.05 $\mu m$ and an uncertainty for the relative width of about 0.05. Subsequently, according to the method of Appendix H, the relative errors in the droplet number density and the droplet mass density can be estimated to be about 0.1 to 0.2.

The analysis continued

It has been explained in the preceding subsections, how the pressure, the density of the gas mixture and the droplet parameters can be obtained from the experimental signals. From these quantities, every other quantity of state can be derived. Some interesting quantities, clearly illustrating the wave-induced relaxation behaviour of the mixture, are shown in Fig. (7.2.4).

On passage of the rarefaction wave, the gas-vapour mixture is set into motion towards the vacuum vessel and is expanded adiabatically. At the measurement station, this is observed as a decrease of the pressure, the temperature and the density. The saturation ratio, which is defined as the ratio of vapour pressure and saturation vapour pressure: $\chi = p_v/p_{vs}(T) = f_v \rho_R v T/p_{vs}(T)$, starts to rise due to the temperature decrease, so the vapour is brought into a supersaturated state. Then, at some critical state, condensation of vapour is initiated on the nuclei and droplets start to grow. With the phase-transition process, latent heat is released and vapour is depleted. As a result, the temperature rises and the vapour mass fraction decreases. Both effects force the saturation ratio towards unity. However, a complete equilibrium cannot be achieved yet, since the reflected rarefaction wave induces a second increase of the saturation ratio. Again, the droplets grow until the saturation ratio has returned to unity. Then, approximately half the amount of vapour has condensed and the droplets have attained a modal radius that lies between 1.55 and 1.60 $\mu m$.

The very first instant during the first expansion, that an attenuation of the red-light laser beam is observed, is indicated in the figures by an open mark. At
Fig. 7.2.4. Experimental observations for the unsteady expansion of a mixture of nitrogen gas and water vapour. Experimental errors are indicated. The marks indicate possible onset points.

a. Pressure. b. Temperature. c. Vapour mass fraction. d. Saturation ratio. e. Modal droplet radius. f. Droplet number density and total mixture density; ●: $n_d$ and $\rho$ are matched at this point.

Experimental run 417, conditions: $p_o = 1.01$ bar, $T_o = 292.9$ K, $f_vo = 1.25 \times 10^{-2}$, $n_{do} = 0.9 \times 10^{12} \text{m}^{-3}$, $\rho_o = 1.15 \text{kg/m}^3$.

that moment, the saturation ratio is equal to 2.2. However, for condensation nuclei with a radius of 15 nm, the instantaneous-wetting model for droplet formation predicts a critical saturation ratio of 1.08. In that case, the onset point is according to the black mark indicated in the figures. It is not possible to decide on the position of the onset point or on the validity of the instantaneous-wetting model,
since there will surely be a time delay between onset and observation, because the droplets have to grow to sufficient size before they can be detected. The instantaneous-wetting model is based on the assumption that the nuclei are perfectly wettable. If this is not the case and we would take the moment of observation as the moment of onset, then this would imply a contact angle of approx. 60° as can be obtained from Fig. (2.3.4). Additional remarks on the state of condensation onset will be made in the second section of the next chapter.

Fig. (7.2.4f) contains the time histories of the total density and the droplet number density. Excluding the coalescence of the droplets as a relevant phenomenon\(^2\), the ratio \(n_d/\rho\) would be expected to be constant. However, as observed from the figure, the droplet number density decreases at a rate that is higher than the rate of decrease of the density. By means of field interferometry, we have been determining the structure and thickness of the thermal boundary layers at different instants of time. It was concluded, that after 40 to 60 ms, these boundary layers have grown to a significant size. Since the measured droplet number density is an average along a line perpendicular to the tube axis, the effect of the thermal boundary layers is that close to the walls droplets have disappeared and no longer contribute to the attenuation of the light beams. One could argue that there is an adiabatic core with a relative dimension equal to the ratio of the measured number density and the number density inside the core.

### 7.3 The evaporation of droplets behind a shock wave

**Introductory remarks**

Execution of the experimental procedure, designed to study the unsteady expansion of a gas-vapour mixture, results in the formation of a stagnant fog in the test tube. At the points of observation, the fog remains stationary for about 15 ms. Therefore, this situation can pre-eminently be used to study the behaviour of a fog after shock-wave compression. A large number of shock-wave experiments have been carried out, using the fog after expansion as initial medium. The pressure, the temperature, the saturation ratio, the droplet mass fraction and the particle size of the stationary state showed some variation between the different experimental

\(^2\) For the conditions of this investigation, i.e. \(r_m = 1 \mu m, \epsilon = 0.1\) and \(n_d = 10^{12} m^{-3}\), the half-life of the droplet population is about 2500 s, when the coalescence occurs by thermal motion, and is approximately 500 s for wave-induced coalescence, as can be deduced on the basis of Eq. (2.4.43).
runs. The modal values and bounds are:

\[-0.01 < p_1 \text{ (bar)} - 0.59 < 0.03\]
\[-4 < T_1 \text{ (K)} - 270 < 3\]
\[-0.2 < x_1 - 1.0 < 0.4\]
\[-10^{-3} < f_{d1} - 7 \times 10^{-3} < 0.5 \times 10^{-3}\]
\[-0.3 < r_{m1} \text{ (\mu m)} - 1.3 < 0.4\]

The pressure in the driver section was varied, so that shock waves of different strengths were generated. The shock Mach number was derived from the measured pressure jump. At the observation station, the partly-frozen shock Mach number ranged from 1.23 to 1.88.

**The first phase of relaxation**

As explained in Chapter 4, the relaxation process behind a shock wave occurs in different phases. Typical examples of recordings of the first phase of the relaxation process are shown in Fig. (7.3.1). The pressure and the extinction coefficient for \(\lambda = 807 \text{ nm}\) are plotted as functions of the laboratory time for partly-frozen shock Mach numbers of 1.26 and 1.80. The arrival of the shock wave is observed as a sudden increase in the pressure. First, the shock compression initiates an increase of the extinction coefficient in a short period of time. This is due to the momentum relaxation of the droplets, which causes the droplet number density to increase. Then, the rise of the extinction coefficient turns into a gradual decrease, due to the evaporation of the droplets, which takes place on a much longer time scale.

For both experiments shown, the characteristic times for momentum relaxation \(\tau_{mom}\) and evaporation \(\tau_{ev}\) are calculated with the theory of Section 4.2. The time constant \(\tau'\) for the observer at a fixed position in the laboratory is related to the time constant \(\tau\) for a droplet traversing the relaxation zone by:

\[
\tau' = (v_2/v_1) \tau = (\rho_1/\rho_2) \tau.
\]

Here, \(v\) refers to the flow velocity in the shock-fixed reference frame, subscript 1 denotes the pre-shock state and subscript 2 denotes the partly-frozen post-shock state, i.e. the state after momentum and wet-bulb relaxation. The values of \(\tau_{mom}'\) and \(\tau_{ev}'\) are indicated in Fig. (7.3.1). The predicted duration of the momentum-relaxation phase agrees surprisingly well with the experimental findings for both
Fig. 7.3.1. Experimental momentum relaxation of droplets behind a shock wave. The pressure and the extinction coefficient for $\lambda = 807$ nm are plotted as functions of the laboratory time. The characteristic times $\tau_{\text{mom}}$ and $\tau_{\text{ev}}$, and the partly-frozen density jump $\rho_2/\rho_1$ are indicated. Test mixture: nitrogen and water.

a. Experimental run 409, conditions: $Ma_{pf} = 1.26$, $Re_d = 16$, $We_{\text{max}} = 0.7$, $r_m = 1.14 \mu m$, $P_1 = 0.59$ bar, $\beta_1 = 8.8$.

b. Experimental run 430, conditions: $Ma_{pf} = 1.80$, $Re_d = 64$, $We_{\text{max}} = 9.3$, $r_m = 1.27 \mu m$, $P_1 = 0.58$ bar, $\beta_1 = 6.3$.

The evaporation of the droplets appears to take longer than predicted by the theoretical time constant. As explained in Section 4.3, this is due to the effect of the droplet size on the rate of evaporation and to the change of the conditions of the gas phase by the subtraction of heat and the addition of vapour.

By comparing the change in the extinction coefficient from its pre-shock value $\beta_1$ to its value $\beta_2$ after momentum relaxation with the change in the density of the fog from $\rho_1$ to $\rho_2$, some qualitative information can be obtained on the processes of evaporation and break-up of droplets during the phase of momentum relaxation. With Eqs. (6.4.5), (6.4.9) and (2.2.16), we can write:

$$\frac{\beta_2 \rho_1}{\beta_1 \rho_2} = \frac{\rho_1 n_{d2} <r_{d2}^2Q>_2}{\rho_2 n_{d1} <r_{d1}^2Q>_1} = \frac{f_{d2} [r_{d1}]_1}{f_{d1} [r_{d2}]_2},$$

(7.3.2)

where:

$$[r_d] = <r_{d}^3>/<r_{d}^2Q>,$$

(7.3.3)

Please note that the subscripts in this equation refer to different states and do not refer to different wavelengths as is the case in Eq. (6.4.6).
If we assume that the droplets do not break up, then \( n_{d_2}/\rho_2 = n_{d_1}/\rho_1 \), and Eq. (7.3.2) shows that a difference of magnitude between the density jump and the jump in the extinction coefficient is directly related to a change in the size of the droplets. Since \( \langle r_d^2 \rangle \) is more or less an increasing function of \( r_d \), a partial evaporation will cause \( \beta_2/\beta_1 \) to be smaller than \( \rho_2/\rho_1 \). On the other hand, if we assume that no evaporation takes place during the stage of the equilibration of momentum, then the droplet mass fractions in states 1 and 2 are the same. In that case, according to Eq. (7.3.2), comparing the ratios gives a conclusion on the break-up of droplets. Droplet break-up will result in \( \beta_2/\beta_1 \) to be larger than \( \rho_2/\rho_1 \). From Fig. (7.3.1), it can be seen, that the ratio of the extinction coefficients is in close agreement with the ratio of the densities, which is derived from the measured pressure jump. Consequently, there is no significant change of droplet size during the stage of momentum relaxation. This conclusion can be drawn for all the experiments performed. Even for the experimental conditions \(\text{Ma}_{pr} = 1.88, r_{d1} = 1.34 \mu m\), which resulted in a maximum Weber number of 13, no droplet break-up could be observed.

**The structure of the evaporation zone**

The results of a complete shock-wave experiment are depicted in Figs. (7.3.2) and (7.3.3). The conditions of the initial medium are derived from the expansion stage of the experiment. For the typical case, the fraction of liquid mass is equal to 0.007 and the droplets have a modal radius of 1.59 \(\mu m\). In addition, the vapour is saturated. During the experiment, the gasdynamic shock wave is only recorded as a jump in the pressure signal. The shock strength and the change of the other quantities across the shock wave are derived from this pressure jump. In this way, the partly-frozen shock Mach number can be calculated to be 1.42.

The time histories of the droplet size and the droplet number density can be obtained from the measured extinction signals, which are plotted in Fig. (7.3.3a). As observed, the extinction coefficients do not diminish to zero after the complete evaporation of the droplets, due to the inaccuracy of the measurement. The size of the error bounds indicated in the \((\beta_{12}, \beta_{32})\) plane of Fig. (7.3.3b) is based on this observation. Only that part of the experimental curve has been drawn in the \((\beta_{12}, \beta_{32})\) plane that can be analysed. For the experiment under consideration, this means that the evaporation process can be observed until the modal radius becomes approx. 0.7 \(\mu m\). The modal droplet radius and the droplet number density, obtained from the measured extinction signals, are shown in Figs. (7.3.3c,d).

The experimental results are compared with the theoretical model of Section
Experimental results on the shock wave in a fog, compared with the theoretical model of Section 4.2. Test mixture: nitrogen and water. Experimental errors are indicated.


Experimental run 417, conditions: \( M_{\text{pt}} = 1.42, r_{\text{m1}} = 1.59 \, \mu m, n_{d1} = 0.3 \times 10^{12} \, m^{-3}, p_1 = 0.58 \, \text{bar}, T_1 = 170.2 \, K, f_{v0} = 1.25 \times 10^{-2}. \)

4.3. The model assumes an instantaneous wet-bulb and momentum equilibrium. Moreover, it deals merely with a monodisperse droplet cloud. As illustrated by Figs. (7.3.2) and (7.3.3), the evaporation behaviour of the droplets and the change in the state of the gas phase due to the relaxation process are well predicted. On disturbance of the fog by the shock wave, the pressure and the temperature are raised abruptly and the vapour is brought into an unsaturated state. The corresponding saturation ratio is as low as 0.04. In response to the shock compression of the gas phase, the droplets change their speed and temperature, and start to evaporate. At this time scale, the relaxation of momentum appears to be an almost instantaneous process as can be concluded from the measured profile of the droplet number density. This is in agreement with the predictions of the theory of Section 4.2 and supports the model that the evaporation of the droplets takes place during a momentum and wet-bulb equilibrium. The initial droplet mass
fraction is sufficiently high to let the evaporation of the droplets affect the state of the gas-vapour mixture. The subtraction of heat gives a decrease in the temperature, while the vapour mass fraction increases due to the addition of vapour. Both processes result in an increase of the saturation ratio. Consequently, as shown in Fig. (7.3.3c), the measured time history of the droplet radius squared deviates from the linear profile proposed in Section 4.2. In addition, another factor

Fig. 7.3.3. Experimental results on the shock wave in a fog, compared with the theoretical model of Section 4.2. Test mixture: nitrogen and water. Experimental errors are indicated.

a. Extinction coefficients at three wavelengths. b. Dispersion coefficients. The error bounds are based on Eq. (6.4.12) with $\xi = 0.02$. c. Modal droplet radius squared. Dashed lines: 1: evaporation curve according to the simple theory of Section 4.2, 2: initial slope of the theoretical evaporation curve. d. Droplet number density.

Experimental run 417, conditions: $Ma_{pf} = 1.42$, $r_{m1} = 1.59 \mu m$, $n_{d1} = 0.3 \times 10^{12} \text{m}^{-3}$, $p_1 = 0.58 \text{bar}$, $T_1 = 170.2 \text{K}$, $f_{v0} = 1.25 \times 10^{-2}$. 

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Fig. 7.3.4. Experimental results on the shock-induced evaporation of droplets, compared with the theoretical model of Section 4.2. Test mixture: nitrogen and water. Dashed lines: 1: evaporation curve according to the simple theory of Section 4.2. 2: initial slope of the theoretical evaporation curve.

a. Experimental run 402, conditions: $M_{pf} = 1.26$, $r_{m1} = 1.28 \mu m$.

b. Experimental run 429, conditions: $M_{pf} = 1.82$, $r_{m1} = 1.35 \mu m$.

is of influence on the rate of evaporation. The transfer of heat and mass to and from the droplets becomes increasingly difficult when the droplet size decreases. Even for the initial droplet size, this Knudsen effect causes a 20% decrease of the evaporation rate.

**Droplet evaporation rates**

Additional experimental evaporation curves, compared with the theoretical model, are depicted in Fig. (7.3.4). Together with the evaporation curve of Fig. (7.3.3c), they are representative examples of the results obtained for the range of Mach numbers investigated. The time of total evaporation appears to vary from 0.45 ms to 5 ms. The evaporation behaviour is well predicted by the theoretical model. However, an experimental curve shows a large variation, so that quantitative results can only be obtained on an overall basis. Therefore, the initial slope of an experimental evaporation curve has been determined by means of a least-squares technique applied to the full period of observation. Based on the initial evaporation rate, a characteristic time $\tau'$ of evaporation can be defined as:

$$\tau' = \frac{r_{m1}^2}{(dr_m^2/dt)_2}, \quad (7.3.4)$$

Here, subscripts 1 and 2 pertain to the pre-shock state and the initial post-shock
Fig. 7.3.5. Evaporation rates. A comparison of the experimental data with the theory. 

(a) Reduced evaporation time in the droplet coordinate system versus the post-shock saturation ratio and temperature. 

(b) Reduced evaporation length versus the partly-frozen shock Mach number.

Test mixture: nitrogen and water. Lines: theoretical bounds based on the variation in the pre-shock conditions: temperature: $270 \pm 3 \, \text{K}$, vapour mass fraction: $(5.5 \pm 1.0) \times 10^{-3}$.

state, respectively. To obtain a more universal evaporation time $\tau$ in the droplet frame of reference, we multiply $\tau'$ with the density ratio across the shock front:

$$
\tau \equiv \tau' \left( \frac{\rho_2}{\rho_1} \right).
$$

Finally, a characteristic length $L$ of evaporation can be defined as:

$$
L \equiv v_2 \, \tau.
$$

For a great number of experimental conditions, corresponding to shock waves of different strengths, this particle evaporation time, reduced with $r_{m1}^2$, is plotted as a function of the initial driving force $\log \chi_2$ and of the initial temperature $T_2$ in Fig. (7.3.5a). The theoretical dependence is indicated with the solid line based on the average pre-shock state. The dashed lines reflect the small but finite variation in the initial pre-shock conditions. It is clear that Gyarmathy's non-linear droplet growth and evaporation model, described in Chapter 2, is of great practical value and describes the experiments accurately. In addition, the experimental data are plotted as a reduced characteristic relaxation length $L/r_{m1}^2$ as a function of the partly-frozen Mach number $\text{Ma}_{pf}$ in Fig. (7.3.5.b).
8.1 Introduction

This chapter discusses the numerical results on unsteady non-linear waves in gas-vapour mixtures with phase transition. To obtain the results, the Random-Choice method, described in Chapter 5, has been used. Three different topics have been addressed. First, the wave experiment has been simulated. The results are presented in the first section and are compared to actual experimental observations. In addition, as described in Section 8.3, the asymptotic behaviour of an unsteady rarefaction wave in a gas-vapour mixture has been calculated and the asymptotic solution is compared to the theoretical self-similar solution. Finally, the reflection of the asymptotic rarefaction wave at a wall has been studied and the results are reported in the last section.

8.2 A numerical simulation of the wave experiment

The simulated wave problem

The experimental study, reported in Chapter 6 of this thesis, on the gasdynamic behaviour of a gas-vapour mixture was carried out by means of a combined expansion-compression experiment. A schematic diagram of the wave experiment was shown in Fig. (6.2.1) of Chapter 6. By opening a diaphragm to a vacuum vessel, an unsteady rarefaction wave is created, running into a test mixture of gas and vapour, seeded with foreign particles. After reflection at the other end of the tube, the expansion wave returns and gives a renewed expansion of the mixture. Due to the double adiabatic expansion, a stagnant and stationary fog develops in the test tube. Then, the diaphragm to the driver section is opened, which causes a shock wave to propagate in the fog, evaporating the droplets present.

The numerical simulation

The wave experiment has been simulated numerically with the Random-
Choice method, explained in Chapter 5. The initial conditions were taken from experimental run 108. In Fig. (8.2.1), calculated and observed quantities are shown as functions of time at a position of 6.55 m from the vacuum-vessel diaphragm. The time origin corresponds to the instant of opening the diaphragm to the vacuum vessel. The size of the condensation nuclei was put equal to $r_n = 15 \text{ nm}$ in the calculation. This value is in accord with what was found by electron microscopy. The initial experimental value of the nuclei number density is not known. Therefore, the value $n_{00} = 2 \times 10^{12} \text{ m}^{-3}$, used numerically, was obtained by matching droplet number densities at a certain instant, indicated in Fig. (8.2.1f), after the first expansion. Experimentally, the depth of the expansion is controlled by an orifice between the test section and the vacuum vessel. This boundary condition is implemented numerically by imposing a prescribed flow Mach number at the location of the orifice. The Mach number value that was used was 0.212 and was obtained by matching the numerical calculation to the data of an experiment with dry nitrogen gas. Finally, in the simulation, the shock strength and the time origin of shock-wave formation were fitted to the experimental observation. The numerical time delay of 66 ms between the rupture of both membranes agrees within 2 ms with the actual time delay.

The results of the simulation depicted in Fig. (8.2.1) give a clear picture of the wave process. Initially, the mixture is in an unsaturated state without droplets. The saturation ratio is 0.83. At the passage of the front of the first rarefaction wave, the mixture is accelerated towards the vacuum vessel and the saturation ratio of the vapour starts to rise. Then, after only 1 ms, the critical value $X_c = 1.08$ is reached and droplets start to grow on the nuclei. However, not until the droplets have attained a radius of about 0.4 $\mu\text{m}$, corresponding to a fraction of 0.035 of vapour mass condensed, are the macroscopic effects of condensation observable. Due to condensation, vapour disappears and heat is released, resulting in a maximum for the saturation ratio and a minimum for the pressure and the temperature. Before passage of the reflected rarefaction wave, the mixture has almost completely returned to an equilibrium state where the droplet radius is about 0.8 $\mu\text{m}$. The reflected wave again disturbs the state of the mixture and droplet growth is continued to a radius of approx. 1 $\mu\text{m}$. It is seen that, before the passage of the shock wave at $t = 80.7 \text{ ms}$, the state of the mixture has more or less become stationary and the velocity of the flow has decreased to a low value of approx. 4 m/s. When the shock wave passes the observation point, the shock Mach number,

\footnote{The computation was performed on a 80386/20 MHz personal computer with a numerical co-processor. Using a mesh of 256 points, the total computation time was about four hours.}
based on the partly-frozen speed of sound, is 1.23. The state of the gaseous carrier changes discontinuously to an unsaturated state and the droplets start to evaporate. Due to the evaporation relaxation of the droplets, the pressure, the velocity and the temperature change continuously. However, the induced increase of the saturation ratio is insufficient to restore the phase equilibrium, so the droplets completely disappear. The time of total evaporation is 5.6 ms.

A comparison with the experiment

Overall, there is a notable agreement between the experimental observations and the numerical calculation shown in Fig. (8.2.1).

The droplet-formation model used in the computation is based on the assumption of instantaneous wetting of the condensation particles, as described in Chapter 2. For an adiabatic expansion, this model connects the critical saturation ratio \( \chi_C \) to the radius \( r_n \) of the nucleus. The values of \( \chi_C \) predicted by this model lie rather close to unity. For the present case, \( r_n = 15 \text{ nm} \) corresponds to \( \chi_C = 1.08 \). Although the critical state for the onset of condensation could not be determined experimentally, the calculation shows that it indeed is close to the saturation condition and that the instantaneous-wetting model gives acceptable results.

As discussed in Chapter 6, the experimental techniques to determine the gas density and the droplet number density are based on measuring the change in the optical properties of the mixture by means of laser beams. Inevitably, these methods give results that are average values along a line perpendicular to the tube axis. Therefore, the effect of the thermal boundary layers that develop behind an unsteady rarefaction wave is that the measured values of gas density and droplet number density are lower than the values that apply to the adiabatic core\(^3\). Moreover, since the boundary layers have no influence on the measurement of the pressure, the derived gas temperature overestimates the core temperature with a relative amount equal to the relative deviation of the gas density. For the state before shock arrival, the differences in the temperature and the droplet number density between the experimental and the numerical profiles correspond to a boundary-layer thickness of the order of one tenth of the tube diameter. This estimate is in agreement with observations of the thermal boundary layer that have been carried out by means of field interferometry. Obviously, as shown by Fig. (8.2.1d), the systematic errors in the temperature and the droplet number density due to the boundary layers cancel one another in the saturation ratio.

\(^3\) See Section 7.2 as well.
Fig. 8.2.1. Numerical simulation of the wave experiment, compared with experimental observations. a. Pressure. b. Velocity. c. Temperature. d. Saturation ratio. e. Droplet radius. f. Droplet number density.

Solid line: simulation, dotted line: experimental run 108. Test gas: nitrogen gas and water vapour. Initial conditions: $p_0 = 1.00$ bar, $a_o = \sqrt{(\gamma R T)_o} = 351$ m/s, $T_o = 294.3$ K, $\chi_o = 0.83$, $n_{no} = 2 \times 10^{12}$ m$^{-3}$, $r_n = 15$ nm. Critical saturation ratio: $\chi_C = 1.08$. Tube length: $\lambda = 12.8$ m. Observation point: $x = -6.55$ m.

Finally, the evaporation of the droplets behind the shock wave is well predicted by the simulation. The deviation between the evaporation curves, when the droplet radius has decreased to about 0.5 $\mu$m, is attributed to the inaccuracy of the measurement.
As already mentioned in Section 6.2, a number of conditions have to be fulfilled for a successful measurement of droplet evaporation rates by means of a shock-wave compression. A requirement is that the shock wave and its relaxation zone should be steady at the point of observation. Further, the pre-shock state should be sufficiently constant and other waves should not interfere with the shock wave during the observation period. To test these conditions, the Random-Choice method can be employed pre-eminently.

First, for the wave problem discussed above, we make a comparison of the evaporation history at the observation point as calculated with the Random-Choice method with the evaporation profile according to the steady-wave model of Section 4.3. As demonstrated by Fig. (8.2.2), the evaporation curves are in perfect agreement, which means that, indeed, at the point of observation the dispersed shock wave has developed its steady structure. The example shown concerns a shock wave with a Mach number that is the lowest within the range of experimental Mach numbers. Therefore, the example can be classified as a worst case with respect to the condition of steadiness.

Next, we want to investigate the remaining conditions. For that, an interesting moment of the wave process is when the shock wave is about to arrive at the observation window. Fig. (8.2.3) shows the situation at \( t = 80 \text{ ms} \). The shock wave is at a distance of 0.40 m from the point of observation and is moving to the right. In the relaxation zone behind the shock wave the droplets evaporate. The width of the relaxation zone is 2.15 m and may be compared to the characteristic lengths of evaporation presented in Fig. (7.3.5). Some distance behind the dispersed shock wave, the contact discontinuity between the driver gas and the test gas is observed in the profile of the saturation ratio. The observation point is approached from the
Fig. 8.2.3. *Numerical simulation of the wave experiment.* a. Pressure. b. Velocity. c. Saturation ratio. d. Droplet radius squared. Time instant: t = 80 ms.

Symbols: C: contact discontinuity, E: end of evaporation zone, S: shock wave, O: observation station, R: rarefaction-wave head. Constants: $p_1 = 0.58$ bar, $a_1 = \sqrt{\gamma \bar{\rho} T}$, $332$ m/s, $r_{d1} = 1.016 \mu$m, $\lambda = 12.8$ m. For additional specifications, consult the figure caption of Fig. (8.2.1).

right by the expansion wave. The expansion causes the saturation ratio to go up again. In between the rarefaction wave and the shock wave, a region exists with a width of 3.40 m, where, more or less, the fog is stagnant and in a state of equilibrium. For a successful experiment, it is important that the shock wave and its trailing relaxation zone have passed the observation point before the rarefaction wave arrives. Fig. (8.2.1) showed that this is the case.

8.3 The asymptotic behaviour of an unsteady rarefaction wave

*Introduction*

As illustrated in the preceding section, the unsteady expansion of a gas-vapour mixture is accompanied by a non-equilibrium condensation process. This process
can be characterized by a relaxation time, or equivalently, a relaxation length. Clearly, for the presented example the characteristic time of the wave and the relaxation time are of the same order of magnitude. During the passage of the wave, when the pressure changes substantially, the saturation ratio remains considerably different from unity. Further, a time delay is observed between the onset of condensation at the critical state and the occurrence of macroscopic relaxation effects of condensation, i.e. a decrease in the saturation ratio towards unity and the formation of a compression wave at the tail of the rarefaction wave. As argued in Section 3.4, the relaxation time changes only slightly with time and eventually attains a constant and finite value. Since the characteristic time of the rarefaction wave is proportional to time, a self-similar wave solution will develop asymptotically as $t/\tau \to \infty$, with $\tau$ as the relaxation time.

The self-similar solution, considered in Section 3.4, differs substantially from the solution at a time of order $\tau$. If the onset of condensation occurs at a saturation ratio of unity, the rarefaction wave is split into two zones separated by a uniform region. This is due to the discontinuous character of the equilibrium speed of sound at the onset point. The specific case presented in the preceding section, which has a critical saturation ratio close to unity, will develop into an asymptotic solution of this type. However, in the case where the condensation is delayed until a fixed critical supersaturation is reached that deviates considerably from unity, then a condensation discontinuity is part of the solution. In our model, the critical saturation ratio is controlled by the size of the nuclei. Therefore, in order to obtain this type of asymptotic solution, the size of the nuclei has to be reduced.

**Conditions of the problem and the way of computation**

The development of the asymptotic solution of a rarefaction wave, propagating into a mixture of water vapour and nitrogen gas, has been studied numerically. Initial conditions of the mixture are identical to those given in the preceding section. However, the size of the condensation nuclei now has been reduced to $r_n = 1.5 \text{ nm}$, implying a critical saturation ratio $\chi_c = 2.2$. This configuration will lead to length and time scales that are not easily realizable in practice. We remark that the length and time scales are proportional to the characteristic length and time of relaxation, which depend on the droplet number density and the material properties. At $t = 0$, a diaphragm is opened, separating a semi-infinite tube and a vacuum vessel. The orifice boundary condition again imposes a Mach number of 0.212. The calculation was performed on a mesh of 500 points. The initial spatial
Fig. 8.3.1. Development of the asymptotic wave solution. The pressure $p$, the temperature $T$, the saturation ratio $\chi$ and the droplet radius $r_d$ are plotted as functions of the similarity variable $x/a_0 t$. Curves 1 to 9 correspond to the instants $t (\text{ms}) = 10, 30, 70, 88, 104, 112, 122, 200$ and 1120. Dotted line: analytical frozen solution, dashed line: analytical asymptotic solution. Test gas: nitrogen gas and water vapour. Initial conditions: $p_0 = 0.997 \text{ bar}, T_0 = 294.3 \text{ K}, \chi_0 = 0.83, a_0 = \sqrt{(\gamma \mathcal{E} T)_0} = 351 \text{ m/s}$. Critical saturation ratio: $\chi_c = 2.2$.

Step size is 0.05 m. At $t = 70 \text{ ms}$, the front of the rarefaction wave is bound to run out of the computational mesh. Then, the spatial step size is doubled, reducing the number of used mesh points by a factor of 2. At time $t (\text{ms}) = 140, 280, 560$, etc. this procedure is repeated. Every cycle took about 2 to 3 hours of computation time.
The results of the computation

The results of the computation are presented in Fig. (8.3.1). The pressure, the temperature, the saturation ratio and the droplet radius are shown as functions of the similarity variable \(x/a_0 t\), where \(a_0\) is the initial sound speed of the mixture. This has the advantage that the development of the wave can be presented easily. On the other hand, it is rather difficult to interpret changes in the wave structure that do not show similarity behaviour. The different curves represent the solution at subsequent instants of time. Both the analytical frozen \((t = 0)\) and the analytical asymptotic \((t/\tau \to \infty)\) solution are depicted. At the first depicted instant, the characteristic extent of the wave is small with respect to the relaxation length. Then, the rarefaction wave itself is described by the fully-frozen solution and the macroscopic effect of condensation is observed beyond the tail of the rarefaction wave. The distance between the point of the critical saturation ratio and the point of the observed condensation is related to the relaxation length. With increasing time, the extent of the rarefaction wave increases. Since the relaxation length is almost invariant, this results in a relative shift of the compression wave towards the head of the rarefaction wave. Consequently, the maximum value of the saturation ratio decreases. At the fourth instant, the front of the compression wave has steepened into a shock wave and the maximum of the saturation ratio has decreased to the critical value. A moment later (instant 5), it is even lower and the initiation of condensation has stopped. A contact discontinuity has formed, observable as a jump in the temperature but not in the pressure, which travels to the right with the speed of the flow. To the right of the contact discontinuity, a material element exists with growing droplets. To the left, no droplets have formed, since this material element has not yet encountered the critical state. The value of the second maximum of the saturation ratio, positioned at the contact discontinuity, increases with time. At time instant 6, it has attained the critical value and the onset of condensation is restarted at the back of the compression wave. Then, the strength of the contact discontinuity diminishes. It has almost completely vanished at instant 7. The process of stopping the onset of condensation and restarting it at a different position in the wave repeats itself, thus forming a critical-state plateau. The condensation process is initiated at the back of the plateau and the droplets grow in a finite time until a new equilibrium is established. Because the rarefaction wave broadens in proportion to the time, the finite and constant relaxation zone can eventually be considered as a condensation discontinuity. The final depicted time shows a solution that closely resembles the analytical asymptotic solution, derived in Section 4.3. The shock wave, as a
Reflection of an asymptotic rarefaction wave. The pressure $p$ and the vapour mass fraction $f_v$ are plotted as functions of position and time. Test mixture: nitrogen and water. Initial conditions: $p_0 = 0.997$ bar, $f_{v0} = 0.0136$, $a_0 = \sqrt{(\gamma R T)_0} = 351$ m/s. Tube length: $L = 400$ m. Marked positions: a: head of the incoming wave, b: condensation discontinuity, c: condensation due to the reflection.

The remainder of the start-up behaviour, has almost completely disappeared. A critical-state plateau has formed, bordered at the rear by a condensation discontinuity that travels at the correct subsonic speed.
8.4 The reflection of an asymptotic rarefaction wave

Finally, we consider the perpendicular reflection of an unsteady rarefaction wave in its self-similar form at a wall. The calculation described in the preceding section was restarted at time $t = 750$ ms. At that time, the spatial step size is 0.8 m. Now, we assume that the tube, which contains the mixture in which the rarefaction wave propagates, has an end wall at $x = -400$ m. The result of the calculation is shown in Fig. (8.4.1). At the initial time of the calculation, the rarefaction wave has the asymptotic wave form. The head of the wave is at the position marked $a$, as can be seen from the pressure diagram. The condensation front is at the end of the plateau at the position marked $b$. Up to point $b$, the mixture is free from droplets, as can be obtained from the vapour mass fraction diagram. At the condensation front, the critical state is established and condensation takes place on the nuclei, giving an almost discontinuous decrease in the vapour mass fraction to a value corresponding to the liquid-vapour equilibrium at post-wave conditions.

The rarefaction wave travels to the left and encounters the wall, resulting in a supersaturation of the vapour. At the position marked $c$, the supersaturation becomes critical and condensation relaxation is triggered almost instantaneously on the whole interval from the wall to the front of the plateau, where a second condensation front is formed that travels to the right. The plateau between the two condensation fronts is now the only part of the wave where the mixture is still without droplets. At the instant of time that the two condensation fronts encounter one another, the essential condition for the existence of a condensation discontinuity disappears, i.e. the supersaturated state in front of the discontinuity disappears, and both discontinuities behave as ordinary diverging rarefaction waves in an equilibrium gas-vapour-droplet mixture.
The unsteady rarefaction wave

An unsteady rarefaction wave propagating in a gas-vapour mixture induces the formation and growth of droplets in the mixture. The addition of foreign particles to the mixture stimulates the onset of the condensation process.

The experimental study does not yield accurate data on the critical state for the onset of condensation, since during that stage of the process the droplets are still too small to be detected by light-attenuation measurement. However, a numerical simulation of the unsteady expansion with the Random-Choice method shows that a model based on the instantaneous wetting of the condensation nuclei gives acceptable results.

For the conditions of the experimental investigation, the wave-induced condensation process appears to be a non-equilibrium relaxation process. The rapid adiabatic expansion results in an increase in the saturation ratio of the vapour to values considerably above unity. This supersaturated state is then broken down by the effects of condensation on the state of the gas-vapour mixture, which are the addition of latent heat and the depletion of vapour. A comparison of the experimental findings with calculations performed with the Random-Choice method gives good agreement. This supports the non-linear droplet-growth model of Gyarmathy, which is based on a dynamic wet-bulb equilibrium and which incorporates the Knudsen and Kelvin effects.

Due to a substantial supply of latent heat to the flow, the rarefaction wave is affected by the condensation process. One can conclude that an important parameter in the description of the behaviour of the wave is the characteristic time $\tau$ of relaxation to the equilibrium state. When time is of the order of $\tau$, which corresponds to the experimental circumstances, the non-equilibrium condensation zone is located at the tail of the rarefaction wave. There, the effect of condensation is to generate a weak compression wave behind the rarefaction wave and to increase the level of the trailing pressure plateau. Since $\tau$ changes only slightly with time, a self-similar wave solution develops asymptotically as $t/\tau \rightarrow \infty$. If the onset of condensation occurs at a saturation ratio of unity, the rarefaction wave is split into two zones separated by a uniform region. This is due to the discontinuous character of the equilibrium speed of sound at the onset point. However, in the case that condensation is delayed until a fixed critical supersaturation is reached that deviates considerably from unity, then a condensation discontinuity of the
expansion type is part of the solution. A numerical simulation of the development of the asymptotic wave with the Random-Choice method indicates that time has to proceed over more than two decades of characteristic times of condensation before the self-similar solution can be recognized. Since a value for $r$ corresponding to the experimental situation is of the order of 10 ms, the asymptotic wave form has not been realized in the laboratory.

The shock wave

A shock wave, moving in a gas-vapour-droplet mixture, induces a state of non-equilibrium by suddenly changing the velocity and state of the gas-vapour mixture, while the droplets are left undisturbed, retaining their original velocity, temperature and size. Subsequently, in a finite region behind the shock wave, the mixture tends to a new equilibrium by the exchange of momentum, heat and mass between the droplets and the gas.

The analysis of the transport processes in the relaxation region behind the shock wave results in expressions for the characteristic times for droplet heat diffusion, momentum relaxation, the formation of the wet-bulb state and for droplet evaporation. A numerical evaluation of the characteristic times for the system of water droplets in humid nitrogen gas shows that, for weak shock waves and relatively small droplet sizes, the relaxation of momentum and droplet temperature takes place at a time scale that is short compared to the total-evaporation time of the droplets. Therefore, shock-induced evaporation can be described as a process governed by heat conduction and vapour diffusion while the droplet temperature remains at its wet-bulb value and the droplets are stagnant with respect to the gas.

The stagnant and stationary fog that forms in the test tube after unsteady expansion is a suitable medium for studying the response of a fog to shock compression. The observations show that the relaxation of momentum is an almost instantaneous process at the time scale of droplet evaporation. Moreover, it is found that the evaporation process cannot satisfactorily be described by a single value for the rate of change of the droplet radius squared. The full steady-wave model, based on the steady conservation laws and the droplet-growth model of Gyarmathy, gives an accurate description of the structure of the relaxation zone. The experimental results on evaporation rates for water droplets in humid nitrogen gas, obtained for an appreciable range of experimental conditions by varying the shock strength, support Gyarmathy's non-linear droplet-growth model.
The experimental method

The expansion-recompression experiment, combined with the application of measuring techniques to observe the pressure, the gas density and the droplet parameters, gives a detailed picture of non-linear waves in a medium with phase transition.

The fog, as a medium for shock-wave propagation, is formed by a double unsteady expansion of a gas-vapour mixture. It is a prerequisite, that the dispersed shock wave is fully developed at the observation station and that the test time for evaporation is sufficiently long. These terms are successfully met by taking a test tube of approx. 13 m and locating the observation station halfway the tube. Due to this configuration, however, the experimental determination of the droplet number density is significantly affected by the development of thermal boundary layers behind the unsteady rarefaction wave. The development of the boundary layers appears to be of little influence on the overall results of the investigation.

Light-extinction measurement proves to be a simple and reliable method to obtain a time-resolved profile of the droplet parameters. For spherical droplets with known refractive index, the use of three different wavelengths in the visible and infrared domain allows a determination of the number density and the modal radius of micron-sized droplets and results in an estimate for the width of the droplet-size distribution, provided the size variation is within certain limits. However, one must be aware that the results obtained are averages along the line of measurement.

The numerical method

The Random-Choice method can be applied successfully to the problem of non-linear wave propagation in gas-vapour mixtures, in which non-equilibrium condensation and evaporation phenomena occur. By a stepwise decoupling of the computation of wave propagation and phase transition, the numerical method presented proves to be effective and efficient for a wide range of ratios of the relaxation time and the characteristic wave time. The whole range in between frozen and equilibrium flow can be handled. Shock waves and contact discontinuities are described with high resolution, which is an inherent feature of the Random-Choice method. When studying the asymptotic behaviour of a rarefaction wave with condensation, the method yields a good description of the development of the condensation discontinuity, while such a discontinuity was not implemented in the numerical method as such.
Appendix A. Material-properties data


<table>
<thead>
<tr>
<th>$N_2$</th>
<th>$a_{ng} = a_{fg} = 1$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$M_g$</td>
<td>0.028013 kg mol$^{-1}$</td>
</tr>
<tr>
<td>$R_g$</td>
<td>296.8 J kg$^{-1}$K$^{-1}$</td>
</tr>
<tr>
<td>$c_{pg}$</td>
<td>1041 J kg$^{-1}$K$^{-1}$</td>
</tr>
<tr>
<td>$\gamma_g$</td>
<td>1.40</td>
</tr>
<tr>
<td>$\eta$</td>
<td>$17.6 \times 10^{-6} (T(K)/295)^{0.767}\text{kg m}^{-1}\text{s}^{-1}$</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>$25.5 \times 10^{-3} (T(K)/295)^{0.828}\text{J m}^{-1}\text{s}^{-1}\text{K}^{-1}$</td>
</tr>
<tr>
<td>$a$</td>
<td>$21.4 \times 10^{-5} (T(K)/295)^{1.858}/p(\text{bar})\text{m}^{2}\text{s}^{-1}$</td>
</tr>
<tr>
<td>$Pr$</td>
<td>$0.718 (T(K)/295)^{-0.071}$</td>
</tr>
<tr>
<td>$\kappa$</td>
<td>$2.395 \times 10^{-4}\text{m}^{3}\text{kg}^{-1}$</td>
</tr>
</tbody>
</table>

Table A1. Material-properties data of nitrogen gas. The calculated thermal diffusivity, $a$, is based on the assumption of a perfect gas.

<table>
<thead>
<tr>
<th>$N_2O - N_2$</th>
<th>$D = 24.5 \times 10^{-6} (T(K)/295)^{2.085}/p(\text{bar})\text{m}^{2}\text{s}^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$D_m$</td>
<td>$18.0 \times 10^{-8} (T(K)/295)^{1.085}\text{kg m}^{-1}\text{s}^{-1}$</td>
</tr>
<tr>
<td>Sc</td>
<td>$0.630 (T(K)/295)^{-0.318}$</td>
</tr>
</tbody>
</table>

Table A2. Material-properties data of a mixture of water vapour and nitrogen gas. The calculated Schmidt number, Sc, is based on the assumption of dry and perfect nitrogen gas.
Table A3. Material-properties data of water: vapour and liquid.
Appendix B. Heat and mass transfer rates

Heat transfer

The energy equation, presented as Eq. (2.4.15), describes the temperature field in the carrier for a steady situation. The temperature field is influenced by a radial vapour mass flow, with $\mathbf{w} = -4\pi r^2 \rho v$. Assuming that $\lambda$ may be evaluated at a given reference temperature, a constant parameter $\zeta$ can be defined as: $\zeta = (\mathbf{M}_{c_p v})/(4\pi \lambda r_d^2)$, and the differential equation, i.e. Eq. (2.4.15), can be rewritten to obtain:

$$-\zeta r_d \frac{dT}{dr} = \frac{d}{dr} (r^2 \frac{dT}{dr}). \quad (B.1)$$

The boundary conditions that must be applied are $T(r=r_d) = T_r$ and $T(r=\infty) = T_\infty$. Then, integration of Eq. (B.1) yields the temperature profile:

$$T - T_\infty = (T_r - T_\infty) \frac{1 - \exp(\zeta r_d/r)}{1 - \exp(\zeta)}. \quad (B.2)$$

The rate of supply of heat to the droplet is given by:

$$H = 4\pi r_d^2 \lambda \frac{dT}{dr} \big|_{r=r_d}, \quad (B.3)$$

so combining the two last equations results in:

$$H = 4\pi r_d \lambda \frac{\zeta}{\exp(\zeta) - 1} (T_\infty - T_r). \quad (B.4)$$

If the wet-bulb approximation, discussed in Section 2.4 and reading $\mathbf{w} = -H/L$, is used, then this equation can be rewritten as:

$$H = 4\pi r_d \lambda \frac{\ln(1 + x)}{x} (T_\infty - T_r), \quad (B.5)$$

where $x$ is the superheat parameter defined by Eq. (2.4.19). Using the last equation for the heat transfer rate and Eq. (2.4.10) defining the Nusselt number, Eq. (2.4.17) is finally obtained.
Mass transfer

The equation that describes the vapour concentration profile in the carrier, i.e. Eq. (2.4.16), needs some substitutions:

\[ v = g_v v + g_g v = g_v v, \]

\[ M = -4 \pi r_d^2 \rho_v v, \] (B.6)

\[ p = (\rho_v + \rho_g) (g_v R_v + (1-\rho_v) R_g) T, \]

in order to be transformed into:

\[ M = 4 \pi r^2 \frac{Dp}{T} \frac{1}{1-g_v (1-\rho_v) R_g + g_v R_v} \frac{dg_v}{dr}. \] (B.7)

The supplemented boundary conditions are \( g_v (r=r_d) = g_v r \) and \( g_v (r=\infty) = g_v \infty \). If we assume that \( Dp/T \) is a constant that may be evaluated at some reference temperature \( T_m \), then integration yields:

\[ M = 4 \pi r_d \frac{Dp}{R_v T_m} \ln(1+\gamma), \] (B.8)

where the parameter \( \gamma \) has been defined by Eq. (2.4.20), and use has been made of:

\[ g_v = \frac{p_v R_g}{p_v R_g + p_g R_v}. \] (B.9)

By combining Eq. (B.8) with the definition of the Nusselt number, i.e. Eq. (2.4.11), Eq. (2.4.18) finally follows.
Appendix C. The unsteady centred rarefaction wave

Consider a left-facing unsteady rarefaction wave in one dimension. The wave is centred with its origin at \( x = 0, \ t = 0 \). The medium consists of a perfect gas with only very few dispersed droplets, so the droplets do not influence the behaviour of the gas. In principle, the droplets have velocities that differ from the velocity of the gas. We make however the a-priori assumption that velocity differences are small, so that the material derivatives of the gas and droplets are approximately identical:

\[
\frac{\partial}{\partial t} + u \frac{\partial}{\partial x} \approx \frac{\partial}{\partial t} + u. \quad (C.1)
\]

As will follow from the conclusions of this section, this simplification is correct.

According to gasdynamic theory (Thompson, 1984), the rarefaction wave is described by:

\[
x = (u - a) t, \quad (C.2)
\]

\[
\frac{2}{\gamma - 1} a = \frac{2}{\gamma - 1} a_0, \quad (C.3)
\]

\[
\frac{dp}{p} = \frac{\gamma}{\gamma - 1} \frac{dT}{T} = \frac{2 \gamma}{\gamma - 1} \frac{da}{a}. \quad (C.4)
\]

The initial state of the medium is referred to by index \( \sigma \). A straightforward manipulation of these equations results in a number of useful relations:

\[
u = \frac{2}{\gamma + 1} \left( x + a_0 \right), \quad a = \frac{2}{\gamma + 1} \left( a_0 - \frac{\gamma - 1 x}{2 t} \right), \quad (C.5)
\]

\[
\frac{du}{dt} = \frac{2}{\gamma + 1} \frac{dx}{dt} \frac{x}{t}, \quad \frac{da}{dt} = \frac{\gamma - 1}{2} \frac{du}{dt}. \quad (C.6)
\]

In order to obtain the material derivatives, we put \( \frac{dx}{dt} = u \), so that:

\[
\frac{Du}{Dt} = \frac{2 a}{\gamma + 1}, \quad \frac{Da}{Dt} = \frac{\gamma - 1 a}{\gamma + 1}, \quad \frac{DT}{Dt} = \frac{2 (\gamma - 1) T}{\gamma + 1}, \quad \frac{Dp}{Dt} = \frac{2 \gamma p}{\gamma + 1} \cdot (C.7)
\]

The solution of a centred rarefaction wave is self-similar. This means that
every quantity is a function of $x/t$ only. Examples of this behaviour are reflected by Eqs. (C.5) for the velocity and the sound speed. We can express this property as:

$$\left[ \frac{dx}{dt} \right]_z = \frac{x}{t}, \quad (C.8)$$

where $z$ denotes an arbitrary quantity. Some implications for the derivatives are:

$$\frac{\partial z}{\partial x} = -t \frac{\partial z}{\partial t} \quad \text{and} \quad \frac{\partial z}{\partial x} = -\frac{1}{a} \frac{\partial z}{\partial t}. \quad (C.9)$$

Consequently, the gradient of any quantity can immediately be deduced from the material derivative.

Characteristic times of environmental change can be defined as:

$$\frac{1}{\tau_u} = \frac{1}{u} \frac{1}{Dt}, \quad \frac{1}{\tau_a} = \frac{1}{a} \frac{1}{Dt}, \quad \frac{1}{\tau_T} = \frac{1}{T} \frac{1}{Dt}, \quad \frac{1}{\tau_p} = \frac{1}{p} \frac{1}{Dt}. \quad (C.10)$$

A combination of these definitions with Eqs. (C.7) gives the following relations:

$$\frac{1}{\gamma-1} \frac{a}{u} \tau_u = -\frac{1}{2} \frac{a}{a} \tau_a = \frac{\gamma}{\gamma-1} \tau_T = \frac{1}{\gamma-1} \tau_p = \frac{1}{2} \frac{(\gamma+1)}{\gamma-1} t. \quad (C.11)$$

The characteristic times, except $\tau_u$, are functions of the time only and are independent of the position inside the wave.

A droplet travels with respect to the gas with a velocity that approximates the drift velocity given by Eq. (2.4.43). Then, the Reynolds number $Re_d$ and the Mach number $Ma_d$ of the droplet can be expressed as:

$$Re_d = \frac{2 \rho a_r_d \tau_{drf}}{\eta (\gamma-1) \tau_T} \approx \frac{1}{2} \frac{[r_d(\mu_m)]^3}{t(\text{ms})}, \quad (C.12)$$

$$Ma_d = \frac{1}{\gamma-1} \frac{\tau_{drf}}{\gamma \tau_T} \approx \frac{1}{100} \frac{[r_d(\mu_m)]^2}{t(\text{ms})}. \quad (C.13)$$

where the time constant $\tau_{drf}$ for attaining the drift velocity is given by Eq. (2.4.44). The estimates are based on $T = 295$ K, $\rho = 1$ kg/m$^3$ and $N_{uf} = 3$, and on numerical data for water and nitrogen from Appendix A.
Appendix D. Explicit droplet-growth models

Consider a droplet of known size in an environment with known pressure, temperature and saturation ratio. The growth of the droplet, expressed by the mass transfer rate $M$, can be obtained for wet-bulb conditions from the set of equations:

$$ M = 2\pi r_d \frac{\lambda N_u T_\infty T_d}{L} (\frac{T_d}{T_\infty} - 1), \quad (D.1) $$

$$ \frac{T_d}{T_\infty} - 1 = \frac{\theta}{T_\infty} p_{v\infty} - p_{v_T}, \quad (D.2) $$

where $p_{v_T} = p_{v_e}(T_d, r_d)$ and $\theta$ is given by Eq. (2.4.52). We assume that the droplet is stagnant, so the Nusselt numbers for heat and mass transfer depend only on the Knudsen number and are given by Eqs. (2.5.4). The set of equations above contains the implicit wet-bulb equation, so finding the growth rate is rather laborious. However, by introducing some simplifications this implicit equation can be replaced by an equation that explicitly relates the droplet temperature to the droplet size and the environmental conditions. Once the droplet temperature is known, the growth rate of the droplet can easily be calculated with Eq. (D.1). In the literature, different simplifying assumptions are proposed to obtain different explicit relations for calculating the growth rate of a droplet. In this appendix we shall consider the explicit growth equations according to Mason (1953), Gyarmathy (1963) and Barrett and Clement (1988).

All authors use the common assumption that the latent heat of evaporation and the transport coefficients are constant within the range $(T_d, T_\infty)$ and may be calculated at the temperature of the environment. In order to include the Kelvin effect into the consideration, the Kelvin number has to be evaluated at the temperature $T_\infty$ rather than the temperature $T_d$. To indicate this we shall add the subscript $\infty$ to the Kelvin number. Then, the vapour pressure at the droplet surface is approximated by:

$$ p_{v_T} = p_{v_S}(T_d) \exp Ke_\infty. \quad (D.3) $$

Using the assumption of constant latent heat, the Clausius-Clapeyron equation (2.2.7) can be integrated to obtain:

$$ \ln\left(\frac{p_{v_S}(T_d)}{p_{v_S}(T_\infty)}\right) = \frac{L_\infty}{R_v T_\infty} \frac{T_d}{T_\infty} \left(\frac{T_d}{T_\infty} - 1\right). \quad (D.4) $$
The basic assumption underlying the equations of Mason (1953) and Barrett and Clement (1988) is identical, whereas the basic assumption made by Gyarmathy (1963) deviates. Therefore, we shall first derive simultaneously the explicit equations according to Mason (1953) and Barrett and Clement (1988) and then return to the derivation of Gyarmathy (1963).

First, Eq. (D.2) is rewritten using Eqs. (2.2.11), (2.2.20) and (D.3) to obtain:

\[
\frac{T_\infty}{\theta_\infty} \left( \frac{p_\infty}{p_{VS}(T_\infty)} - x_\infty \right) e^{-K_\infty} \frac{T_d}{T_\infty} = \left( x_\infty e^{-K_\infty} - 1 \right) - \frac{p_{VS}(T_d)}{p_{VS}(T_\infty)}. \tag{D.5}
\]

The parameter \( \theta \) has received a subscript \( \omega \), since it has to be evaluated at the temperature \( T_\infty \). The logarithmic term of Eq. (D.4) can now be related to the last term of Eq. (D.5) by:

\[
\ln \left( \frac{p_{VS}(T_d)}{p_{VS}(T_\infty)} \right) = \zeta_1 \ln \left( \frac{p_{VS}(T_d)}{p_{VS}(T_\infty)} - 1 \right), \tag{D.6}
\]

where \( \zeta_1 = 1 \) is used by Mason (1953) and Barrett and Clement (1988), which corresponds to a Taylor series expansion with truncation after the first-order term. For now we shall retain \( \zeta_1 \) in our equation. By use of Eq. (D.4) and the second-order term of the Taylor series expansion, its magnitude can be estimated to be:

\[
\zeta_1 \approx 1 - \frac{1}{2} \frac{L_\omega}{K_v T_\infty} \frac{T_d}{T_\infty}. \tag{D.7}
\]

Finally, combining Eqs. (D.4) to (D.6) the following equation for the droplet temperature is obtained:

\[
\left( \frac{T_d}{T_\infty} - 1 \right) = \left( x_\infty e^{-K_\infty} - 1 \right) \left[ \frac{T_\infty}{\theta_\infty} \left( \frac{p_\infty}{p_{VS}(T_\infty)} - x_\infty \right) e^{-K_\infty} + \frac{L_\omega}{R_v T_\infty} \frac{T_\infty}{T_d} \right]^{-1}. \tag{D.8}
\]

This equation is not yet an explicit equation for the droplet temperature, since the second term between square brackets still contains the droplet temperature. Therefore, it is assumed that \( T_\infty \approx T_d \) and \( \zeta_1 \approx 1 \) in order to obtain the explicit equation for the droplet temperature according to Barrett and Clement (1988):

\[
\left( \frac{T_d}{T_\infty} - 1 \right) = \left( x_\infty e^{-K_\infty} - 1 \right) \left[ \frac{T_\infty}{\theta_\infty} \left( \frac{p_\infty}{p_{VS}(T_\infty)} - x_\infty \right) e^{-K_\infty} + \frac{L_\omega}{R_v T_\infty} \right]^{-1}. \tag{D.9}
\]
In addition, Mason (1953) uses the extra assumption $\chi_\infty \ll \frac{p_\infty}{p_{\infty S}(T_\infty)}$ to find:

$$\frac{T_d}{T_\infty} = \left(\frac{T_\infty}{T_d} - 1\right) \left(\frac{T_\infty}{T_\infty} - \frac{L_\infty}{R_v T_\infty}\right)^{-1} \left(\frac{\theta_\infty p_\infty}{\theta_\infty p_{\infty S}(T_\infty)} e^{-Ke_\infty} + \frac{L_\infty}{R_v T_\infty}\right).$$ (D.10)

The approach of Gyarmathy (1963) is slightly different from that of the previous authors. First, we rewrite Eq. (D.2) to:

$$\frac{T_\infty}{\theta_\infty} \left(\frac{p_\infty}{p_{\infty S}(T_\infty)} - \chi_\infty\right) \frac{T_d}{T_\infty} - 1 = -\left(\frac{p_{VR}}{p_{\infty S}} - 1\right),$$ (D.11)

and transform the last term of this equation into a logarithm with:

$$\ln\left(\frac{p_{VR}}{p_{\infty S}}\right) = \xi_{\ln}\left(\frac{p_{VR}}{p_{\infty S}} - 1\right),$$ (D.12)

where the factor $\xi_{\ln}$ can be estimated with the help of Eq. (D.11) to be:

$$\xi_{\ln} \approx 1 + \frac{1}{2} \frac{T_\infty}{\theta_\infty} \left(\frac{p_\infty}{p_{\infty S}(T_\infty)} - \chi_\infty\right) \frac{T_d}{T_\infty}.$$ (D.13)

Combining Eqs. (D.3), (D.4), (D.11) and (D.12) to eliminate $p_{VR}$ the result is:

$$\frac{T_d}{T_\infty} = \left(\ln \chi_\infty - Ke_\infty\right) \left[\frac{T_\infty}{\theta_\infty} \left(\frac{p_\infty}{p_{\infty S}(T_\infty)} - \chi_\infty\right) \xi_{\ln} + \frac{L_\infty}{R_v T_\infty} T_d\right]^{-1}. (D.14)$$

Again to obtain an explicit equation, Gyarmathy (1963) uses the assumptions $T_\infty \approx T_d$ and $\xi_{\ln} \approx 1$:  

$$\frac{T_d}{T_\infty} - 1 = \left(\ln \chi_\infty - Ke_\infty\right) \left[\frac{T_\infty}{\theta_\infty} \left(\frac{p_\infty}{p_{\infty S}(T_\infty)} - \chi_\infty\right) + \frac{L_\infty}{R_v T_\infty}\right]^{-1}. (D.15)$$

In order to compare the explicit solutions with the exact solution an error parameter $\delta$ is defined as:

$$\delta = \frac{(T_d/T_\infty - 1)_{\text{explicit}}}{(T_d/T_\infty - 1)_{\text{exact}}}. (D.16)$$

Now, consider first the model of Barrett and Clement (1988). If the deviations due to the approximations of the latent heat and the Kelvin number are neglected, then...
Eq. (D.8) still represents the exact solution. Then, a first-order approximation of the error parameter can be obtained with the help of Eq. (D.7) by comparing Eqs. (D.8) and (D.9):

\[ \delta_{\text{barrett}} \approx \frac{\frac{1}{2} C_2^2 - C_2}{(C_1 + C_2)^2} \left( \chi_{\infty} e^{-Ke_{\infty}} - 1 \right). \]  

(D.17)

Here, the parameters \( C_1 \) and \( C_2 \) represent, respectively, the first and second term between the square brackets of the explicit equation (D.9). The first-order approximation of the error parameter for the model of Mason (1953) deviates from that for the model of Barrett and Clement (1988) due to the extra assumption made by Mason:

\[ \delta_{\text{mason}} \approx \frac{\frac{1}{2} C_2^2 - C_2}{(C_1 + C_2)^2} \left( \chi_{\infty} e^{-Ke_{\infty}} - 1 \right) - \frac{1}{C_1 + C_2} \frac{T}{\theta} \chi_{\infty} e^{-Ke_{\infty}}. \]  

(D.18)

Now, \( C_1 \) and \( C_2 \) represent the terms between square brackets of Eq. (D.10). Finally, the first-order error approximation for the model of Gyarmathy (1963) can be obtained by using Eq. (D.13) and comparing Eqs. (D.14) and (D.15):

\[ \delta_{\text{gyarmathy}} \approx \frac{\frac{1}{2} C_1^2 - C_2}{(C_1 + C_2)^2} (\ln \chi_{\infty} - Ke_{\infty}), \]  

(D.19)

where \( C_1 \) and \( C_2 \) are equal to the terms between square brackets of Eq. (D.15).
Appendix E

The production of entropy

Consider a mass element of the mixture containing exactly one droplet. The mass element has a mass $m$ and a volume $V$. The droplet has a mass $m_d$, a volume $V_d$, a surface area $A_d$ and a radius $r_d$. The temperature of the gas-vapour mixture and the droplet may be different. The fundamental equations of each constituent can be written as:

\[
\begin{align*}
\text{d}U_g &= T_d S_g - p_g d(V - V_d), \\
\text{d}U_v &= T_d S_v - p_v d(V - V_d) + \mu_v dm_v, \\
\text{d}U_d &= T_d dS_d - p_d dV_d + \sigma dA_d + \mu_\ell dm_\ell,
\end{align*}
\]

(E.1)

where $U$, $S$ and $\mu$ denote the internal energy, the entropy and the specific thermodynamic potential, respectively. Using: $U = U_g + U_v + U_d$, $S = S_g + S_v + S_d$, $p_d = p_g + p_v + 2\sigma/r_d$, $dA_d = 2\sigma/r_d dV_d$ and $dm_\ell = -dm_d$, we obtain:

\[
\text{d}S = \frac{\mu_v - \mu_\ell}{T} dm_d - \frac{T_d}{T} (1 - 1) dS_d,
\]

(E.2)

where we have applied the extra condition of adiabaticity: $\text{d}U + pdV = 0$. The entropy change of the droplet obeys:

\[
\text{d}S_d = m_d c_\ell \frac{dT_d}{T_d} + s_\ell dm_\ell.
\]

(E.3)

Starting from the fundamental relation: $\text{d}\mu = -s\text{d}T + \rho^{-1}\text{d}p$, the specific thermodynamic potentials of the vapour and the liquid can be expressed with respect to a reference state of temperature $T$ and vapour pressure $p_v = p_{vs}(T)$:

\[
\begin{align*}
\mu_v(T, p_v) &= \mu_v(T, p_{vs}) + R_v T \ln \chi, \\
\mu_\ell(T_d, p_d) &= \mu_\ell(T, p_{vs}) - s_\ell T (1 - 1) + \frac{p - p_{vs}}{\rho_\ell} + \frac{2\sigma}{\rho_\ell r_d}.
\end{align*}
\]

(E.4)

(E.5)

Here, we have approximated $s_\ell$ by a constant, since we may readily assume that $T_d \approx T$. Due to the special choice of the reference state, we have: $\mu_v(T, p_{vs}) = \mu_\ell(T, p_{vs})$. Using this and neglecting the penultimate term of Eq. (E.5), a
substitution of Eqs. (E.3) to (E.5) into Eq. (E.2) yields:

\[ \frac{ds}{ds} = R_v \left[ \ln \chi - K e(T) \right] \frac{df_d}{dT} + \frac{T_d}{T} - 1 \right] \frac{f_d}{T} \frac{dT_d}{T_d} \]

(E.6)

where \( ds = dS/m \) and \( f_d = m_d/m \). According to the explicit droplet-growth model of Gyarmathy (1963), described in Section 2.5, the droplet temperature and the saturation ratio are related by:

\[ \frac{T_d}{T} - 1 = \left[ C_1 + C_2 \right]^{-1} \left[ \ln \chi - K e(T) \right] \]

(E.7)

where \( [C_1 + C_2] \sim 10 \). Then, the ratio of the first and the second term of Eq. (E.6) is of the order of \( (df_d/f_d)/(dT_d/T_d) \), which in many cases may be estimated to be appreciably greater than unity. Therefore, we finally approximate:

\[ ds \approx R_v \left[ \ln \chi - K e(T) \right] \frac{df_d}{dT} \]

(E.8)
Appendix F. A computational flow chart

The numerical code, that is written to compute non-linear wave propagation in gas-vapour mixtures, is based on the Random-Choice method combined with a splitting technique. The calculation of phase transition during one time step occurs while the density and the internal energy of the mixture are kept constant. The computational flow chart, listed in this appendix, illustrates the procedure of the computation of the phase change.

**INPUT:**
- Constant values of $\rho$, $e$, $f_g$ and $n_d$
- Initial value of $r_d$
- Parameter values of $\Delta t$ and $r_n$

**OUTPUT:**
- Final value of $r_d$

**DEFINITIONS:**
- $a = r_d^2$ and $\phi = \phi_{r_d^2}$
- $\phi_a = d\phi/da$ and $\phi_{aa} = d^2\phi/da^2$
- $\phi_p(a') = \frac{1}{2} \phi_{aa} (a' - a)^2 + \phi_a (a' - a) + \phi$
- $\alpha_{\text{max}} = \left[\frac{3}{4\pi\rho_e} (1-f_g) (\rho/n_d)\right]^{2/3}$

**NUMERICAL CONSTANTS:**
- $\epsilon_1 = 0.25$
- $\epsilon_2 = 0.10$
- $\xi_1 = 1.1$
- $\xi_2 = 0.8$

**(SUB)ROUTINES:**
- MAIN ALGORITHM
- RELAXATION
- POSITIVE FIRST DERIVATIVE
- NEGATIVE FIRST DERIVATIVE
- EVAPORATION
Main Algorithm

BEGIN

calculate CRITICAL RADIUS $r_c = r_c(x, T)$

IF DROPLETS EXIST

$T$

IF $r_c > r_n$

END

ELSE

NUCLEATION

$r_d = \xi_1 r_n$

RELAXATION

END

IF $r_d > r_n$

T

RELAXATION

END

ELSE

F

DROPLETS DISAPPEAR

END

IF $r_c > r_n$

T

NUCLEATION

$r_d = \xi_1 r_n$

RELAXATION

END

ELSE

F

END
Relaxation

calculate $\phi, \phi_a, \phi_{aa}$ → BEGIN

$\phi > 0$ → EVAPORATION → END

$T$ → POSITIVE FIRST DERIVATIVE → END

$\phi_a > 0$ → NEGATIVE FIRST DERIVATIVE → END

Negative First Derivative

BEGIN → calculate EQUILIBRIUM SOLUTION $(a_{eq}, \phi_{eq})$

calculate $\Delta a$ with:

$$\int_a^{a + \Delta a} \frac{da'}{aa'^2 + ba' + c} = \Delta t$$

→ END

calculate PARABOLA FIT $\phi = aa^2 + ba + c$, using $a, \phi, \phi_a$ and $a_{eq}, \phi_{eq}$ → END
Positive First Derivative

\[
\begin{align*}
\text{calculate } \delta a \text{ with: } & \left| \frac{1}{2} \phi_{aa} \delta a^2 \right| = \epsilon_1 |\phi + \phi_a \delta a| \\
\delta a &= \max(a + \delta a, a_{\max}) - a \\
\text{calculate } \delta t \text{ with: } & \delta t = \int \frac{\phi_p(a')^{-1}da'}{a} \\
\end{align*}
\]

BEGIN

\[ \delta t > \Delta t \]

\[ \delta t = \Delta t \]

\[ \text{calculate } \delta a \text{ with: } a + \delta a \]

\[ \int \frac{\phi_p(a')^{-1}da'}{a} = \delta t \]

\[ \delta t = \Delta t - \delta t \]

BEGIN

\[ \phi_a < 0 \]

\[ a = a + \delta a \]

\[ \text{calculate } \phi, \phi_a, \phi_{aa} \]

\[ \Delta t = 0 \]

NEGATIVE FIRST DERIVATIVE

END
Evaporation

\[
\delta a = \frac{1}{2} \phi_{aa} \delta a^2 = \epsilon_1 |\phi + \phi_a \delta a|
\]
\[
\delta a = \min(a+\delta a, 0) - a
\]

\[
\delta t = \int \frac{\phi_p(a')^{-1} da'}{a}
\]

\[
\delta t > \Delta t \quad T
\]

\[
\delta a = \xi_2 \delta a
\]

\[
\frac{|\phi(a+\delta a) - \phi_p(a+\delta a)|}{|\phi(a+\delta a)| + |\phi(a)|} < \epsilon_2 \quad T
\]

\[
\Delta t = \Delta t - \delta t \\
\Delta t > 0 \quad T
\]

\[
\Delta t = 0 \text{ or } a = 0 
\]

\[
\phi, \phi_a, \phi_{aa}
\]

\[
\text{BEGIN}
\]

\[
\text{END}
\]
Consider a beam of monochromatic linearly polarized light with a cross-sectional area $A$ and an intensity $I_0$, incident perpendicular to a slab of droplets with thickness $l$. The droplets have a radius $r_d$ and a number density $n_d$. The attenuation of the light beam is measured with a detector having a half angle of detection $\theta_{det}$.

At a location behind the slab, the intensity of the scattered light due to a droplet at a depth $\Delta x$ in the slab can be shown to be (Bohren and Huffman, 1983):

$$I_s = I_0 e^{-\beta \Delta x} \left( k \rho \right)^{-1} \left[ i_1(a, m, \theta) \sin^2 \varphi + i_2(a, m, \theta) \cos^2 \varphi \right] e^{-\beta (l-\Delta x)}, \quad (6.1)$$

with $\rho$ as the radial distance to the droplet, the angles $\theta$ and $\varphi$ defined according to Fig. (G.1), $k$ as the wave number, and $i_1$ and $i_2$ as the scattering functions depending on $a$, $m$, and $\theta$. Then, the power scattered by the droplet and received by the detector follows to be:

$$P_s^1 = I_0 e^{-\beta l} \left( \frac{r}{k^2} \right) i_0(a, m) \theta_{det}^2, \quad (6.2)$$

provided that the angle of detection is small. In this equation, we have introduced: $i_0(a, m) = i_1(a, m, \theta=0) = i_2(a, m, \theta=0)$. It may be noted that the power given by Eq. (6.2) is independent of the position of the droplet. Therefore, the total received power due to scattering can simply be obtained by multiplying the power scattered by one droplet with the volume of the illuminated slab and the number density of the droplets:

$$\frac{P_s}{P_b} = n_d l \pi r_d^2 \frac{i_0(a, m)}{a^2} \theta_{det}^2. \quad (6.3)$$

Here $P_b$ is the power of the attenuated light beam: $P_b = Al_0 \exp(-\beta l)$. Equation (6.3), predicting the effect of scattering on light-extinction measurement, is used in Section 6.4.

Appendix H. Experimental errors

This appendix discusses the method to estimate the errors in the droplet number density and the droplet mass density due to the uncertainty $\xi$ in the determination of the transmitted light intensities. As already discussed in Section 6.3, the error $\Delta \beta_{i,j}/\beta_{i,j}$ can be calculated with Eq. (6.4.12) and the errors $\Delta r_m/r_m$ and $\Delta \epsilon$ can be obtained graphically from Fig. (6.4.4). To estimate the errors in the remaining droplet parameters, we use:

$$\langle r_d^n \rangle = r_m^n \exp \left[ \frac{[(n+1)^2 - 1]}{2} \epsilon^2 \right], \quad (H.1)$$

which is valid for a ZO\ldots function (Kerker, 1969). Assuming that:

$$\Delta \langle r_d^n \rangle /\langle r_d^n \rangle \approx \Delta \langle r_d^n \rangle /\langle r_d^n \rangle, \quad (H.2)$$

and applying Eq. (H.1), we can deduce by differentiation from Eqs. (6.4.8) and (6.4.9) that:

$$\frac{\Delta n_d}{n_d} = \left| \frac{\Delta \beta_1}{\beta_1} \right| + 2 \left| \frac{\Delta r_m}{r_m} \right| + 8 \epsilon \left| \Delta \epsilon \right|, \quad (H.3)$$

$$\frac{\Delta \rho_d}{\rho_d} = \left| \frac{\Delta \beta_1}{\beta_1} \right| + \left| \frac{\Delta r_m}{r_m} \right| + 7 \epsilon \left| \Delta \epsilon \right|, \quad (H.4)$$

The relative error in the dispersion quotient is related to the uncertainty $\xi$ in the transmitted light intensities by:

$$\frac{\Delta \beta_1}{\beta_1} = \frac{\xi}{\epsilon \beta_1}, \quad (H.5)$$
List of symbols

This list of symbols contains an explanation of the important symbols used in this thesis. Sometimes, in a given (sub)section, the meaning of a symbol may deviate from the definition in this list. Then, at those places, the local meaning is explicitly mentioned. Symbols, that are used only locally in the text or that are used solely in the appendices, are not included in this list.

Latin symbols

\( \text{a} \) \( \text{m s}^{-1} \) sound speed, defined by Eq. (3.2.12), Eq. (3.2.13), Eq. (4.2.2) or Eq. (4.3.2)

\( \text{c} \) \( \text{m}^{2} \text{s}^{-1} \) thermal diffusivity

\( B_F \) - coefficient, defined by Eq. (2.4.38)

\( B_M \) - coefficient, defined by Eq. (2.4.36)

\( B_M \) - coefficient, defined by Eq. (2.4.37)

\( c_L \) \( \text{J kg}^{-1} \text{K}^{-1} \) specific heat of liquid

\( c_p \) \( \text{J kg}^{-1} \text{K}^{-1} \) specific heat at constant pressure

\( c_v \) \( \text{J kg}^{-1} \text{K}^{-1} \) specific heat at constant volume

\( D \) \( \text{m}^{2} \text{s}^{-1} \) diffusion coefficient

\( D_m \) \( \text{kg m}^{-1} \text{s}^{-1} \) modified diffusion coefficient, defined by Eq. (2.4.13)

\( e \) \( \text{J kg}^{-1} \) specific internal energy

\( f \) - mass fraction, defined by Eq. (2.2.16)

\( f \) - function, defined by Eq. (4.2.8)

\( F \) \( \text{N} \) drag force on a droplet

\( F(r_d) \) \( \text{m}^{-1} \) droplet-size distribution function

\( g \) - mass fraction, defined by Eq. (2.2.17)

\( g \) - function, introduced in Eq. (4.2.14)

\( h \) \( \text{J kg}^{-1} \) specific enthalpy

\( H \) \( \text{J s}^{-1} \) rate of heat transfer to a droplet

\( H_{int} \) \( \text{J s}^{-1} \) rate of heat transfer into a droplet

\( I \) \( \text{J m}^{-2} \text{s}^{-1} \) light intensity

\( K_e \) - Kelvin number, defined by Eq. (2.2.10)

\( K_n \) - Knudsen number, defined by Eq. (2.4.27)

\( \ell \) \( \text{m} \) molecular mean free path, defined by Eq. (2.4.28)

\( l \) \( \text{m} \) geometric path length

\( L \) \( \text{J kg}^{-1} \) latent heat of evaporation, defined by Eq. (2.2.6)

\( L_o \) \( \text{J kg}^{-1} \) latent heat of evaporation at zero temperature
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Unit</th>
<th>Meaning</th>
</tr>
</thead>
<tbody>
<tr>
<td>( L )</td>
<td>m</td>
<td>characteristic width</td>
</tr>
<tr>
<td>( m )</td>
<td>kg</td>
<td>mass</td>
</tr>
<tr>
<td>( m )</td>
<td>-</td>
<td>refractive index</td>
</tr>
<tr>
<td>( M )</td>
<td>kg s(^{-1})</td>
<td>rate of mass transfer to a droplet</td>
</tr>
<tr>
<td>( M )</td>
<td>kg mol(^{-1})</td>
<td>molar mass</td>
</tr>
<tr>
<td>( Ma )</td>
<td>-</td>
<td>Mach number, defined by Eq. (2.4.21), Eqs. (3.4.7), Eq. (4.2.2) or Eq. (4.3.2)</td>
</tr>
<tr>
<td>( n )</td>
<td>m(^{-3})</td>
<td>number density</td>
</tr>
<tr>
<td>( Nu_F )</td>
<td>-</td>
<td>Nusselt number for momentum transfer to a droplet, defined by Eq. (2.4.12)</td>
</tr>
<tr>
<td>( Nu_H )</td>
<td>-</td>
<td>Nusselt number for heat transfer to a droplet, defined by Eq. (2.4.10)</td>
</tr>
<tr>
<td>( Nu_M )</td>
<td>-</td>
<td>Nusselt number for mass transfer to a droplet, defined by Eq. (2.4.11)</td>
</tr>
<tr>
<td>( p )</td>
<td>Pa</td>
<td>pressure</td>
</tr>
<tr>
<td>( Pr )</td>
<td>-</td>
<td>Prandtl number, defined by Eq. (2.4.24)</td>
</tr>
<tr>
<td>( Q )</td>
<td>-</td>
<td>extinction efficiency, introduced on page 105</td>
</tr>
<tr>
<td>( r )</td>
<td>m</td>
<td>radius</td>
</tr>
<tr>
<td>( r_m )</td>
<td>m</td>
<td>modal droplet radius, introduced on page 107</td>
</tr>
<tr>
<td>( \tilde{R} )</td>
<td>J kg(^{-1})K(^{-1})</td>
<td>specific gas constant</td>
</tr>
<tr>
<td>( Re_d )</td>
<td>-</td>
<td>Reynolds number, defined by Eq. (2.4.21); ( \tilde{Re}<em>d ) and ( \tilde{Re}</em>{d1} ) are introduced on page 74</td>
</tr>
<tr>
<td>( s )</td>
<td>J kg(^{-1})K(^{-1})</td>
<td>specific entropy</td>
</tr>
<tr>
<td>( S )</td>
<td>-</td>
<td>photometric signal of the Mach-Zehnder interferometer</td>
</tr>
<tr>
<td>( Sc )</td>
<td>-</td>
<td>Schmidt number, defined by Eq. (2.4.24)</td>
</tr>
<tr>
<td>( t )</td>
<td>s</td>
<td>time</td>
</tr>
<tr>
<td>( T )</td>
<td>K</td>
<td>temperature</td>
</tr>
<tr>
<td>( T_m )</td>
<td>K</td>
<td>temperature, intermediate between ( T_r ) and ( T_w ), introduced on page 31</td>
</tr>
<tr>
<td>( u )</td>
<td>m s(^{-1})</td>
<td>velocity</td>
</tr>
<tr>
<td>( U )</td>
<td>m s(^{-1})</td>
<td>velocity of a discontinuity or a condensation front</td>
</tr>
<tr>
<td>( v )</td>
<td>m s(^{-1})</td>
<td>velocity in the reference frame fixed to a discontinuity or a condensation front</td>
</tr>
<tr>
<td>( We )</td>
<td>-</td>
<td>Weber number, defined in footnote on page 73</td>
</tr>
<tr>
<td>( x )</td>
<td>m</td>
<td>space coordinate</td>
</tr>
<tr>
<td>( X )</td>
<td>-</td>
<td>parameter, defined by Eq. (2.4.19)</td>
</tr>
<tr>
<td>( Y )</td>
<td>-</td>
<td>parameter, defined by Eq. (2.4.20)</td>
</tr>
<tr>
<td>( Z )</td>
<td>?</td>
<td>arbitrary quantity</td>
</tr>
</tbody>
</table>
Greek symbols

\( \alpha \) - size parameter, introduced on page 105
\( \alpha_F \) - diffuse-reflection coefficient
\( \alpha_M \) - thermal-accommodation coefficient
\( \beta \) - condensation coefficient
\( \beta \) \( \text{kg m}^{-2} \text{s}^{-1} \) - molecular mass flux, or
\( \beta \) \( \text{m}^{-1} \) - extinction coefficient, introduced on page 105
\( \beta_{ij} \) - dispersion coefficient, defined by Eq. (6.4.6)
\( \gamma \) - ratio of specific heats
\( \epsilon \) - relative width of the droplet-size distribution,
introduced on page 107
\( \eta \) \( \text{kg m}^{-1} \text{s}^{-1} \) - dynamic viscosity
\( \kappa \) \( \text{m}^3 \text{kg}^{-1} \) - Gladstone-Dale constant
\( \lambda \) \( \text{J m}^{-1} \text{s}^{-1} \text{K}^{-1} \) - thermal conductivity, or
\( \lambda \) - wavelength of light
\( \nu \) \( \text{m}^2 \text{s}^{-1} \) - kinematic viscosity
\( \xi \) - relative error, defined by Eq. (6.4.13)
\( \rho \) \( \text{kg m}^{-3} \) - density
\( \rho_c \) \( \text{kg m}^{-3} \) - density of carrier gas
\( \sigma \) \( \text{N m}^{-1} \) - surface tension
\( \tau \) \( \text{s} \) - characteristic time
\( \tau_{\text{mom}} \) \( \text{s} \) - characteristic time, defined by Eq. (4.2.9)
\( \tau_{\text{int}} \) \( \text{s} \) - characteristic time, defined by Eq. (4.2.10)
\( \tau_{\text{wb}} \) \( \text{s} \) - characteristic time, defined by Eq. (4.2.13)
\( \tau_{\text{ev}} \) \( \text{s} \) - characteristic time, defined by Eq. (4.2.16)
\( \varphi \) \( \text{rad} \) - phase factor, according to Eq. (6.4.2)
\( \phi_{\text{f,}} \) \( \text{s}^{-1} \) - condensation rate, defined by Eq. (3.2.19)
\( \phi_{\text{r,d}} \) \( \text{m}^2 \text{s}^{-1} \) - droplet-growth function, defined by Eq. (2.5.1)
\( \chi \) - saturation ratio, defined by Eq. (2.2.11)

Subscripts

\( \omega \) - pertains to far field
\( \omega \) - pertains to critical, but see also \( \rho_c \) above
\( \omega \) - pertains to droplet
\( \omega \) - pertains to phase equilibrium or to equilibrium flow
\( \omega \) - pertains to frozen flow
Various symbols

\( D/Dt \quad \text{s}^{-1} \)

\( = \partial/\partial t + u \partial/\partial x \)

\( <Z> \quad ? \)

average droplet quantity, defined by Eq. (2.2.13)
References


Roth, P. and R. Fischer (1985): An experimental shock wave study of aerosol drop-


Summary

Non-linear wave phenomena in compressible fluids with internal relaxation due to non-equilibrium phase-transition processes are encountered in many technological and scientific environments. Examples are the unsteady periodic wave patterns that occur in steam turbines and combustion engines, or vapour explosions due to gas-tanker or gas-transport pipeline accidents. In these cases, the wave phenomenon itself is the object of interest. However, one can also use steady and unsteady gasdynamic flow as an experimental tool to create rapid changes in the thermodynamic state of a vapour or a gas-vapour mixture in order to study nucleation phenomena or to investigate droplet growth and evaporation. This dissertation reports on an experimental and theoretical investigation of two types of gasdynamic flow with phase transition. The flow phenomena studied are: 1. the unsteady non-linear rarefaction wave in a gas-vapour mixture with stimulated condensation on foreign particles added to the mixture, and: 2. the shock wave in a fog, which causes the evaporation of droplets that are present. The intention of the study was to investigate the structure of these types of waves, influenced by the phase-transition process, and to establish the validity of models to describe droplet formation and growth.

An unsteady rarefaction wave propagating in a mixture of gas and vapour results in an adiabatic cooling down of the mixture, which may produce a supersaturated vapour, depending on the expansion ratio and the initial state of the vapour. If a state of sufficiently large supersaturation is reached, droplets will be formed due to a nucleation process. This nucleation process is stimulated by the presence of condensation nuclei. The subsequent growth of the droplets by condensation results in a release of latent heat and a depletion of vapour. The condensation process can be considered as a non-equilibrium process, heading towards the establishment of a new equilibrium state of droplets in a saturated vapour environment. The structure and the behaviour of the rarefaction wave are substantially influenced by the addition of the heat of condensation to the flow.

A shock wave, moving in a gas-vapour mixture with droplets, induces a state of non-equilibrium by a sudden acceleration, compression and heating of the gas-vapour mixture, while the droplets are left undisturbed, retaining their original velocity, temperature and size. Subsequently, in a finite region behind the shock wave, the mixture tends to a new equilibrium by a relaxation of momentum and temperature and by evaporation of the droplets. The post-shock gas state may change appreciable in the relaxation zone due to the phase-transition process.

The unsteady rarefaction wave has been studied experimentally by means of
an expansion tube with a length of approx. 13 m. The test mixture consisted of water-saturated nitrogen gas at atmospheric conditions, seeded with chromium-oxide particles with radii in the range 15 ± 5 nm and with an initial number density of the order of $10^{12} \text{m}^{-3}$. At the observation point, approx. 6.5 m from the birth point of the wave, the cooling rate experienced by a fluid element did not exceed the value of 5000 K/s. Time-resolved measurements were performed of the pressure and the gas density, and of laser light attenuation at three different wavelengths. The light-extinction signals were used to determine the droplet number density and the modal droplet radius, and to obtain an estimate of the width of the droplet-size distribution. From the measured quantities, every other thermodynamic quantity of the mixture could be obtained.

For the specified experimental conditions, the wave-induced condensation process appears to be a non-equilibrium relaxation process with a characteristic time that is of the same order of magnitude as the characteristic time of expansion. Even after the onset of condensation, the rapid adiabatic expansion results in a further increase in the saturation ratio of the vapour, but thereupon the supersaturated state is broken down by the effects of condensation on the state of the gas-vapour mixture. The experimental study does not yield accurate data on the critical state for the onset of condensation, since during that stage of the process the droplets are still too small to be detected by light-attenuation measurement.

A comparison of the experimental findings with calculations performed with the Random-Choice method gives good agreement. This supports the non-linear droplet-growth model of Gyarmathy, which is based on a dynamic wet-bulb equilibrium and which incorporates the influence of the droplet size on the exchange of heat and mass between a droplet and its environment. Moreover, the calculation shows that a droplet-formation model based on the instantaneous wetting of the condensation nuclei gives acceptable results.

A theoretical study is presented on the asymptotic solution that develops when $t/\tau \rightarrow \infty$, with $\tau$ as the relaxation time for condensation. It is argued that the asymptotic wave is self-similar and that two types of solutions exist. If the onset of condensation occurs at a saturation ratio of unity, the rarefaction wave is split into two zones separated by a uniform region. This is due to the discontinuous character of the equilibrium speed of sound at the onset point. However, in the case that condensation is delayed until a fixed critical supersaturation is reached that deviates considerably from unity, then a condensation discontinuity of the expansion type is part of the solution. A simulation with the Random-Choice method confirms that the experimentally observed wave structure develops to the self-similar form derived for the asymptotic limit.
The treatment of the shock wave in a fog starts with an analysis of the transport processes in the relaxation region behind the shock wave. Expressions are obtained for the characteristic times for droplet heat diffusion, for momentum relaxation, for the formation of the wet-bulb state and for droplet evaporation. A numerical evaluation of the characteristic times for the system of water droplets in humid nitrogen gas gives as a result that, for weak shock waves and relatively small droplet sizes, the relaxation of momentum and droplet temperature takes place at a time scale that is short compared to the total-evaporation time of the droplets. Therefore, for this regime, shock-induced evaporation can be described as a process governed by heat conduction and vapour diffusion to and from the droplets, while the droplet temperature remains at its wet-bulb value and the droplets are stagnant with respect to the gas.

The stagnant and stationary fog that forms in the test tube after a twofold unsteady expansion is found to be a suitable medium for studying the response of a fog to shock compression. Experimental results on shock-induced relaxation are presented for water droplets in humid nitrogen gas. The shock Mach number varies from 1.23 to 1.88, the initial droplet mass fraction is approximately equal to $7 \times 10^{-3}$ in all cases, and the initial droplet radius is in the range $1.0 - 1.7 \, \mu m$. The observations show that the relaxation of momentum is an almost instantaneous process at the time scale of droplet evaporation. In the relaxation zone, the gas state variables change due to the evaporation of the droplets. The change is appreciable for the weaker shock waves. The rate of evaporation appears to vary strongly with the shock strength. A variation of about a decade was observed for the Mach-number range studied. The evaporation process cannot satisfactorily be described by a simple model that neglects the evaporation-induced change in the gas state and that does not account for the droplet-size dependence of the transfer coefficients for heat and mass transfer. An extended model, based on the steady conservation laws and on the droplet-growth model of Gyarmathy, gives an accurate description of the structure of the evaporation zone and of the rate of evaporation for the appreciable range of experimental conditions obtained by varying the shock strength.

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Samenvatting

Niet-lineaire golfverschijnselen in compressibele media met interne relaxatie ten gevolge van fasentransitie onder niet-evenwichtscondities worden waargenomen in veel technologische en wetenschappelijke situaties. Enkele voorbeelden zijn de instationaire periodieke golfpatronen die optreden in verbrandingsmotoren en stoomturbines, of dampexplosies bij gastankers- of gastransportleiding-ongevallen. In deze gevallen richt de interesse zich op het golfverschijnsel zelf. Echter, stationaire en instationaire gasdynamische stromingen kunnen ook worden toegepast als experimenteel gereedschap voor het genereren van snelle veranderingen in de thermodynamische toestand van een damp of een gas/damp-mengsel voor het bestuderen van nucleatieverschijnselen of voor het onderzoeken van druppelgroei en verdamping. Dit proefschrift beschrijft een experimentele en theoretische studie naar twee verschillende gasdynamische verschijnselen met fasentransitie. Het betreft hier: 1. de instationaire niet-lineaire expansiegolf in een gas/damp-mengsel met gestimuleerde condensatie op aan het mengsel toegevoegde vreemde deeltjes, en: 2. de schokgolf in een nevel, welke leidt tot het verdampen van aanwezige druppels. Het onderzoek was gericht op een beschrijving van de structuren van beide golftype, beïnvloed door het fasetransitieproces, en een vaststelling van de geldigheid van modellen voor druppelvorming en groei.

Een instationaire expansiegolf, zich voortplantend in een mengsel van gas en damp, heeft een adiabatische afkoeling van het mengsel tot gevolg. Afhankelijk van de mate van expansie en de begintoestand van de damp, leidt dit tot een oververzadigde toestand voor de damp. Is de graad van oververzadiging voldoende hoog, dan ontstaan er druppels ten gevolge van een nucleatieproces. Dit proces wordt bevorderd door de aanwezigheid van condensatiekernen. De daaropvolgende groei van de druppels door condensatie gaat gepaard met het vrijkomen van latente warmte en met het afnemen van de dampconcentratie. Het condensatieproces kan men beschouwen als een proces dat beheerst wordt door de verstoring van het evenwicht en dat er naar streeft een nieuw evenwicht van druppels in een ver­zadigde omgeving van damp te bewerkstelligen. De structuur en het gedrag van de expansiegolf worden in belangrijke mate beïnvloed door de absorptie van de condensatiewarmte door de stroming.

Een schokgolf, zich verplaatsend door een gas/damp-mengsel met druppels, veroorzaakt een toestand van niet-evenwicht, welke ontstaat door een plotselinge versnelling, compressie en verhitting van het gasmengsel, terwijl de druppels hun oorspronkelijke snelheid, temperatuur en grootte behouden. Daaropvolgend neigt het mengsel naar een nieuwe evenwichtstoestand via de relaxatie van impuls en
temperatuur en via de verdamping van de druppels. In de relaxatiezone kan de toestand van het gas een aanzienlijke variatie vertonen ten gevolge van het proces van fase transitie.

De instationaire expansiegolf is experimenteel bestudeerd met behulp van een expansiebuis met een lengte van ong. 13 m. Het testmengsel bestond uit met waterdamp verzadigd stikstofgas onder atmosferische condities, waaraan chroomoxide-deeltjes waren toegevoegd met stralen in het bereik van 15 ± 5 nm en met een iniëtiële concentratie van de orde van \(10^{12} \text{m}^{-3}\). De afstand tussen het punt van observatie en het ontstaanspunt van de golf was ong. 6.5 m. De koelsnelheid ter plaatse van het observatiepunt, zoals ondervonden door een stromingselementje, kwam niet boven 5000 K/s uit. Tijdsopgeloste metingen werden verricht van de druk, de gasdichtheid en van laserlichtverzwakking bij drie verschillende golflengten. De lichtextinctie-signalen werden gebruikt ter bepaling van de druppelconcentratie en de modale druppelstraal en ter verkrijging van een schatting voor de breedte van de druppelgroottesverdeling. Met behulp van de gemeten grootten kon elke andere thermodynamische grootte van het mengsel worden bepaald.

Voor de specifieke experimentele omstandigheden blijkt het golfgeinduceerde condensatieproces een relaxatieproces te zijn waarvan de karakteristieke tijd van dezelfde orde van grootte is als de karakteristieke tijd van expansie. Zelfs na initialisatie van het condensatieproces leidt de snelle adiabatische expansie tot een verdere toename van de verzadigingsgraad van de damp. Maar vervolgens wordt deze toestand van oververzadiging afgebroken door de invloeden van condensatie op de toestand van het gas/damp-mengsel. De experimentele studie geeft geen uitsluitend omtrent de kritische toestand behorende bij de initialisatie van het condensatieproces, omdat de druppels tijdens deze fase van het proces nog te klein zijn om te worden waargenomen d.m.v. een registratie van de lichtverzwakking.

Een vergelijking van de experimentele bevindingen met berekeningen uitgevoerd met de "random choice"-methode geeft een goede onderlinge overeenstemming. Dit ondersteunt het niet-lineaire druppelgroveimodel van Gyarmathy, dat gebaseerd is op een dynamisch natteboulevendrecht en dat de invloed van de druppelgrootte op de uitwisseling van warmte en massa tussen een druppel en zijn omgeving in rekening brengt.

Een theoretische beschrijving wordt gegeven van de golfvorm, die ontstaat wanneer \(t/\tau \rightarrow \infty\), waarbij \(\tau\) staat voor de karakteristieke tijd voor condensatie. Er wordt getoond dat de asymptotische oplossing gelijkvormig is en dat er twee verschillende golfoplossingen bestaan. Als het ontstaan van druppels optreedt bij een verzadigingsgraad die gelijk is aan 1, dan bestaat de expansiegolf uit twee gedeelten gescheiden door een uniform gebied. Dit is een gevolg van het discontinue karakter.
van de evenwichtsgeluiddensnelheid in the transitiepunt. Echter in het geval dat het begin van condensatie uitblijft, totdat een gegeven kritische verzadigingsgraad wordt bereikt die aanzienlijk afwijkt van 1, dan zal een condensatiediscontinuïteit van het expansietype onderdeel zijn van de oplossing. Een simulatie met de "random choice"-methode bevestigt dat de experimenteel waargenomen golfstructuur evolueert naar de gelijkvormigheidssstructuur afgeleid voor de asymptotische limiet.

De behandeling van de schokgolf in een nevel wordt begonnen met een analyse van de transportprocessen in het relaxatiegebied achter de schokgolf. Dit leidt tot uitdrukkingen voor de karakteristieke tijden voor de warmtediffusie in de druppel, voor de relaxatie van impuls, voor de vorming van de natteboltoestand en voor de verdamping van de druppels. Een numerieke evaluatie van deze karakteristieke tijden voor een systeem van waterdruppels in een mengsel van waterdamp en stikstoöf gas voorspelt dat, voor zwakkere schokgolven en relatief kleine druppels, de relaxatie van impuls en van de druppeltemperatuur plaatsvindt op een tijdschaal die kort is vergeleken met de tijd benodigd voor een volledige verdamping van de druppels. Voor het betreffende regiem kan daarom de schokgolfgeëinduceerde verdamping worden beschreven als een proces dat bepaald wordt door diffusie van damp en geleiding van warmte van en naar de druppels, terwijl de druppeltemperatuur gelijk blijft aan de nattebolwaarde en de druppels in rust zijn ten opzichte van het gas.

De stil staande en stationaire nevel, die zich ontwikkelt in de testbuis na een tweevoudige instationaire expansie van het gas/damp-mengsel, blijkt een geschikt medium te zijn ter bestudering van het gedrag van een nevel na schokcompressie. Experimentele resultaten met betrekking tot schokgeëinduceerde relaxatie worden gepresenteerd verkregen voor een mengsel van waterdamp en vochtig stikstof gas. Het Machgetal van de schokgolf varieert van 1.23 tot 1.88, de initiële massafractie vloeistof is in alle gevallen bij benadering gelijk aan 7 10^-3 en de initiële druppelstraal ligt in het bereik 1.0 - 1.7 µm. De waarnemingen tonen aan dat op de tijdschaal van verdamping de relaxatie van impuls nagenoeg als een instantaan proces kan worden beschouwd. In de verdampingzone treedt een variatie op van de groot heden van de gasfase, veroorzaakt door de verdamping van de druppels. De variatie is aanzienlijk voor de zwakkere schokgolven. De verdampingsnelheid blijkt sterk te variëren met de schoksterkte. Een variatie van ongeveer één decada wordt gevonden voor het bestudeerde bereik van Machgetallen. Het verdampingsproces kan niet bevredigend worden beschreven met een eenvoudig model dat de verdampingseïnduceerde verandering van de gasfase verwaarloost en dat geen rekening houdt met de afhankelijkheid van de overdrachtscoëfficiënten voor warmte- en massaoverdracht van de druppelgrootte. Een vollediger model, dat gebaseerd is op
de stationaire behoudswetten en op het druppelgroeimodel van Gyarmathy, geeft een nauwkeurige beschrijving van de structuur van de verdampingszone en van de snelheid van verdamping voor het behoorlijke bereik van experimentele condities verkregen door variatie van de schoksterkte.
Nawoord

Een promotie-onderzoek kan goed worden vergeleken met een overlevingstocht door onbekend en onherbergzaam terrein. Voordat men aan de reis begint, bestaat er slechts de spanning van het onbekende en de ambitie om te slagen. Door oefening en training heeft men getracht zich zo goed mogelijk voor te bereiden, maar of de capaciteit en conditie voldoende zijn is nog een open vraag. Dan begint de tocht met een voorzichtige verkenning van het terrein, al zwakkend zonder goed te weten waar men heen moet. Gaandeweg ontstaat het reisritme en struikelt men niet meer over elke steen. De tocht is zwaar en vergt veel van lichaam en geest. Af en toe vraagt men zich af of het dit allemaal wel waard is. Maar des te sterker is de bevrediging als na een zware en vermoeiende klim zich een prachtig vergezicht openbaart. Tenslotte, staande op de laatste berg, ziet men het bestemmingspunt en weet men dat de reis zal worden volbracht, mits de kiezen nog even op elkaar worden gebeten. Na de tocht heerst de voldoening over de prestatie en blijft vooral de opgedane ervaring met betrekking tot eigen kunnen als waardevol element voor de toekomst.

Diegenen, die ooit een overlevingstocht hebben aangedurft, weten hoe belangrijk de reisgenoten zijn, bijvoorbeeld voor het meedenken over de te kiezen route, of voor het overnemen van de rugzak als het even niet meer gaat, het verzorgen van de blaren of het inspreken van moed, of om gewoon samen mee te lachen. De vergelijking met promoveren houdt hier dan ook niet op. Ook promoveren is teamwork. Daarom zou ik al die mensen die hebben bijgedragen aan dit werkstuk van harte willen bedanken voor hun inzet, stimulans en betrokkenheid. Met name wil ik dank zeggen aan prof. Vossers, voor zijn ook tijdens zijn emeritaat steeds aanwezige interesse voor dit onderzoek, aan Rini van Dongen, Harm Jager, Eep van Voorthuizen en Jan Willems, als onmisbare en enthousiaste medewerkers van de vakgroep Transportfysica, aan de studenten Kees Braun, Corné de Kok, Rick de Lange, Ed Niessen en Peter de Wit, welke als afstudeerders een wezenlijke bijdrage hebben geleverd aan dit onderzoek, en aan de stagiaires Rob Bergmans, Eric Brentjens, Rinus Broekmans, Huub Dankers, Bart de Graaf, Bart van Hest, Erik Korevaar, Bruno Mulder, Marina Theeuwen en Johnny Zweegers, die altijd voor de nodige drukte en bedrijvigheid rond de experimentele opstelling hebben gezorgd.
Curriculum Vitae


1968 - 1974 Basisonderwijs aan de Maria-school te Someren.


29/5/1980 Diploma atheneum.


11/2/1987 Doctoraal examen.


1. De tegenwoordige beschikbaarheid van handzame spectrometers en snelle CCD-arrays biedt veelbelovende mogelijkheden voor de experimentele registratie van druppelgroei-processen door middel van lichttransmissie-metingen.


3. Bij zijn beoordeling van het algoritme van Phillips en Twomey ter herleiding van een verdelingsfunctie uit een verzameling meetgegevens weegt Walters onvoldoende dat het voornoemde algoritme een arbitraire keuze van de waarde van een weegfactor vereist.


4. Een alternatief voor de schokbuismethode voorgesteld en gebruikt door Peters ter bestudering van het proces van homogene nucleatie bestaat uit het toepassen van een plaatselijke verwijding in de lagedruk-sectie van de schokbuis. Dit alternatief verdient de voorkeur.


5. De energiebalans opgesteld door Paikert voor de interface van een druppel en zijn vrije-moleculaire omgeving bevat een onjuiste dubbele bijdrage van de enthalpie toegevoerd aan de druppel door condenserende damp.

   B. Paikert, proefschrift Universitàt Essen, 1990.

6. Bij zijn beschrijving van condensatie en verdamping in binaire mengsels als stromingsprobleem, gaat Oswatitsch er ten onrechte van uit dat de arbeid verricht door de druk op de vloeistof in een controlevolume mede bepaald wordt door de beweging van de rand van het volume.

7. Bij de instationaire uitstroming uit een spleetvormige opening met scherpe randen kan de invloed van wrijving voor hoge waarden van het getal van Reynolds in rekening worden gebracht door het toepassen van de z.g. Kutta-voorwaarde aan beide randen. Toepassing van een soortgelijke conditie op de centrale stroomlijn in het uitstroomvlak, zoals gesuggereerd door S.A. Elder, leidt niet tot correcte resultaten.


8. Ook indien de porievloeistof van een poreus materiaal bestaat uit een mengsel van water en luchtbellen, kunnen twee afzonderlijke Biot-golven worden geïdentificeerd.

_D.M.J. Smeulders, proefschrift Technische Universiteit Eindhoven, 1992._

9. Met het oog op een vergroting van de vaardigheid in de engelse taal zal een vervuilde aandacht voor het engels in het basisonderwijs meer effect sorteren dan het voeren van deze taal in het universitair onderwijs.