Optimal design of spiral sterilisers

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Optimal design of spiral sterilisers

PROEFSCHRIFT

ter verkrijging van de graad van doctor aan de
Technische Universiteit Eindhoven, op gezag van de
Rector Magnificus, prof.dr. R.A. van Santen, voor een
commissie aangewezen door het College voor
Promoties in het openbaar te verdedigen
op maandag 13 januari 2003 om 16.00 uur

door

Jasper David Hidde Kelder

geboren te Groningen
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Subject headings: food technology; sterilization / food gelling; starch / food rheology; viscous flow / curved flow and heat transfer / physicochemical simulation and modelling

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to my parents
Summary

This work reports numerical simulations of the thermal processing of laminar gelatinising and gelatinised starch suspensions in coiled tubular ultra high temperature sterilisers. A 3-dimensional model of the flow, heat transfer, and related physico-chemical changes in toroidal pipes of extreme length was developed in the commercially available Star finite volume code. To account for gelatinisation at high shear and high heating rates typical of industrial scale processing, we propose a rheological model employing gelatinisation classes.

Curvature was shown to increase both frictional losses and heat transfer, both in developed and in developing power-law flow. Pseudoplasticity decreases friction factors and improves heat transfer relative to Newtonian flow, whereas dilatancy effects the opposite. Pitch and free convection are unimportant under the current processing conditions. Hydrodynamic and thermal flow development are accelerated in curved tube flow.

In toroidal heater flow, heat transfer and viscous development of a gelatinising model food were simulated. A gelatinisation class model incorporated the direct shear and temperature dependency, as well as the history effects of both parameters. Curvature improves heat transfer, and reduces the severity of the channeling phenomenon associated with gelatinisation. For minimum starch consumption, the smallest practical coil diameter and the lowest possible product velocity are recommended.

In the ultra high temperature section consisting of a heater, a holder and a cooler, the impact of centrifugal and buoyant forces on viscous power-law foods regarding lethality development and thiamine concentration was analysed. Curvature and pseudoplasticity substantially increase heat transfer and render the axial velocity profile more uniform. As a result, processing times in the heater and cooler are shorter, greatly improving thiamine retention.

In addition to a thorough experimental validation of the current results, further areas of interest include the prediction of local fouling rates for milk based products, and the incorporation of the turbulent-laminar transition expected in gelatinising suspensions.
Samenvatting

Dit proefschrift doet verslag van de numerieke simulatie van de thermische behandeling van verstijfzende zetmeelsuspensies in gespiraliseerde ultra hoge temperatuur sterilisatoren. Voor buizen van extreme lengte werd in de Star eindige volume code een 3-dimensionaal model geconstrueerd van de strooming, de warmteoverdracht en de veranderende stofeigenschappen. Daarbij is gebruik gemaakt van een verstijfzeldingsklassenmodel om de rheologie van een verstijfzandelende suspensie voor de hoge verhittings- en afschuifnelheden te beschrijven.

Kromming verhoogt de stromingsweerstand en verbetert de warmteoverdracht, zowel voor ontwikkelde als ontwikkelende power-law vloeistoffen. Pseudoplasticiteit vermindert de stromingsweerstand en verhoogt de warmteoverdracht ten opzichte van de Newtonse rheologie. Het omgekeerde is het geval voor dilataties. De spoed van de spiraal en de invloed van vrije convectie zijn onbelangrijk voor de beschouwde procescondities. Tenslotte versnelt kromming de hydrodynamische en thermische ontwikkeling van de stroming.

Een verstijfzeld modelproduct is gesimuleerd in een spiraalverhitter zonder spoed. Daarbij werd de invloed van zowel de directe als de cumulatieve invloed van de afschuifnelheden en temperatuur gemodelleerd door gebruikmaking van een verstijfzeldingsklassenmodel. Kromming verbetert de warmteoverdracht en onderdrukt het optreden van de kortsluitstroming als gevolg van een meer uniforme verstijfseling. Het laagste zetmeelverbruik wordt verkregen voor de kleinste mogelijke spiraaldiameter, en voor de laagste stroomsnellheid.

In het hoge temperatuurtraject bestaande uit een verhitter, een houder en een koeler is de invloed van centrifugale en opwaartse krachten op de steriliteit en de thiamine concentratie van een hoogviscose power-law vloeistof gesimuleerd. Kromming en pseudoplasticiteit verbeteren de warmteoverdracht aanzienlijk en verkleinen de snelheidsverschillen in de doorsnede. Hierdoor zijn de verblijftijden in de verhitter en de koeler korter, en is de afbraak van thiamine geringer.

Naast een grondige experimentele validatie van de huidige simulaties, vormen de vervuilingssnelheid van producten gebaseerd op melk en de turbulent-laminaaire omslag voor verstijfzellende producten interessante onderzoeksgebieden.
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Chapter 1

Introduction

1.1 Continuous aseptic processing

Most food products that are consumed some time after preparation need protection against the activity of enzymes or microbes. Throughout history, foods have been dried, frozen, salted, sugared or acidified to achieve this (Hugo, 1996). However, when the product is a liquid and the original fresh flavour needs to be retained, for example when dairy products or fruit juices are concerned, different conservation techniques are required. One such aseptic procedure (derived from the Greek adjective 
\textit{aseptikos} - not liable to putrefy) is heat treatment, involving heating, holding and cooling of the food to kill pathogens and inactivate enzymes.

As early as 1810, heating of food products in glass jars was applied to extend their shelf-life up to months (Hugo, 1996). However, it was not until the work of Pasteur (Pasteur, 1857) that the underlying fundamentals of bacterial growth and spoilage were understood. By 1900, in-can pasteurisation (a mild treatment at 55 - 70°C killing most bacteria) and sterilisation (a more severe treatment at 110 - 120°C, inactivating bacteria and their heat resistant spores) were well established. Unfortunately, heating rates were limited in the absence of convective heat transfer, and long processing times and modest product throughputs were inevitable. To ensure sufficient heating of the coldest point, the heat treatment could be harsh, resulting in excessive losses of valuable constituents (e.g. vitamins), accompanied by unacceptable changes in taste or colour.

To avoid such over-processing and to increase throughput, continuous flow sterilisers were patented and constructed before the turn of the nineteenth century, but it was not before 1921 that the first successful aseptic canning system was developed. Though many different continuous flow type
heat exchangers continued to be developed between 1900 and 1950, only the introduction of aseptic cartoning systems in 1961 enabled the world-wide success of aseptic continuous processing (Burton, 1988).

It was soon appreciated that at higher temperatures the inactivation of microbes and their spores exhibited higher rates than the reduction in vitamins or changes of other food properties. High temperatures applied for short times were therefore found to have the same aseptic effect as longer processing times at lower temperatures, whereas the destruction of nutrients was dramatically reduced. Further exploitation of this phenomenon led to the concept of Ultra High Temperature (UHT) sterilisation, a name applicable to continuous processes using maximum temperatures in the 130 - 150 °C range for 1 - 10 seconds (Holdsworth, 1992).

Because of the required heating rates, UHT-sterilisation can only be performed beneficially using continuous heat exchangers. Two main categories of continuous processing equipment can be discerned: direct and indirect. Direct systems involve a heating stage where steam condenses into the product ('injection') or product is injected into condensing steam ('infusion'), a holding period and subsequent cooling by flashing of the condensate. Indirect heating can be performed in tubular, plate or scraped surface heat exchangers, and contrary to direct systems, the heating medium is never in direct contact with the product. Though fouling is generally reduced in direct UHT-plants and the product quality is slightly better, indirect heating processes are generally less complex, and feature lower equipment and operating costs (Burton, 1988; Lewis and Heppell, 2000).

The simplest possible flow diagram of an indirect UHT-plant is shown in figure 1.1. An untreated product is heated (in a series of heat exchangers) to the sterilising temperature, led through a holding section and cooled (in a series of heat exchangers). Usually the pumping power is provided by a positive displacement pump having 3 - 5 pistons, as the frictional losses may be large, especially in the case of viscous foods. Some products (e.g. milk, cream) require homogenisation (or redispersion of fat globules), which is achieved by pressing the liquid through a narrow slit. To prevent boiling in the system, a reducing valve is located prior to the aseptic filling machine. Of course the schematics of an actual UHT-plant are far more complex, as considerable piping is installed to recover energy.

Of the indirect heat exchangers, perhaps the most versatile regarding the range of products and of operation, are tubular heat exchangers. Mainly necessitated by a sufficient heat recovery, heating and cooling pipes are usually long (on the order of 100 m). To arrive at compact steriliser designs, these heat exchangers consist of a large number of straight tubes in series, or they can be wound into coils. Since the late 1940-ties, the latter strategy
has been employed by Stork Food & Dairy Systems B.V. in their concentric tube Sterideal steriliser (Stork B.V. (1998), figure 1.2).

Originally, concentric coils were chosen for reasons of robustness and compactness. However, the curvature of the Sterideal proved to bring unexpected benefits in the processing of viscous products as a result of its higher heating rates relative to straight tube heat exchangers. It is amongst the objectives of this dissertation to quantify such benefits.

Lately, novel techniques are being developed that promise safe products of an even better quality than UHT-sterilisation. Among those are Ohmic heating, Pulsed Electric Field (PEF) sterilisation, membrane processes, irradiation and Ultra High Pressure (UHP) sterilisation (Barbosa-Canovas, Pothakamury, Palou, and Swanson, 1998). Though these techniques may offer specific benefits for some products, conventional continuous UHT-sterilisation still offers, and will continue to offer, an inexpensive and efficient means to produce bacteriologically safe product of a high nutritional quality (Lewis and Heppell, 2000).
1.2 Aim and outline

The design of continuous straight tube heat exchangers for turbulent, low-viscosity foods is well established. However, sterilisation and preparation of viscous foods such as sauces and desserts, where thickening agents cause dramatic changes in viscosity in the course of the process, is little explored. The main focus is therefore on the following:

*It is the aim of this work to simulate the flow, heat transfer and changing functional properties (viscosity, sterility and nutritional content) of a viscous model dessert containing a thermally activated thickener (starch), as it is heated, sterilised and cooled in a coiled UHT-steriliser.*

Hence, a 3-dimensional model of the flow and heat transfer in loosely coiled sterilisers of extreme length (200 - 400 m) was built in the commercially available Star finite volume code\(^1\). Adopting a body-force formulation, the complex rheology of gelatinising starch suspensions could be incorporated at a reasonable computational cost.

In chapter 2, curved developed and developing flow and heat transfer of constant property power-law fluids are treated. Chapter 3 covers the time-shear- and temperature dependent changes associated with gelatinisation in dilute starch suspensions. The practical implications for the processing of a gelatinising model food in the heater of an UHT-steriliser are presented in chapter 4. Chapter 5 is dedicated to the heat transfer, sterilisation and nutrient retention in the high temperature section \((T > 100 \, ^{\circ}\text{C})\) of a coiled UHT-steriliser. Finally, chapter 6 provides some recommendations regarding validation and further research.

Chapter 2 to 5 have been laid out as scientific papers, and can, apart from additional references to the appendices, be read as self-contained units. Inherent to this approach is some overlap between chapters and absence of separate final conclusions. I would like to apologise to the reader for any inconveniences thus caused.

\(^1\)www.cd.co.uk
Bibliography

Chapter 2

Flow and heat transfer in coils

Flow and temperature fields for developed and developing viscous toroidal power-law flow were simulated and validated. Friction factors and heat transfer were shown to be significantly higher in curved flow as a result of the distorted axial velocity field and the occurrence of Dean-vortices. Pseudoplasticity decreases frictional losses while further increasing heat transfer, whereas dilatancy effects the opposite. In dilatant high Dean number flows, a periodic unsteady phenomenon is predicted. Helicity and buoyancy are briefly discussed and are unimportant for the processing conditions expected in industrial coiled heaters.

2.1 Introduction

Curvature is a salient feature of the Stork Sterideal coiled steriliser, and this chapter is dedicated to its effect on performance in terms of frictional losses and heat transfer. Berger, Talbot, and Yao (1983) and Ito (1987) have reviewed the large body of analytical and numerical work on the flow in curved tubes, and experimental correlations for friction factors were discussed by Srinavasan, Nandapurkar, and Holland (1968) and Gnielinski (1986). Bau- rmeister and Brauer (1979) reviewed early analytical and numerical studies of heat transfer in curved pipes, and Manlapaz and Churchill (1981) discussed the experimental work. General overviews of the implications of curvature for flow and heat transfer engineering can be found in Shah and Joshi (1987) and Ebadian and Dong (1998).

Laminar flow and temperature fields in curved pipes deviate from those in straight tubes because of centrifugal forces. As the axial velocity profile in pipe flow is non-uniform and centrifugal forces are proportional to the
square of the axial velocity, fast flowing fluid near the axis of the tube will experience a greater force perpendicular to the axis of the coil than slow fluid closer to the wall. This results in a secondary flow in the cross-section, which consists of two symmetric vortices (figure 2.1, middle). These vortices are termed Dean vortices, after W.R. Dean, who published the first mathematical treatise on curved flow (Dean, 1927). For developed and strongly curved flow, the distribution and magnitude of the centrifugal forces is such that the maximum of the axial velocity is moved from the tube centerline towards the outside of the coil (figure 2.1, left).

Dean vortices have a marked influence on the temperature field in a curved tube, which is illustrated for the case of a heated fluid (figure 2.1, right). For sufficiently high Prandtl number $Pr$, the single temperature minimum of straight pipe flow is replaced by two symmetrically located temperature minima away from the tube centerline. The Dean vortices transport fluid along the hot wall from the outside to the inside of the coil, and this heated fluid subsequently penetrates the cold core. The center of each vortex roughly coincides with a temperature minimum, as it is excluded from this convective heat transfer mechanism.

Dean vortices can have significant practical implications for the design of curved tubular heat exchangers. First, friction factors in curved pipes are higher than those in equivalent straight pipes. This results from the additional dissipation of the secondary flow, but more importantly from the steeper gradients in axial velocity at the outer wall of the curved pipe. The local friction factor at the inner wall of the pipe decreases, but over-all an increase in peripherally averaged friction factor occurs (figure 2.2).

Second, heat (and mass) transfer rates are usually higher in curved than in straight pipes. In straight developed laminar pipe flow, conduction is the
only heat transfer mechanism in the fluid, whereas in curved pipes the Dean vortices provide additional convective transport. Another benefit comes from the shift of the temperature extrema to the pipe wall. Temperature gradients and local heat transfer towards the outer wall of the curved tube increase, while at the inner wall gradients and transfer rates decrease. Over-all a substantial increase of the peripherally averaged heat transfer may result (figure 2.3).

Third, axial dispersion is strongly reduced by the secondary flow in curved pipes (Trivedi and Vasudeva, 1975), even for very weak curvature (Singh and Nigam, 1981). Reduction of the axial dispersion and a more uniform axial velocity profile at higher curvature result in narrower residence time distributions (Ranade and Ulbrecht, 1981; Saxena, Nigam, and Nigam, 1983).

Fourth, curvature reduces the hydrodynamic and thermal development lengths. Though hydrodynamic entrance length is usually unimportant in design calculations, it is further reduced in curved flows (Berger et al., 1983). Thermal development in laminar flow however can be slow, but curvature may reduce the entrance length by 20 - 90%. For engineering purposes, use of developed heat transfer correlations is therefore often recommended (Ebadian and Dong, 1998).

Finally, curvature stabilises the flow and transition from laminar to turbulent flow is delayed. Straight developed pipe flows usually becomes unstable and turbulent above a Reynolds number $Re = 2100$, but for practical values of pipe diameter $d$ and coil diameter $D$, this may be as high as 6000 - 8000. The exact explanation for this is still lacking (Webster and Humphrey, 1993; Yamamoto, Akita, Ikuechi, and Kita, 1995) but perhaps the secondary convection suppresses the axial propagation of initial turbulent fluctuation at the outer wall. Empirical equations for the critical Reynolds number $Re_c$ were given by Ito (1959), Schmidt (1967) and Srinavasan et al. (1970); because of their very similar predictions, only the latter is presented (equation 2.1).

$$Re_c = 2100(1 + 12\sqrt{d/D}) \quad (2.1)$$

Beyond the critical Reynolds number the flow is fully turbulent. Dean vortices are present here as well, but due to the turbulent fluctuations of the flow, the transfer of momentum and heat across the fluid is much higher than in laminar flow, making the effects of the secondary flow on the friction factor and heat transfer less important. Next to this, turbulent axial velocity profiles are more uniform than those in laminar flow, reducing the driving force behind the Dean vortices.
Reynolds number $Re$, the thermal entrance length $z^*$ and the Prandtl number $Pr$ characterise the flow and heat transfer in straight tubes when dissipation is neglected. In curved flows the Reynolds number is usually replaced by the Dean number $De$ (equation 2.2), and for strong curvature ($d/D > 0.1$) the radius of curvature $d/D$ is added. The Reynolds and Prandtl number are used in a generalised form (Metzner and Reed, 1955) applicable to power-law flows. In that case Reynolds is derived from the relation $f = 64/Re$, in which $f$ is the friction factor. The Prandtl number is defined such that the product of the Reynolds and the Prandtl number yields the Péclet number $Pe$.

\[
De = Re \sqrt{\frac{d}{D}} \quad (2.2)
\]

Friction factor correlations are often presented as the normalised friction factor $f_n$, which is the ratio of the curved ($f$) to the straight ($f_s$) tube friction factor (equation 2.3).

\[
f_n = \frac{f}{f_s} = \mathcal{F}(De, d/D, z^*) \quad (2.3)
\]

Heat transfer correlations are usually presented as the dimensionless Nusselt number $Nu$ (equation 2.4). For the developed flow and temperature field in a straight tube $Nu$ is equal to 3.657 for constant wall temperature ($T$-boundary) and 4.364 for constant heat flux ($H_1$-boundary) (Shah and London, 1978).

\[
Nu = \mathcal{F}(De, d/D, z^*, Pr) \quad (2.4)
\]

For loosely coiled pipes ($d/D \leq 0.1$) and hydrodynamically and thermally developed flow, flow and heat transfer in curved tubes are a function of $De$ or $De$ and $Pr$ respectively.

Figure 2.2 shows six experimental friction factor correlations for developed, isothermal, Newtonian flow in curved pipes within their ranges of validity. It should be noted that the variation of the Dean number has been obtained by variation of the fluid viscosity, keeping the other fluid properties, the coil geometry ($d = 0.025$ m and $D = 1$ m) and the average fluid velocity ($\bar{\omega} = 0.5$ m/s) constant. This is a realistic approach, since during heating and cooling density, heat capacity and thermal conductivity remain roughly constant,
Figure 2.2: Developed normalised friction factors for curved laminar flow $(d/D = 0.025)$.

whereas the viscosity may change substantially. Especially in the case of physical changes in the fluid flow (e.g. gelatinisation, please refer to chapter 3), the viscosity may change several orders of magnitude.

Relationships for friction factors generally agree fairly well, as measuring pressures in tubes is a rather straightforward matter. For developed flows, Gnielinski (1986) recommends the relationship of Mishra and Gupta (1979a) after correction for non-isothermal effects. This correlation was chosen as a reference in this thesis as it has by far the widest validated range and correctly predicts friction factors for straight pipe flow $(De \to 0)$.

Figure 2.3 presents eight experimental heat transfer correlations for developed, Newtonian flow in curved pipes. $De$ is again made a function of the fluid viscosity implying extreme Prandtl numbers for low Dean numbers. In this range no correlation applies, though in industrial practice these conditions are entirely feasible.

Contrary to friction factors, relationships for developed heat transfer yield a wide range of heat transfer coefficients under identical process conditions. Reasons for this variety are different types of thermal boundary condition (constant wall temperature and two types of constant heat flux, denoted by $T$, $H_1$ and $H_2$ respectively, Shah and London, 1978), various definitions of
the temperature used to evaluate fluid properties and the fact that local heat transfer coefficients are derived from a number of other measured quantities, each with its measuring inaccuracy. Another explanation may be an insufficient thermal development, as this requires 5 - 10 turns (please refer to section 2.4), despite the accelerating effect of curvature.

Two correlations due to Manlapaz and Churchill, one for constant wall temperature and one for constant heat flux (6 and 7 respectively in figure 2.3), were derived by combining a perturbation solution for low Dean numbers with boundary layer flow asymptotes at high Dean numbers, and the assumption of an intermediate asymptote at moderate $De$. The coefficients of these composite correlations were fit to experimental data and to numerical results. Though Dravid, Smith, Merrill, and Brian (1971) questioned the independence of the heat transfer on Prandtl number for high Dean numbers, Manpalaz’ correlations are recommended by Shah and Joshi (1987) and Ebadian and Dong (1998), and will be used as the reference heat transfer correlations throughout this thesis.

In the remainder of this chapter, developed and developing laminar flow and heat transfer for toroidal flow of constant property power-law fluids are simulated and validated. Helical flow and the impact of variable fluid properties are briefly discussed.
2.2 Equations and numerical considerations

The coordinate system for a curved pipe of zero pitch (torus) is given in Figure 2.4 (left). For arbitrary curvature ratio, the equations of continuity, motion and energy in a toroidal coordinate system can be found in e.g. Berger, Talbot, and Yao (1983).

\[ \nabla \cdot \mathbf{v} = 0 \]  \hspace{1cm} (2.5)

\[ \rho \left( \frac{d\mathbf{v}}{dt} + \left( \mathbf{v} \cdot \nabla \right) \mathbf{v} \right) = -\nabla p + \mu \nabla ^2 \mathbf{v} + \mathbf{F} \]  \hspace{1cm} (2.6)

\[ \rho c_p \left( \frac{dT}{dt} + \mathbf{v} \cdot \nabla T \right) = \lambda \nabla ^2 T \]  \hspace{1cm} (2.7)

For steady flow the time-dependent terms in the momentum and energy equations are dropped. Body forces enter the model through the term \( \mathbf{F} \) in the equations of motion. The effect of curvature on the flow and temperature field for example is included by term \( F_c \) (equation 2.8) acting in the x-direction as shown in Figure 2.4 (right).
\[ F_c = \rho \frac{w^2}{x} \]  

(2.8)

Using a centrifugal momentum source term and a straight pipe to model flow and temperature fields in a curved pipe disregards tertiary Coriolis forces in the pipe cross-section and the fact that streamlines are shorter on the inside than on the outside of the curved tube. However, Bauke and Brauer (1979) state that developed friction factors and Nusselt numbers are no longer a function of \( d/D \) if this ratio is smaller than 0.1. Shah and Joshi (1987) even recommend a value of 0.33. For such weakly curved pipes all terms involving \( d/D \) in the equations of motion in the toroidal coordinate system are of higher order, except the terms containing the centrifugal force. In appendix A it is shown that a weakly curved toroidal coordinate system is equivalent to a Cartesian system including a centrifugal force term as given by equation 2.8.

Numerical trials for power-law fluids over the entire range of process conditions for the current \( d/D = 0.025 \) confirmed this approach. They revealed a maximum difference of 0.17 % in the developed friction factor and 1.6 % in the developed Nusselt numbers between the curved and the straight formulation (table 2.1). All further flow simulations were therefore carried out using straight pipes and centrifugal body forces.

<table>
<thead>
<tr>
<th>( \Delta f_n ) (%)</th>
<th>( \Delta N_u ) (%)</th>
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<tr>
<td>( \Delta f_n ) (%)</td>
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<td>0.5</td>
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<td>0.05</td>
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Table 2.1: Change of \( f_n \) and \( N_u \) for 'straight' relative to curved formulation.

2.2.1 Boundary conditions

Different sets of boundaries were imposed for developed (table 2.2) and developing (table 2.3) flow. Developed flow and heat transfer were obtained on a short tube section using partially cyclic boundaries in the tube cross
section. On these boundaries, all velocity components on corresponding cell faces at the inlet and the exit of the tube were set equal, and temperature profiles were coupled by a fixed offset. At the wall, the no-slip condition was imposed, and the $H_1$ thermal boundary (axially constant heat flux, peripherally constant temperature (Shah and London, 1978)) was implemented using a constant temperature difference between the wall and the bulk. The $H_1$-boundary is a good approximation of the developed heat transfer in the counter-current regenerative sections of the heaters and coolers of a steriliser as the heat capacities of both streams are approximately equal.

To obtain the $H_1$-boundary, the temperature difference between the wall and the bulk was kept constant. As the heat transfer rate and the axial gradient of the bulk temperature are not known a priori, establishing the required axial gradient of the wall temperature is an iterative process. Fortunately, the axial temperature rise was very small compared to the temperature difference between the wall and the bulk, and axial gradients of the wall temperature calculated using the reference correlation of Manlapaz and Churchill (1981) were satisfactory.

For developing flow and heat transfer simulations, a developed power-law velocity profile and a uniform temperature were adopted at the inlet. At the tube wall, the no-slip condition and the $T$-boundary were applied. At the outlet, static pressure as well as all axial gradients were set to zero. For Newtonian ($n = 1$), straight ($Da = 0$) pipe-flow these boundaries amount to the Graetz-Nusselt problem. The constant temperature boundary approximates heat transfer by condensing steam at the walls of the steam heaters of a steriliser.

### 2.2.2 Fluid properties

The rheology is described by the Ostwald-de Waele or power-law model (Bird, Stewart, and Lightfoot, 1960). For 3-dimensional flows, the apparent viscos-

<table>
<thead>
<tr>
<th>inlet</th>
<th>wall</th>
<th>outlet</th>
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<tr>
<td>$\varrho_{\text{in}}(r, \theta) = \varrho_{\text{out}}(r, \theta)$</td>
<td>$\varrho = \bar{\varrho}$</td>
<td>$\varrho_{\text{out}}(r, \theta) = \varrho_{\text{in}}(r, \theta)$</td>
</tr>
<tr>
<td>$\bar{T} = \text{const.}$</td>
<td>$q(z) = \text{const.}$</td>
<td>-</td>
</tr>
<tr>
<td>$T_{\text{in}}(r, \theta) = T_{\text{out}}(r, \theta) - \Delta T$</td>
<td>$T(r) = \text{const.}$</td>
<td>$T_{\text{out}}(r, \theta) = T_{\text{in}}(r, \theta) + \Delta T$</td>
</tr>
</tbody>
</table>

Table 2.2: Boundaries used in the developed simulations.
ity $\mu$ depends on power-law index $n$, consistency index $m$, and the magnitude of the rate of strain tensor $\Delta_{ij}$ (2.9).

$$\mu = m \left| \frac{1}{2} \sum \sum \Delta_{ij} \Delta_{ij} \right|^{\frac{m+1}{2}}$$  

(2.9)

For the developed simulations, $d/D$ was fixed and $De$ was set changing $m$, implying a varying Prandtl number. This approach is realistic, as viscosity is very sensitive to temperature, especially in the case of gelatinisation (please refer to chapter 3).

For developing flow, $m$ was given two constant values over the range $De = 0 - 100$ and for $De = 500 & 1000$. The Dean number itself was varied using the coil diameter $D$. The Prandtl number was kept constant ($Pr = 104.5$) adjusting the fluid conductivity ($\lambda = 0.5$ W/mK for $De = 0 - 100$; $\lambda = 0.1$ W/mK for $De = 500 & 1000$). This seemingly cumbersome approach was necessitated by the wide range of Dean numbers investigated. Straight flow ($De = 0$) requires a minimum viscosity to ensure laminar flow, whereas strongly curved flows ($De > 100$) cannot be obtained without violating the loosely coiled assumption ($d/D \leq 0.1$) using this viscosity. A fivefold decrease in viscosity was therefore introduced for the two highest Dean numbers.

Density $\rho = 1000$ kg/m$^3$ and heat capacity $c_p = 4180$ J/(kg °C) were constant. The impact of temperature dependent fluid properties is discussed in section 2.6.

### 2.2.3 Solution strategy

Solutions to equations 2.5 - 2.7 have been approximated using the Star finite volume package (CD Ltd., 2001). The SIMPLE algorithm was employed to decouple the equations after discretisation, which was performed using a second order accurate scheme (please refer to appendix B). The sets of

<table>
<thead>
<tr>
<th>inlet</th>
<th>wall</th>
<th>outlet</th>
</tr>
</thead>
<tbody>
<tr>
<td>$w = \overline{w}^{\frac{3n+1}{n+1}} \left( 1 + \frac{2n}{d} \frac{\Delta_{ij}}{\Delta_{ij}} \right)$</td>
<td>$v = 0$</td>
<td>$\partial w/\partial z = 0$</td>
</tr>
<tr>
<td>$T = \text{const.}$</td>
<td>$T = \text{const.}$</td>
<td>$\partial T/\partial z = 0$</td>
</tr>
</tbody>
</table>

Table 2.3: Boundaries used in the developing simulations.
algebraic equations for each dependent variable were solved using a conjugate gradient type solver and various preconditioning techniques (CD Ltd., 2001). To promote convergence whilst preserving stability, problem and variable specific under-relaxation factors were employed.

Solutions were considered converged when all variables in a cell at the maximum axial velocity had become constant at double machine precision, and the relevant sums of normalised residuals $R_\phi$ had reached a small value (typically $10^{-7}$).

\[ R_\phi = \frac{\sum |r_\phi|}{M_\phi} \] (2.10)

In equation 2.10, the cell residual $r_\phi$ is the local imbalance in the finite volume transport equation of variable $\phi$ (e.g. $u, v, w, T$), resulting from the incomplete solution. Summation of $r_\phi$ over all computational cells, and division by the inflow rate $M_\phi$, yields $R_\phi$.

The numerical procedure in the case of unsteady computations is outlined in appendix C.

### 2.2.4 Mesh and discretisation for developed flow

The independence of the numerical solutions on mesh and discretisation scheme was established over the entire range of flow conditions and fluid properties. For five successively refined butterfly meshes, four Dean numbers (0, 10, 100 and 1000) and three power-law indices ($n = 0.2, 1$ and 2) the developed normalised friction factor $f_n$ and the developed Nusselt number $Nu$ were computed. The diffusive fluxes were discretised using central differencing, whereas the convective fluxes were discretised employing a first, a second and a third order accurate scheme. For detailed results on these 180 cases, and a more extensive discussion of the mesh structure and the schemes employed, the reader is referred to appendix B. As an example, the mesh dependence of $f_n$ and $Nu$ of a Newtonian fluid ($n = 1$) using the second order MARS scheme is shown in table 2.4.

For engineering purposes the developed simulations are considered independent for the $r \times \theta = 35 \times 80$ mesh. In table 2.5, the three discretisation schemes are compared for this mesh. It is clear that the first order Upwind scheme (UD) may be satisfactory for flow computations, but it fails to yield the analytical straight pipe flow heat transfer, and overestimates heat transfer at all other Dean numbers. The second order MARS scheme and third
<table>
<thead>
<tr>
<th>mesh</th>
<th>$f_n$</th>
<th>$Nu$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$r \times \theta$</td>
<td>$De$</td>
<td>$De$</td>
</tr>
<tr>
<td>23 x 32</td>
<td>1.009</td>
<td>1.012</td>
</tr>
<tr>
<td>27 x 64</td>
<td>1.002</td>
<td>1.005</td>
</tr>
<tr>
<td>35 x 80</td>
<td>1.001</td>
<td>1.005</td>
</tr>
<tr>
<td>54 x 128</td>
<td>1.001</td>
<td>1.004</td>
</tr>
<tr>
<td>70 x 160</td>
<td>1.001</td>
<td>1.005</td>
</tr>
<tr>
<td>anal.</td>
<td>1</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 2.4: Dependence of $f_n$ and $Nu$ on the mesh for MARS and $n = 1$.

order QUICK perform satisfactorily, with the QUICK scheme approaching independence slightly faster due to its higher order.

<table>
<thead>
<tr>
<th>$f_n$</th>
<th>$Nu$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$r \times \theta$</td>
<td>$De$</td>
</tr>
<tr>
<td>UD</td>
<td>1.006</td>
</tr>
<tr>
<td>MARS</td>
<td>1.001</td>
</tr>
<tr>
<td>QUICK</td>
<td>1.002</td>
</tr>
<tr>
<td>anal.</td>
<td>1</td>
</tr>
</tbody>
</table>

Table 2.5: Dependence of $f_n$ and $Nu$ on scheme for $n = 1$ and $r \times \theta = 35 \times 80$.

It should be remarked that all meshes possess a limited radial refinement from the tube axis to the wall (radial dimension of bulk cell only 1.5 times that of wall cell). High radial refinement ratios were found to be detrimental to the solutions accuracy, probably as a result of excessive radial-tangential aspect ratios near the wall. In rheologically complex flows (chapter 4), the highest velocity and temperature gradients do not necessarily occur at the pipe wall, and equidistant discretisation is in fact desirable.

2.2.5 Mesh and discretisation for developing flow

For Newtonian flow, three meshes having 50, 100 and 200 unequally spaced axial cells were tested for the MARS and QUICK scheme. A detailed comparison of the developing friction factors and Nusselt numbers can be found in appendix B. As shown in figure 2.5, both MARS and QUICK yield independent solutions for 100 axial cells. However, the QUICK scheme produces
Figure 2.5: Developing heat transfer for MARS and QUICK, 100 axial cells.

...minor temperature undershoots in the entrance of the tube, and the MARS scheme is therefore favoured.

2.3 Developed flow and heat transfer

Developed velocity and temperature fields for a heated constant property power-law fluid were calculated for $De = 0 - 1000$ and $n = 0.2 - 2$ using the $r \times \theta = 35 \times 80$ mesh established in section 2.2.4.

Figure 2.6 shows the developed axial velocities over the range of curvature and rheological properties. For straight pipe flow ($De = 0$), the numerical solution reproduces the analytical axial velocity field perfectly. Newtonian flow ($n = 1$) under increasing curvature ($De > 10$) yields the distorted axial velocity distribution experimentally observed by e.g. Adler (1934). As confirmed by laser Doppler anemometry (Ranade and Ulbrecht, 1983), increasing $De$ and increasing pseudoplasticity ($n < 1$) both serve to flatten the axial velocity profile. Curvature and pseudoplasticity also increase the axial velocity gradients at the outside of the curved tube, while decreasing those on the inside.

All dilatant flows ($n > 1$) fail to converge to a steady solution for $De > 250$. As explained in appendix C, this may be due to a physical instability of the flow. The axial velocity profiles shown in figure 2.6 are therefore snapshots of the flow obtained using the transient procedure outlined in appendix C.
Figure 2.6: Axial velocity for curved developed flow, $\overline{w} = 0.5 \text{ m/s}$.
Figure 2.7: Magnitude of secondary velocity $v_s = \sqrt{v^2 + v_0^2}$. 
Figure 2.8: Cross-section temperature, $\overline{T} = 90^\circ C, T_w = 100^\circ C.$
Figure 2.7 presents the magnitude of the secondary velocity \( v_s \). In straight pipe flow, centrifugal forces are absent and no secondary velocity exists. As curvature increases, the intensity of the secondary flow passes through a maximum (Bauernfeind and Brauer, 1979), ultimately yielding the boundary layer flow regime for \( De > 100 \). In boundary layer flow, the secondary flow from the inside to the outside of the curved pipe approaches uniformity in the core of the flow, whereas the secondary flow from the outside to the inside is confined to two thin layers at the upper and lower wall of the tube.

Power-law rheology affects the secondary flow in two ways. First, pseudoplasticity flattens the axial velocity field, decreasing differences in the centrifugal forces and hence the strength of the secondary flow. Second, pseudoplasticity increases axial velocity gradients at the wall on the outside of the curved tube, locally lowering the apparent viscosity. Friction experienced by the secondary flow is reduced, and the maximum of the secondary velocity moves closer to the wall, decreasing the thickness of the boundary layer on the outside of the curved tube. Dilatancy on the other hand increases the intensity of the secondary flow and decreases the gradient of the secondary velocity in the boundary layer. These phenomena agree with the numerical work of Hsu and Patankar (1982) and Agrawal, Srivastava, Jayaraman, and Nigam (1994), and with the experimental observations of Ranade and Ulbrecht (1983).

Figure 2.8 shows the temperature distribution in the cross-section. For straight pipe flow the expected temperature minimum at the centerline is found. If the tube is curved and for a sufficiently high Prandtl number, this minimum is replaced by two minima roughly coinciding with the centers of the Dean vortices. As \( De \) increases, the center of each vortex moves closer to the pipe wall and the temperature gradients at the wall increase.

The impact of rheology can be similarly explained: pseudoplasticity decreases the thickness of the boundary layer and moves the center of the vortices closer to the wall. Again the distance between the temperature minima and the wall is reduced and the temperature gradients increase. The effect of dilatancy is opposite.

The peripherally averaged normalised friction factor \( f_n \) (figure 2.9) is calculated from the peripherally averaged shear stress at the wall. For curvature up to \( De = 10 \), the increase in shear stress at the outside of the curved tube is balanced by the decrease in shear stress at the inside, and the friction factors for straight and curved flow are identical (\( f_n = 1 \)). As curvature further increases, the local friction factor at the outside continues to rise, while
keeping approximately constant at the inside. The friction factor in curved flow is significantly higher than in straight flow \((f_n > 1)\), and agrees well with the reference correlation for Newtonian flow due to Mishra and Gupta (1979a).

Pseudoplasticity lowers the apparent viscosity in the high shear region at the outside of the curved tube, decreasing local wall shear stresses and averaged friction factors for \(De > 10\). Contrarily, dilatancy strongly increases \(f_n\). Few experimental correlations for friction factors in curved power-law flow are available, and most of these do not express the effects of the Dean number and the power-law index separately. Those that do (Mashelkar and Devarajan, 1976; Mujawar and Rao, 1978; Mishra and Gupta, 1979b) are only valid over a very small range of indices, and yield erroneous results for smaller and larger values of \(n\). However, these correlations qualitatively agree within their ranges with the current numerical results, and the predicted impact of \(n\) on \(f_n\) at fixed Dean number is confirmed by the numerical studies of Hsu and Patankar (1982), Takami, Sudou, and Tomita (1986) and Agrawal, Srivastava, Jayaraman, and Nigam (1994).

For \(De > 250\) and \(n > 1\) the flow seems unsteady, and simulations were carried out on the full cross-section of the pipe using a transient algorithm (Issa, 1986). The values for \(f_n\) and \(Nu\) are therefore time-averaged over one oscillation (appendix C). No significant impact of the unsteadiness on the friction factor was found.
Newtonian curved heat transfer agrees satisfactorily with the reference correlation of Manlapaz and Churchill (1981) (figure 2.10). For intermediate Dean numbers a plateau value (termed intermediate asymptote by Manlapaz and Churchill) for the Nusselt number is apparent. The relative independence of heat transfer on curvature at intermediate Dean numbers can be understood from the shape of the Dean vortices and the resulting temperature profiles. As long as the Dean vortices remain oval in shape, the location of the temperature minima and the magnitude of the temperature gradients hardly change. Only when curvature is sufficiently strong to induce a boundary layer flow ($De > 100$), the thickness of the boundary layer decreases with curvature, and heat transfer is proportional to the square root of the Dean number (Mori and Nakayama, 1965, 1967).

Concerning curved developed power-law heat transfer, only numerical results of Hsu and Patankar (1982), and an analytical boundary layer approximation by Kawase and Moo-Young (1987) express the separate effects of $De$ and $n$. At first inspection, the impact of rheology on heat transfer found by Hsu and Patankar (1982) seems to contradict the present results. However, these authors use a different definition of the Prandtl number, and taking this into account, a positive correlation between heat transfer and pseudoplasticity is revealed here as well. Dilatancy on the other hand lowers the value of the Nusselt numbers at fixed $Pr$ and $De$, as was confirmed for the boundary layer flow regime ($De > 100$) by Kawase and Moo-Young (1987).
The values of $Nu$ are time-averaged for $De > 250$ and $n > 1$ (appendix C), and heat transfer proved significantly higher in oscillatory flow. Unfortunately no experimental verification is available. Simon, Chang, and Chow (1977) performed a perturbation analysis for small Dean number flows and found an increase in heat transfer, but Rabadi, Chow, and Simon (1982) later discredited these findings in their report on numerical simulations of insta-


tory high Dean number flows. However, these studies are fundamentally different from the current work as they concern Newtonian flow having an oscillating mass flux.

2.4 Developing flow and heat transfer

Developing velocity and temperature fields for a heated, constant property power-law fluid were calculated for five Dean numbers (0, 10, 100, 500 and 1000) and three power-law indices ($n = 0.2$, 1 and 2). As in section 2.3, the $r \times \theta = 35 \times 80$ mesh was used for the cross-sectional discretisation. In the axial direction 100 cells were used, and for the first $1/10$ of the pipe the axial cell length increased according to a fixed ratio.

Figure 2.11 shows the developing axial velocity, secondary flow and the temperature field in strongly curved Newtonian flow. As the developed straight pipe flow enters the pipe, the unbalanced centrifugal forces accelerate the fast moving fluid outwardly. A strong secondary displacement flow is induced, which is capable of transporting fluid of a high axial momentum from the outside of the curved pipe, along the pipe wall, back to the inside of the pipe. For sufficiently strong curvature, this gives rise to a dou-

bly peaked axial velocity profile along the the line of symmetry (Austin and Seader (1974); Takami, Sudou, and Tomita (1990); figure 2.11, $z/d = 12$).

Further down the pipe, the balance between the centrifugal forces and the viscous forces in the secondary flow is established, and fluid of high axial mo-

mentum that was transported to the inside of the curved pipe, is moved back to the outside. The secondary flow field passes through a 6-vortex pattern (figure 2.11, $z/d = 20$) and the flow develops into the weaker Dean vortices.

Temperature fields develop much slower than the axial and secondary veloc-

ity. Because of the secondary flow reversal following the displacement flow, little fluid is initially transported from the hot wall into the cold core, and the double temperature minimum is established only beyond $z/d = 400$. 

26
Figure 2.11: From top to bottom: developing axial velocity, secondary flow vectors and temperature field as a function of $z/d$ ($De = 500$).

Figure 2.12: Axial velocity, secondary flow vectors and temperature for curved developed bifurcating flow, $z/d = 400$ ($De = 1000$).

For extreme curvature ($De > 800$), the 6-vortex flow develops into the 4-vortex pattern of figure 2.12 when a plane of symmetry is used. Irrespective of discretisation scheme or axial mesh density, restarts from a 2-vortex flow at $De = 800$ eventually branched into the 4-vortex type.

Some controversy exists in the literature whether such bifurcations have a physical basis, or whether they are numerical artefacts. The 4-vortex flows were first reported in numerical studies of Dennis and Ng (1982) and Nandakumar and Masliyah (1982), but were later denounced by Liu, Acafan,
Nasr-El-Din, and Masliyah (1994) as being unphysical. Though flow visualisation experiments by Cheng, Inaba, and Akiyama (1985) confirmed the existence of additional 4-vortex solutions after a 180 degree turn, Daskopoulos and Lenhoff (1989) attributed the occurrence of such numerical solutions to the use of planar symmetry. Goering and Humphrey (1993) numerically found 4-vortex solutions on the full cross-section to be unstable to asymmetric perturbations, as they eventually decayed to the 2-vortex pattern.

Current simulations using the full cross-section of the tube for $De = 1000$ indeed yielded only 2-vortex solutions. Incomplete convergence of the steady simulations prompted the use of the transient approach as outlined in appendix C. Some unsteadiness was apparent on the outside of the curved tube in the hydrodynamic entrance region. This unsteadiness is probably not related to the travelling wave instability in curved tube flow at Reynolds numbers close to the transition to turbulence, as the unsteadiness does not occur on the inside of the coil.

Webster and Humphrey (1997) pointed out that secondary centrifugal forces may give rise to a periodic, laminar unsteadiness on the inside of the coil, the so-called travelling wave instability. For very high Dean numbers, the boundary layer flows collide on the inside of the coil and sharply turn to form a planar jet in the plane of symmetry. A combination of high secondary flows and this sharp turn may produce secondary centrifugal forces strong enough to dominate the local force balance. An increase of the secondary flow increases these centrifugal forces, which create a local positive pressure gradient encountered by the secondary flow. As a result the secondary circulation is inhibited, and the secondary centrifugal forces decrease.

Pseudoplasticity and dilatancy do not qualitatively change the development of the velocity and temperature fields. Velocity and temperature fields in the developed part of the pipe were very similar to the developed fields shown in figures 2.6 to 2.8. Due to the unsteady nature of the flow, no solutions could be obtained for dilatant flow when the Dean number exceeded 250.

The peripherally averaged developing normalised friction factor $f_n(\zeta)$ is given in figure 2.13. In the inlet of the tube, the hydrodynamic development causes the friction factor to oscillate, except in the case of pseudoplastic flow ($n = 0.2$). So far, this oscillation has only been demonstrated numerically (Lin, Zhang, and Ebadian, 1997), but its existence seems plausible given the dramatic evolution of the axial velocity in the developing region. As corroborated by the measurements of Austin and Seader (1974), increasing curvature decreases the hydrodynamic entrance length. Further down the curved tube the normalised friction factors are of course equal to the developed friction factors shown in figure 2.9.
Hydrodynamically developed, thermally developing Newtonian heat transfer in a straight tube (the Graetz-Nusselt problem), agrees excellently with the analytical results presented by Shah and London (1978) as the numerical and the analytical values coincide (figure 2.14, $De = 0$). Correction of these analytical results for power-law rheology using the method employed by e.g. Metzner, Vaughn, and Houghton (1957), yielded an excellent agreement with the numerical results, except for developed dilatant heat transfer.

As experimentally observed by Dravid et al. (1971) and Janssen and Hoogendoorn (1978), heat transfer fluctuates in the entrance of a curved pipe. This is a result of the alternating exposure of the wall to hot and cold fluid as the flow thermally develops. Thermal development length is shortened by curvature (Dravid, Smith, Merrill, and Brian, 1971; Janssen and Hoogendoorn, 1978; Xin and Ebadian, 1997), but expressed in the dimensional axial coordinate $z$, it is still two orders of magnitude longer than the hydrodynamic entrance length, which is due to the high Prandtl number ($Pr = 104.5$, please refer to section 2.2.2). Care should therefore be taken in applying developed heat transfer correlations to coils having less than 5 - 10 turns. Incomplete thermal development may explain the differences between the experimental heat transfer correlations in figure 2.3.

Pseudoplasticity decreases the strength of the secondary flow in the inlet and in the developed region, resulting in dampened oscillations of the Nusselt number. For $De = 10$ and $n = 0.2$ the secondary flow has become so weak,
Figure 2.14: Developing heat transfer in curved power-law flow.

that the first oscillation only occurs near the end of the section, and thermal development is actually retarded.

In table 2.6, the asymptotic Newtonian Nusselt numbers are compared to the reference correlation for the $T$-boundary of Manlapaz and Churchill (1981). The agreement is good, except for the largest curvature ($De = 1000$). According to Dravid et al. (1971), heat transfer is a monotonically ascending function of the Prandtl number, also at high Dean numbers. As the reference correlation assumes a Prandtl number independence in strongly curved flows, it is likely to underestimate heat transfer for high Prandtl number flows. Four vortex flows exhibit an even higher heat transfer, but this is the result of a qualitatively different temperature field.

<table>
<thead>
<tr>
<th>$De$</th>
<th>0</th>
<th>10</th>
<th>100</th>
<th>500</th>
<th>1000</th>
<th>1000</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>numerical</td>
<td>3.66</td>
<td>7.62</td>
<td>11.2</td>
<td>24.6</td>
<td>45.0</td>
<td>55.0</td>
</tr>
<tr>
<td>experimental</td>
<td>3.66</td>
<td>7.52</td>
<td>11.8</td>
<td>23.7</td>
<td>33.3</td>
<td>33.3</td>
</tr>
</tbody>
</table>

Table 2.6: Developed Newtonian Nusselt numbers for the $T$-boundary, $Pr = 104.5$. 
2.5 Helical flow

If the angular length $\psi$ exceeds $2 \cdot \pi$, the toroidal pipe turns into a helical coil ascending with a coil pitch $p' \geq d$ every full turn. Pitch introduces an additional torsional force in the $\theta$-direction in the cross-section. Axial velocity and temperature fields are rotated with respect to the pipe axis, and the symmetry of the Dean vortices is broken (Liu and Masliyah, 1994; Yang, Dong, and Ebadian, 1995).

However, the effect of pitch on pressure drop and heat transfer can usually be neglected for practical purposes provided the Dean number is replaced by the Helical number $He$ (please refer to the list of symbols). Experimental work of Srinavasan, Nandapurkar, and Holland (1970) and Mishra and Gupta (1979a) shows that pitch only significantly affects the developed friction factors when $p' > D$. In numerical work, Yang, Dong, and Ebadian (1995) reported a decrease up to 10% in the developed Nusselt number for $p' = 0.15 \cdot D$, but in later experimental work, Xin and Ebadian (1997) could find no apparent pitch effects on heat transfer.

Having a pitch on the order of $p' = 0.05 \cdot D$, the effect of the torsional force is insignificant for the Stork Sterideal. However, as the torsional force is essentially a Coriolis force, it can be incorporated into the body force formulation of the curved pipe.

2.6 Variable fluid properties

For all real fluids, properties are functions of the temperature. In steady, incompressible flow without reactions, $c_p$, $\lambda$ and $\rho$ are functions of the local temperature only. The dependence on $T$ is usually not very strong, and it can be incorporated using simple polynomial relationships.

Next to its dependence on shear rate, the apparent viscosity usually exhibits a strong dependence on temperature, which affects both flow and heat transfer. In friction factor and heat transfer relationships for closed channel flows, the property ratio method (Kakaç, 1987) can be employed to allow for variations in viscosity. Andrade and Zarpoli (2001) have numerically shown that this method also yields reasonable results for developed toroidal flows. Of course such corrections are implied by the 3-dimensional numerical approach if the viscosity is a function of temperature.

For complex fluids such as gelatinising suspensions, rheological properties are also a function of time, as gelatinisation and shear destruction increase and decrease viscosity in the course of heating, holding and cooling. Chapter 3 is devoted entirely to the modelling of gelatinising suspensions.
When density is a function of temperature, buoyancy forces may give rise to secondary Morton (Morton, 1959) or mixed secondary vortices (figure 5.3). Additional buoyancy forces in curved flow further increase frictional losses (Goering, Humphrey, and Greif, 1997) and heat transfer (Abul-Hamayel and Bell, 1979). As is shown in appendix D and in chapter 5, flow and heat transfer in coiled sterilisers are clearly dominated by centrifugal effects and for practical purposes buoyancy forces may be neglected.

2.7 Conclusions

A brief review of the literature shows that curvature can have a profound impact on flow, heat- and mass-transfer, residence time distribution and on the transition to turbulent flow. Whereas friction factor relationships generally agree well, heat transfer correlations may yield predictions differing by a factor of three. Most correlations concern Newtonian flow, and little experimental data is available for power-law fluids.

The commercially available Star package was used to simulate flow and heat transfer of developed and developing power-law flows in moderately curved tubes. For developed flow and the $H_1$-boundary, flow fields strongly resemble those presented in the literature, and the predicted friction factors and Nusselt numbers agree well with two experimental reference correlations. In developing flow for the $T$-boundary, the oscillatory behaviour of the friction factor and the heat transfer are confirmed, and friction factors and heat transfer for long tube lengths agree well with the developed reference correlations. Pseudoplasticity was shown to further improve heat transfer while reducing frictional losses, and dilatant flow proved unsteady for high Dean numbers.

Pitch is unimportant in the Stork coiled steriliser and buoyancy effects can be accounted for using a partial Boussinesq approximation. However, buoyancy proved not relevant in the case of the heat exchangers under consideration.
Bibliography


Y. Mori and W. Nakayama. Study on forced convective heat transfer in curved


Chapter 3
Rheology of starch suspensions

Gelatinisation and gelation cause dramatic changes in the rheological properties of dilute starch suspensions. The viscosity may rise several orders of magnitude, and upon cooling solid-like gels are prone to form. Two models describing the developing viscosity during gelatinisation were critically assessed. A model due to Morgan, Steffe, and Ofoke was shown to be comprehensive, as it incorporates both instantaneous and history effects of the shear rate and temperature applied during pasting. A second, more practical model due to Yang and Rao requires less extensive rheometry, but essentially regards gelatinisation to be at equilibrium, as kinetics of gelatinisation and shear destruction are omitted.

We adapted Yang’s model to yield a non-equilibrium model of the shear-, temperature- and time dependent rheology of starch suspensions. We introduce the concept of gelatinisation classes to enable incorporation of gelatinisation-lag due to high heating rates, and shear destruction resulting from high shear stresses. The immediate shear dependency was made to conform to power-law rheology above a critical level of shear. Thus, we obtained a mathematical model of the apparent viscosity of gelatinising starch suspensions suitable for flow and heat transfer simulations under industrial processing conditions.
Figure 3.1: Rice (top left), corn (top right), wheat (bottom left) and potato (bottom right) granules 180×. Reproduced from Fitt and Snyder (1984).

### 3.1 Starch suspensions

Starch is widely used as a thickening agent to increase the viscosity and to contribute to the texture of liquid foods such as sauces, soups and desserts. For this application, the amount of starch added is usually in the 2 - 5% w/w range. Further discussions therefore apply to dilute starch systems in which water is present in large excess. Doughs and other low moisture pastes will not be treated.

#### 3.1.1 Starch granules

Starch is extracted from a variety of plants in which sugars are stored in granules sizing 2 - 100 μm (figure 3.1). Starch granules primarily consist of a mixture of linear (amylose) and branched (amylopectine, figure 3.2) polysaccharides made up of 1000 to 100000 anhydroglucose units. Typically starch granules contain 20% amylose by weight, though high amylopectine
(‘waxy’, >95% w/w amylpectine) and high amyllose (e.g. amylomaize, 50 - 70% w/w amyllose) varieties are available (Zobel, H.F., 1984; Morris, V.J., 1986; Blanshard, 1987; Sivak and Preiss, 1998).

Starting at a growth nucleus, or hilum, amyllose and amylpectine are deposited concurrently in concentric rings on the surface of the starch granule. According to a recent model (figure 3.3) put forward by Cameron and Donald (1993) and Jenkins and Donald (1998), these rings are alternatingly semi-crystalline and amorphous, possibly corresponding to a diurnal growth cycle. The semi-crystalline rings in turn consist of crystalline and amorphous filaments associated with the straight and branching sections respectively of the amylpectine molecules. Amylose is primarily located in the amorphous regions (Blanshard, 1987). Atkin, Abeyseker, and Robards (1998b) state that the surface layer of starch granules constitutes a special type of amylpectine having a higher lipid content. In some starches, channels running from the surface to the hilum can be observed, increasing the accessibility of the granule interior (Huber and BeMiller, 2000). The partial crystallinity of starch granules is reflected in the appearance of an X-shaped pattern under polarised light, termed birefringence (figure 3.4).
3.1.2 Gelatinising suspensions

When suspended in water at temperatures below 55 °C, a starch granule imbibes a limited amount of water and its diameter increases by 10 - 20%. Depending on damage sustained during starch extraction, a small amount of amylose leaches from the granule into the aqueous phase. For suspensions below 10% on a w/w basis, the increase in viscosity on cold swelling is usually small (Dengate, 1984).

As the starch granule is heated above a threshold temperature $T_g$, an irreversible process termed gelatinisation takes place. Water enters the amorphous rings and disrupts the intermolecular hydrogen bonds between the polysaccharide chains, increasing stress on the semi-crystalline rings. On
further temperature increase, the stresses associated with swelling break and aid in solvating the amorphous and crystalline filaments as well (Atkin, Abeysekera, Cheng, and Robards, 1998a; Jenkins and Donald, 1998). All birefringence is lost and a strong uptake of water causes the granule to swell to several times its original diameter. At maximum swelling, the surface layer of amylpectin serves as a bag containing a solution of solvated amyllose and partially entangled amylpectin molecules (Atkin et al., 1998b).

The average value of $T_g$ for a population of native granules is usually around 60°C, but differences in composition, internal structure (crystallinity), shape, size, pre-treatment and damage lead to a gelatinisation range of 5 - 15°C between individual granules (Lund, 1984; Liu and Lelievre, 1993). For low heating rates the extent of gelatinisation depends on the temperature only, and the partially gelatinised suspension can be considered at equilibrium (Blanshard, 1987).

Prolonged exposure to temperatures above 70°C and the application of shear cause the swollen granule to rupture and release its content into the aqueous phase. The amylpectin surface layer then collapses into a ghost remnant which eventually dissolves, yielding a molecular polysaccharide solution (Atkin, Abeysekera, and Robards, 1998b).

Over the course of gelatinisation, dramatic changes in the rheology of the suspension occur. The apparent viscosity increases several orders of magnitude as the granules swell and amyllose exuded from the granules forms a filamentous network in the aqueous phase (Miller, Derby, and Trimbo, 1973). On reaching a peak, the viscosity decreases again as the paste structure disintegrates and the remaining ghosts solubilise. As the suspension cools, a gel-like network starts forming in the aqueous phase and the deformability of the (remnants) of the swollen granules diminishes, increasing the over-all viscosity. The developing viscosity during heating and cooling in a low shear environment is shown in figure 3.5.

### 3.1.3 Gelling and recrystallisation

Fully gelatinised starch suspensions consist of swollen and ruptured granules suspended in a macromolecular solution of amyllose. Depending on starch type and pasting procedure, some amyllose remains in the granules, and amylpectin may co-leach into the continuous phase. When cooled, amyllose solutions tend to form elastic gels, which increase in crystallinity over time (Morris, V.J., 1986; Lii, Tsai, and Tseng, 1996). Amylose gelation of gelatinised suspensions starts on cooling, and concentrations exceeding 1 - 1.5% w/w suffice to trap all remaining water. Amylose solutions gel irreversibly
Figure 3.5: Apparent viscosity of 6% w/w corn starch suspensions in Brabender units during a typical pasting cycle, time given in minutes. Comparison of native, moderately (1 cross-link per 440 anhydroglucose units) and highly cross-linked (1 cross-link per 440 AGU) modifications. Reproduced from Wurzburg (1986).

and very high temperatures are required for resolubilisation (Morris, V.J., 1986; Okechukwu and Rao, 1998).

On prolonged and cool storage, the amylopectine of the granular ghosts also partly recrystallises, and the side chains of the amylopectine molecules in different granules may associate to produce thermo-reversible gels. Both processes are very slow when compared to amylose gelation. Gelation of amylopectine requires concentrations exceeding 10% w/w to occur (Durrani and Donald, 1995; Okechukwu and Rao, 1998).

Starch gels can be pictured as 3-dimensional visco-elastic amylose networks reinforced by deformable granular amylopectine ghosts (Morris, V.J., 1986; Carnali and Zhou, 1996). The gel strength therefore depends on the volume fraction and rigidity of the swollen granules, and the presence and strength of the amylose network. Closely packed, cross-linked granules in an amylose matrix exhibit solid-like behaviour with a high storage modulus $G'$ and a low loss modulus $G''$ when tested in small strain oscillatory rheometry (please refer to appendix E). On the other hand, deformable, waxy starches behave liquid-like after gelatinisation and require a long and cool resting period to develop weak gels.
Temperature- and shear history of the gelatinisation govern the eventual granular strength and amylose distribution, and the same initial suspension therefore produces a range of gels or visco-elastic liquids for different pasting conditions (Doublier, Llamas, and Le Meur, 1987; Hårröd, 1989; Svegmark and Hermansson, 1990, 1991).

### 3.1.4 Other constituents

Gelatinisation and gelation properties of starch suspensions can be strongly influenced by other components, either native to the granule, or inserted into it during extraction or modification. Usually additional solutes such as sugars, salts, proteins and flavour components are added to the continuous phase to tailor the eventual product properties.

Generally, sugars and other non-ionic solutes increase the gelatinisation temperature, and inhibit the extent of gelatinisation through their effect on water activity in the swelling granules. The effect of salts varies from promotion to complete inhibition, which is probably related to the stabilising or destabilising action of the different ions on the hydrogen bonds in the granule (Lund, 1984; Blanshard, 1987).

Gelation and gel properties were also shown to depend on added flavours (Nuessli, Conde-Petit, Trommsdorff, and Escher, 1995), lactose and other skim milk components (Matser and Steeneken, 1997) and whey proteins (Mleko, 1997). Often specific gelation agents such as carrageenans, pectins and gums are added to the continuous phase to provide a required consistency at minimum starch consumption (Abdulmola, Hember, Richardson, and Morris, 1996; Mleko, 1997; Okechukwu and Rao, 1998).

Summarising, the number of possible food formulations involving starch is staggering, and their behaviour during pasting and gelation complex. However, the starch-water interaction is the determining factor in e.g. dilute starch based sauces (Thebaudin, A.C., and Doublier, 1998), and the work in this thesis is limited to pure starch-water systems.

### 3.1.5 Cross-linked starches

Native starches are usually insufficiently stable for use in the UHT-process as the high temperatures and shear stresses involved destroy the paste structure and most of the initial viscous increase is lost. Starch granules are therefore modified by chemical cross-linking of the polysaccharides (Wurzburg, 1986).
Each anhydroglucose unit of the amylose and amyllopectine molecules contains hydroxyl groups that may form chemical bonds with compounds capable of reacting with alcoholic hydroxyl groups. If a reagent is able to form two or more such bonds, a chemical cross-link may result between hydroxyls on different amylose or amyllopectine chains. Many agents can be employed for cross-linking, but in foods only adipic acetic mixed anhydrides and sodium trimetaphosphate are used. Soaking starch for several hours in dilute solutions below the gelatinisation temperature followed by washing, yields distarch adipates and distarch phosphates (Wurzburg, 1986).

After gelatinisation, cross-linking greatly increases stability of the granular structure under thermal and mechanical load (Wurzburg, 1986; Abdelrahim, Ramaswamy, and van de Voort, 1995; Tattiyakul and Rao, 2000). As the granules swell, the hydrogen bonds between the polysaccharide chains are broken, but the chemical cross-links prevent rupture and dissolution of the granules.

The effect of the cross-linking depends on the number of cross-links per anhydroglucose unit. Light cross-linking (e.g. corn starch having 1 cross-link per 1300 anhydroglucose units) yields a thermally stable starch of a great thickening power (figure 3.5). Higher temperatures and higher shear loads evidently require more cross-links for the granule to retain its integrity. Very strong cross-linking (e.g. 1 cross-link per 100 glucose units) severely inhibits swelling and greatly reduces thickening capability (Wurzburg, 1986).

3.2 Modelling gelatinising suspensions

As is clear from the previous section, the rheology of a starch suspension during heating, cooling and storage is a function of a great number of variables. The remainder of this chapter is dedicated to the development of a mathematical model of the apparent viscosity suitable for process simulations.

Two classes of models can be discerned: fundamental and phenomenological. Fundamental models attempt to predict the macroscopic rheology of a gelatinising suspension from the size, rigidity and volume fraction of the swelling granules, and from the properties of the aqueous phase. A simple example is the Einstein equation for the viscosity of dilute suspensions of rigid, monosized spheres (Launay, Dublier, and Cuvelier, 1986).

However, starch has a distinct size distribution and each granule has its unique, irregular shape. Due to differences in structure and composition of individual granules, a population of starch granules gelatinises over a temperature range. In the course of gelatinisation the granule size (distribution)
increases (Rao, Okechukwu, Da Silva, and Oliveira, 1997), the granules become increasingly deformable and amyllose (if present) leaches out of the granule. Eventually starch granules may rupture and solvate. Additionally, the rate of gelatinisation and amyllose network formation are influenced by the composition (sugar, salts, proteins) of the aqueous phase (Lund, 1984).

For a successful fundamental model, the gelatinisation properties of each granule in conjunction with its aqueous environment would have to be determined along with the interaction between the granules in varying state of gelatinisation. Construction of a fundamental model for gelatinising suspensions seems therefore unfeasible, if not impossible.

Phenomenological models correlate the rheology of a suspension to a limited number of key parameters e.g. shear rate, temperature and concentration. Such correlations may be derived from assumptions regarding the underlying physics and their coefficients are fit to macroscopic rheological data obtained from the gelatinising suspension. This approach implicitly includes the distributed characteristics of the specific starch and any interactions with other ingredients. On the downside, for every suspension data will have to be fit to the model and extensive and difficult rheological measurements may be required.

Phenomenological models expressing the rheology of gelatinising suspensions in units suitable for engineering calculations are rare (Lagarrigue and Alvarez, 2001). Traditionally, the Brabender amylograph is used to characterise starch suspensions. In the course of a pre-set temperature-time profile (pasting cycle, figure 3.5) the torque on an impeller is recorded as the starch suspension is rotated in a bowl. However, due to the complicated geometry of the impeller, shear rates are poorly defined and viscosity data is given in machine dependent units.

Two promising phenomenological gelatinisation models given in regular SI units are discussed in the next sections.

### 3.2.1 Morgan’s model

To model the apparent viscosity $\mu$ of starch suspensions in the course of a pasting cycle Dolan, Steffe, and Morgan (1989), and Dolan and Steffe (1990) applied a five term model originally developed for protein doughs (Morgan, Steffe, and Ofoli, 1989). They regarded shear rate $\gamma$, temperature $T$, starch concentration $c$, integral temperature history $\Psi$ and integral strain history $\Phi$ the independent variables in equation 3.1. The constants of this model were fit to data obtained from steady shear measurements.
\[ \mu(\gamma, T, c, \Psi, \Phi') = \mu_{ref} f(\gamma) f(T) f(c) f(\Psi) f(\Phi') \] (3.1)

In equation 3.1, \( \mu_{ref} \) is the consistency index of the ungelatinised and un-sheared suspension at reference temperature \( T_{ref} \) and reference concentration \( c_{ref} \). Equation 3.2 represents the immediate impact of shear rate on viscosity and is described by a power-law model, where \( n \) is the power-law index.

\[ f(\gamma) = \gamma^{n-1} \] (3.2)

An Arrhenius relationship accounts for the instantaneous temperature dependence with \( E_a \) the activation energy and \( R \) the universal gas constant (equation 3.3).

\[ f(T) = e^{\frac{E_a}{RT}} \] (3.3)

Dolan and Steffe (1990) modelled the dependency of viscosity on concentration using an exponential relationship in which \( c \) is the concentration compared to a reference concentration \( c_{ref} \) and \( b \) a constant (equation 3.4).

\[ f(c) = e^{b(c-c_{ref})} \] (3.4)

Equation 3.5 reflects the impact of temperature history on the apparent viscosity.

\[ f(\Psi) = 1 + A' \alpha' (1 - e^{-\Psi})^{\alpha'} \] (3.5)

The temperature history term was originally derived to model the viscous increase of denaturing protein dough. It was drawn upon the similarity between the increasing viscosity due to unfolding and complexing of protein molecules, and the viscous increase associated with polymer molecular weight growth during a polymerisation process (Morgan et al., 1989). In equation 3.5, \( A' \) is the ratio of gelatinised to ungelatinised viscosity, \( \alpha' \) a constant and \( \Psi \) the integral temperature-time function defined by equation 3.6.

\[ \Psi = \int_0^T T(t) k'_0 e^{\left(\frac{E_a}{RT(t)}\right)} dt \] (3.6)
Equation 3.6 was derived from absolute reaction theory and \( \Psi \) can be interpreted as the extent of gelatinisation during a non-isothermal heat treatment. The activation energy of gelatinisation is given by \( E_g \), and its rate constant by \( k'_{g0} \). For the reaction to advance, a minimum temperature \( T \) exceeding the gelatinisation temperature \( T_g \) is required.

Thixotropy resulting from shear was modelled by an asymptotic decay of viscosity (Morgan et al., 1989).

\[
f(\Phi') = 1 - B(1 - e^{-d\phi'})
\]  
(3.7)

In equation 3.7 \( B = (\mu_{\text{init}} - \mu_{\infty})/\mu_{\text{init}} \) is the relative viscosity reduction, \( \mu_{\text{init}} \) the viscosity before shearing, \( \mu_{\infty} \) the viscosity at infinite shear history and \( d' \) a constant. Integrating the average shear rate \( \tau \) experienced by the suspension yields \( \Phi' \).

\[
\Phi' = \int_0^t \tau dt
\]  
(3.8)

Dolan and Steffe (1990) determined the 11 constants of equation 3.1 through extensive measurements on native corn starch solutions which were gelatinised, held and cooled in a mixer viscometer.

However, their initial assumption of independence between the five terms of the model proved incorrect, as the temperature dependence \( f(T) \) and the shear history \( f(\Phi') \) proved strong functions of \( \Psi \), and of \( \Psi \) and \( T \) respectively. No mention was made of dependence of the shear rate term \( f(\gamma) \) on \( \Psi \), but it is known that gelatinising suspensions (increasing \( \Psi \)) usually change from dilatant to pseudoplastic behaviour (Selj, Wilkins, Morley, and Bailey, 1990; Rao, Okechukwu, Da Silva, and Oliveira, 1997). Such dependencies imply that at least some of the constants in equation 3.1 are functions of the independent variables, requiring a much greater number of data points.

Another difficulty of the Morgan model is the crude assumption of one gelatinisation temperature \( T_g \) for all granules. From equation 3.5 follows that a starch suspension held just above its gelatinisation temperature gelatinises completely. An actual suspension however, in which the granules gelatinise over a temperature interval, only partly gelatinises at \( T_g \) and hence exhibits a lower apparent viscosity.

Furthermore, given the range of gelatinisation temperatures for the individual granules, the rapidity of the process, and the inevitable temperature distribution inside the rheometer, the exact meaning of the apparent viscosity derived from such measurements is unclear.
A final remark concerns the strain history term of equation 3.7 and 3.8. The irreversible breakdown of the paste structure is responsible for the thixotropy term \( f(\Phi') \). Instead of defining \( \Phi' \) as the time-integral of the average shear rate, a physically better justified model would emerge if this breakdown would be related to the momentary strength of the paste structure (which is a decreasing function of \( \Psi \)), and the effective shear stresses in the suspension (which are a function of the local shear rates and the apparent viscosity).

Notwithstanding the previous criticism, the Morgan model provides useful clues. When the temperature- and shear history terms are omitted, it accurately describes the viscosity of many dilute gelatinised suspensions at moderate shear rates.

### 3.2.2 Yang’s model

An alternative way to obtain the rheology of a gelatinising suspension is the application of the Cox-Merz rule (Cox and Merz, 1958; Okechukwu and Rao, 1998). The Cox-Merz rule (equation 3.9) links dynamic and steady experimental data, based on the observation that for some polymeric liquids the complex viscosity \( \mu^* \) equals the apparent viscosity \( \mu \) at equal values of oscillatory frequency \( \omega \) and shear rate \( \gamma \). An important advantage of this method concerns the small strains involved in oscillatory rheometry, largely avoiding damage to the paste structure. Oscillatory rheometry is briefly discussed in appendix E.

\[
\mu^*(\omega) = \mu(\gamma) \bigg|_{\omega=\gamma} \tag{3.9}
\]

Yang and Rao (1998a) obtained the complex viscosity of an 8% w/w corn starch suspension for several moderate heating rates (1.6 to 6 °C/ min). When plotted against the temperature, their results collapsed into a single curve revealing the equilibrium nature of the gelatinisation process. Furthermore, for increasing oscillatory frequency the complex viscosity decreased, reflecting the pseudoplastic nature of many (partly) gelatinised suspensions. A representative series of graphs as a function of temperature and oscillatory frequency is shown in figure 3.6. The complex viscosity curves have very similar shapes, and Yang and Rao constructed a complex viscosity-temperature ‘master’ curve by scaling with an arbitrary reference frequency \( \omega_{\text{ref}} \), \( \beta \) a constant (equation 3.10).
Figure 3.6: Complex viscosity of an 8% w/w corn starch suspension, reproduced from Yang and Rao (1998a).

\[ \mu^*_{r}(T) = \mu^*(\omega, T) \cdot \left( \frac{\omega}{\omega_{\text{ref}}} \right)^\beta \]  

(3.10)

Next, Yang and Rao applied a modified Cox-Merz rule to obtain the apparent viscosity of these gelatinising suspension (equation 3.11).

\[ \mu^*(\omega, T) = C \left[ \mu(\gamma, T) \right]^\alpha \bigg|_{\omega=\gamma} \]  

(3.11)

As neither constant \( C \) nor shift factor \( \alpha \) were much affected by temperature, both could be determined using steady shear measurements at room temperature.

Combining equation 3.11 and 3.10 yielded a functional model for the apparent viscosity of gelatinising corn starch suspensions as a function of shear rate and temperature (equation 3.12). For modelling purposes, the reduced complex viscosity master curve was represented by one or more algebraic equations fit to the experimental data.
\[
\mu(\gamma, T) = \alpha \left[ \frac{\mu_e^*(T)}{C} \left( \frac{\omega_{\text{ref}}}{\gamma} \right) \right]^{\beta}
\]

This thermo-rheological model was successfully applied by Yang and Rao (1998b,c) to transient simulations of the natural convective heat transfer to a 3.5% w/w corn starch suspension in a cylindrical can. Liao, Rao, and Datta (2000) applied a similar model of a gelatinising 4% w/w waxy rice starch suspension in numerical simulations of the continuous sterilisation in a tubular heat exchanger.

However, Yang’s model has been derived for moderate heating rates (at most 6°C/min), in which case gelatinisation takes place on shorter time-scales than the heating itself. For all temperatures, the (partially) gelatinised starch granules can then be assumed to be in equilibrium with their environment. When higher heating rates are employed, this assumption is no longer valid, and the degree of gelatinisation and viscous development of the suspension lags behind that of the equilibrium \( T - \mu_e^* \) curve. In the most extreme case, an instantaneous raise to a temperature \( T > T_g^* \), the viscosity does not change at all during heating. When subsequently held at this temperature, the suspension gelatinises to its equilibrium state, and the viscosity rises to the equilibrium value at \( T \). In the Stork sterilisers, the heating rate can be on the order of degrees per second and it is known that gelatinisation kinetics are important.

A second drawback of the Yang model is the omission of irreversible shear destruction. Though highly cross-linked starches possess a high structural integrity, even after gelatinisation, most starch suspensions lose part of their viscosity on prolonged shear due to rupture and fragmentation of the swollen granules (Doublier, Llamas, and Le Meur, 1987; Svegmark and Hermansson, 1991; Loh, 1992). In small amplitude oscillatory rheometry, the shear load is minimal, but in the high temperature and high shear environment of food processing equipment, e.g. scraped surface heat exchangers, the loss of viscosity can be significant (Härröd (1989), figure 3.7). Often such equipment is selected to minimise shear destruction and to minimise the expected starch consumption to reach a target consistency.

A third problem of Yang’s model is the questionable shear dependence of the viscosity in high shear flow. As the values of \( \alpha \) and \( \beta \) in equation 3.12 are close or equal to 1 (Yang and Rao, 1998a; Liao, Tattiyakul, and Rao, 1999), the shear rate \( \gamma \) and the apparent viscosity \( \mu \) are almost inversely related. In terms of power-law rheology this implies a power-law index close to zero, resulting in near plug-flow velocity profiles. However, pseudoplasticity was seen to decrease and turn into dilatancy beyond a critical shear...
rate $\gamma_c$ (Bagley and Christianson, 1982; Okechukwu and Rao, 1996b; Rao, Okechukwu, Da Silva, and Oliveira, 1997; Rao and Tattiyakul, 1999). This dilatancy was observed in the early stages of the gelatinisation, but it was also found for fully gelatinised cross-linked waxy maize in the high temperature region of a UHT-steriliser (Dail and Steffe, 1990). Moreover, Yang and Rao (1998a) and Liao et al. (1999) reported lower values of $\beta$ as $\omega$ and $\gamma$ increased, implying decreasing pseudoplasticity. As the rheology of a starch suspension is governed by $\gamma$ in this complex manner, the shear rate dependence of the apparent viscosity itself must be a function of $\gamma$.

Finally, the Yang model does not take the irreversibility of the gelatinisation process into account. In straight pipe flow where the temperature/time profiles of the fluid elements are monotonically ascending functions, this is not relevant, but in curved pipe flows fluid elements follow corkscrew trajectories and may experience a fluctuating temperature. According to equation 3.12, the viscosity of the fluid would also fluctuate, implying a reversal of the gelatinisation process. To properly account for irreversibility, the suspension needs a memory for the highest temperature it has experienced.

### 3.2.3 Yang's model extended

We propose the concept of gelatinisation classes to adapt and extend Yang’s model to the high heating- and shear rates common in industrial scale heaters. First, the equilibrium reduced complex viscosity master curve obtained using oscillatory shear rheometry is divided into equal contributions of gelatinisation classes. Secondly, the contribution of each class to the non-equilibrium
Figure 3.8: Gelatinising part of the equilibrium reduced complex viscosity master curve of a 5% w/w cross-linked waxy maize suspension. Contribution of gelatinisation class $i$ to the master curve. Data taken from Tattiyakul and Rao (2000).

The reduced complex viscosity is determined taking into account the gelatinisation and shear destruction kinetics. Finally, the apparent non-equilibrium viscosity is calculated from the non-equilibrium complex viscosity master curve using the modified Cox-Merz rule. For high shear rates, the apparent viscosity is forced to follow a power-law rheology.

Starch granules possess a distribution of gelatinisation temperatures and when a starch suspension is held at a temperature within this gelatinisation interval, the gelatinisation process does not complete even for very long holding times (Lund, 1984; Pravisani, Califano, and Calvelo, 1985; Okechukwu and Rao, 1996a; Yang and Rao, 1998a). To describe such partial gelatinisation, we divide the gelatinising part ($T_{g1} \leq T \leq T_{g_n+1}$) of the equilibrium master curve (figure 3.8) into $n'$ classes each contributing $\Delta \mu^*_r$ to the maximum reduced complex viscosity $\mu^*_{r_{\text{max}}}$.

A gelatinisation class only contributes to the suspension’s viscosity if the temperature exceeds or has exceeded the gelatinisation temperature $T_{g_{i}}$ of that particular class $i$.

Gelatinisation classes do not necessarily coincide with sets of individual starch granules, but include contributions from partially gelatinised granules as well. Though loss of birefringence of individual granules usually occurs
over a narrow temperature interval $\Delta T = 1^\circ C$ (French, 1984), total loss of crystallinity and complete gelatinisation occur over a larger temperature range up to $\Delta T = 10^\circ C$ (Liu, Lelièvre, and Ayoung-Chee, 1991; Jenkins and Donald, 1998). Starch granules therefore participate in a number of classes, extending the temperature interval on which the granule fully gelatinises beyond that of a single class.

The actual contribution of a gelatinisation class to the master curve depends on its degree of gelatinisation, and the extent to which the paste structure has been modified by shear destruction. The kinetics of gelatinisation and shear destruction are modelled using the function $f'_i = f'_i(t, T)$ and $g'_i = g'_i(t, \gamma, T)$ respectively (equation 3.13).

$$\Delta \mu_{\gamma}^*(t, \gamma, T) = f'_i(t, T) g'_i(t, \gamma, T) \Delta \mu_{\gamma}^*$$ (3.13)

The functions $f'_i$ monotonically increase from 0 to 1 in the course of the process and represent the lag of gelatinisation as a result of high heating rates relative to the equilibrium curve. As soon the granules start swelling ($f'_i > 0$) the structure is susceptible to fragmentation, and the shear destruction functions $g'_i$ asymptotically decrease from 1 to $g'_i \geq 0$. The terminal value of $g'_i$ depends on the equilibrium viscosity $\mu_\infty$ after prolonged shearing at a given shear rate and temperature.

For $n'$ classes, the temperature-, time- and shear dependent non-equilibrium reduced complex viscosity $\mu_{\gamma}^*(t, \gamma, T)$ is given by equation 3.14.

$$\mu_{\gamma}^*(t, \gamma, T) = \begin{cases} 
\mu_{\gamma}^* & \forall \ T < T_{g_1} \\
\mu_{\gamma}^* f'_1 g'_1 (T) & \forall \ T_{g_1} \leq T < T_{g_2} \\
\mu_{\gamma}^* f'_i g'_i (T) - \Delta \mu_{\gamma}^* & \forall \ T_{g_2} \leq T < T_{g_3} \\
\sum_{i=1}^{n'-1} \left( f'_i g'_i (T) \right) + f'_i g'_i (T) \left( \mu_{\gamma}^* (T) - \left( n' - 1 \right) \Delta \mu_{\gamma}^* \right) & \forall \ T_{g_{n'}} \leq T < T_{g_{n'}+1} \\
\frac{\mu_{\gamma}^* (T)}{\sum_{i=1}^{n'-1} \Delta \mu_{\gamma}^*} \cdot \sum_{i=1}^{n'-1} \left( f'_i g'_i (T) \right) & \forall \ T \geq T_{g_{n'}+1}
\end{cases}$$ (3.14)

To complete the model, expressions for the gelatinisation function $f'_i$ and the shear destruction functions $g'_i$ are needed.
3.2.3.1 Kinetics of gelatinisation: the $f^*$-function

Despite extensive research into gelatinisation, little is known about its true kinetics (Lund, 1984; Lai and Kokini, 1991). Most kinetic data applies to the later stages ($t > 30$ s) of gelatinisation, when the greatest changes in viscosity have already occurred (Lund, 1984; Dolan and Steffe, 1990; Okechukwu and Rao, 1996a). Though gelatinisation is generally acknowledged to involve a number of consecutive and parallel physico-chemical processes, it is usually modelled using first order kinetics (Lund, 1984; Okechukwu and Rao, 1996a).

Data on the kinetics of gelatinisation in the early stages ($t \leq 30$ s) is virtually nonexistent. Lund (1984) states that the initial process follows multiple-order kinetics and observed gelatinisation to be complete within the thermal lag time (10 s) of the measuring equipment at 80°C. A substantial degree of gelatinisation occurred during this interval at lower temperatures. Most authors employ heating rates much lower than the rates of gelatinisation, rendering their kinetics a reflection of the distribution of the gelatinisation temperatures, rather than a description of the true rate of swelling and structure formation. Okechukwu and Rao (1996a) applied a flash gelatinisation procedure to instantaneously yield the target gelatinisation temperature, but their first sample point was taken only after 30 seconds.

Only the work of Marchant, Chapman, and Blanshard (1977); Marchant and Blanshard (1978) can be regarded approximately real-time, as they monitored the rate of loss of birefringence in wheat starch suspensions using a photocell. An arrangement of a temperature controlled air-heated chamber and a small sample size enabled precise and rapid (2°C/s) temperature jumps in the 2 - 8°C range. For small ($\Delta T = 2°C$) temperature jumps, a virtually instantaneous (within 2 - 3 s) loss in birefringence was observed, followed by a slow decrease occurring over much longer times ($\Delta t > 100$ s). For the large temperature jumps ($\Delta T = 8°C$) only the fast response was apparent. Marchant and Blanshard concluded this to be resulting from fast semi-cooperative melting of the crystalline regions, mediated by a slower rearrangement and disentanglement of (amylopectine) chains in the amorphous phase for small temperature jumps or for gradual heating (Marchant and Blanshard, 1978; French, 1984).

Gelatinisation of cross-linked starches in a UHT-process having high heating rates and short processing times is very likely governed by the fast melting process. Of course, loss of birefringence is only an approximate measure of gelatinisation, and Liu et al. (1991) and Jenkins and Donald (1998) state that all birefringence (long range order) is lost well before the complete loss of crystallinity (short range order), which in its turn has finished before complete granule swelling and gelatinisation. The macroscopic increase in
viscosity is therefore expected to have a substantially greater time-constant than the birefringence kinetics, though no accurate data is available.

To estimate the kinetics of the viscous development of the gelatinisation classes, we assume a linear temperature increase employed during dynamic rheometry to be the highest heating rate for which the suspension is still at equilibrium. Higher heating rates result in lower complex viscosities at equal temperature, as was observed by Yang and Rao (1998a), whereas lower heating rates of course yield the equilibrium curve.

Secondly, the extent of gelatinisation is expressed as its contribution to the reduced complex viscosity. Doing so, any complicated relationship between a rather arbitrarily defined, and a posteriori determined degree of gelatinisation (e.g. granule volume fraction (Bagley and Christianson, 1982) or average granular diameter (Okechukwu and Rao, 1995)) and the viscosity is avoided. Moreover, the equilibrium curve contains no information on any degree of gelatinisation, which would have to be monitored simultaneously.

Thirdly, we assume the reaction to be zero-order in the reduced complex viscosity. Granules gelatinise independently in dilute suspensions, and a higher order mechanism yields a qualitatively wrong development of viscosity. Gelatinisation must have progressed considerably before any increase in viscosity can be measured (Evans and Haisman, 1979), whereas any higher order mechanism would exhibit the greatest rise in viscosity in the initial stages of the process.

Finally, for the rate of gelatinisation $k_g$ an Arrhenius dependence on temperature is adopted and the activation energy $E_g$ is taken equal and constant for all gelatinisation classes. This latter simplification may seem crude, but activation energies for starch suspensions presented in the literature show a wide variety (from 79 kJ/mol for rice starch (Kubota, Hosokawa, Suzuki, and Hosaka, 1979) to 740 kJ/mol for corn starch (Dolan and Steffe, 1990)), and any estimate is therefore somewhat arbitrary.

Heating rate $dT/dt = \dot{T}$ and the rate of increase of reduced complex viscosity $d\mu^*_r/dt = k_g$ can be combined into equation 3.15.

\[
\frac{d\mu^*_r}{dT} = \frac{k_g}{\dot{T}} \tag{3.15}
\]

Substitution of the Arrhenius model and integrating over the temperature boundaries of each gelatinisation class yields the rate constant $k_{g0i}$ of each class $i$ (equation 3.16).
\[ \Delta \mu_r^* = \int_{T_{g_i}}^{T_{g_i+1}} \frac{k_{g_0}}{T} e^{-\frac{E_g}{\pi T}} dT \]  

(3.16)

Finally, integrating the gelatinisation kinetics and dividing by the maximum viscous contribution of a class, we obtain the gelatinisation function \( f'_i \) (equation 3.17). Of course \( f'_i \) only increases for \( T \geq T_{g_i} \) and for incomplete gelatinisation (\( f'_i \leq 1 \)).

\[
f'_i(t, T) = \frac{1}{\Delta \mu_r^*} \int k_{g_0} e^{-\frac{E_g}{\pi T}} dT \quad \forall \quad \frac{1}{\Delta \mu_r^*} \int k_{g_0} e^{-\frac{E_g}{\pi T}} dT \leq 1
\]

(3.17)

\[
f'_i(t, T) = 1 \quad \forall \quad \frac{1}{\Delta \mu_r^*} \int k_{g_0} e^{-\frac{E_g}{\pi T}} dT > 1
\]

### 3.2.3.2 Kinetics of shear destruction: the g’-function

Swollen starch pastes are vulnerable to shear, especially native starches in the later stages of gelatinisation (Dengate, 1984; Doublier et al., 1987; Sveggmark and Hermansson, 1991; Loh, 1992). This is not surprising since their structural strength diminishes as the granules swell and gradually dissolve. Cross-linking reduces swelling and solubilisation, and such starches exhibit a much higher structural resistance to mechanical and thermal load (Loh, 1992; Abdelrahim et al., 1995; Rao et al., 1997; Tattiyakul and Rao, 2000). Furthermore, below the close packing point, cross-linked starches show a lower viscosity at equal weight fraction than their native counterparts (Steeneken, 1989), reducing the detrimental shear stresses.

Quantitative data on the kinetics of shear destruction in suspensions of swollen starch granules is rare (Lagarrigue and Alvarez, 2001). Dolan and Steffe (1990) fit the exponential viscous decay of 6.4% w/w native corn starch suspensions to the thixotropy term \( f(\Phi') \) of the Morgan model (equation 3.7).

Using a Brookfield RVTD mixer rheometer, they found that shear destruction only occurred after considerable gelatinisation and at a sufficiently high shear rate. Furthermore, shear destruction was a comparatively slow process and its severity depended strongly on temperature and temperature history.

Data obtained under well-defined shear conditions in a concentric rheometer were provided by Nguyen, Jensen, and Kristensen (1998). Gelatinised native maize and waxy maize starch suspensions exhibited considerable thixotropy (up to 50%) during the first 10 minutes of steady shear in the 100 - 200 1/s range. For higher temperatures the relative decrease in viscosity was lowest. After prolonged shearing, no thixotropy remained which was attributed to irreversible breakdown of the paste structure. At final equilibrium the pastes could be described by the standard power-law model.
Analogous to some models developed for the extrusion of doughs (Lai and Kokini, 1991), we propose to relate the shear destruction to the specific mechanical power input, or simply, the dissipation (equation 3.18). This is a reasonable assumption, as the highest dissipation is accompanied by the highest shear stresses, which play a key role in granule rupture.

\[ \frac{d \mu}{dt} \propto -\mu \Phi \]  \hspace{1cm} (3.18)

Integrating equation 3.18 and defining a rate constant of shear destruction \( k_s \) yields an exponentially decaying function for the apparent viscosity (equation 3.19). This equation bears some similarity to the shear degradation term of the Dolan model (equation 3.7), but closer inspection reveals that the dependence on shear rate is different. Whereas in the Dolan model the average shear rate \( \overline{\gamma} \) is the key parameter, in equation 3.19 this part is played by \( \Phi \). For steady flow in a concentric rheometer \( \overline{\gamma} \) is equal to \( \gamma_r \), whereas \( \Phi \) is equal to \( \gamma_r^2 \). Equation 3.19 therefore depends on \( \gamma \) in a non-linear fashion.

\[ \mu(t, \gamma) = \mu_{\text{init}}(\gamma) e^{-\int k_s \Phi \, dt} \]  \hspace{1cm} (3.19)

Structural breakdown only occurs for sufficiently high shear stresses, and \( k_s \) in equation 3.19 is set zero when the structural strength \( \tau_s \) exceeds the effective shear stress \( \tau_e \) in the fluid. The effective shear stress is calculated from the local apparent viscosity and the magnitude of the rate of strain tensor \( \Delta_{ij} \) (equation 3.20).

\[ \tau_e = \mu \left| \frac{1}{2} \sum \sum \Delta_{ij} \Delta_{ij} \right|^\frac{1}{2} \]  \hspace{1cm} (3.20)

Transforming the relative decrease of apparent viscosity to the relative decrease of complex viscosity, we can now implement the shear destruction function \( g'_i \) (equation 3.21).

\[ g'_i(t, \gamma, \Theta) = \left( \frac{\mu}{\mu_{\text{init}}} \right)^\alpha = \left( e^{-\int k_s \Phi \, dt} \right)^\alpha \]  \hspace{1cm} (3.21)

In first approximation, all gelatinisation classes are assumed to respond identically to mechanical loading. The structural strength and the kinetic constant of the shear destruction can be determined from separate steady shear measurements (please refer to appendix E).
3.2.3.3 Shear dependence in high shear flow

As noted in paragraph 3.2.2, the degree of pseudoplasticity or dilatancy depends on the shear rate. General agreement exists on the importance of the swollen granules for the viscosity, as only near the close packing point major increases in viscosity are observed (Evans and Haisman, 1979; Bagley and Christianson, 1982; Okechukwu and Rao, 1995). The rheology in terms of pseudoplasticity or dilatancy is thought to depend primarily on granule deformability (Bagley and Christianson, 1982; Okechukwu and Rao, 1996b; Rao et al., 1997; Rao and Tattiya kul, 1999). Rigid spheres in the close packing configuration exhibit dilatancy and an increase in volume when shear is applied (Bagley and Christianson, 1982; Frith and Lips, 1995). Deformable particles such as swollen starch granules are able to adapt and move smoothly relative to one another, resulting in macroscopic pseudoplasticity.

As the shear rate increases, collisions between granules occur over a shorter time span, and for reasons of inertia and relaxation time, the granules cannot adapt sufficiently quick. The apparent rigidity of the granules increases, and a shift from pseudoplastic towards dilatant behaviour can be expected, especially for cross-linked starches (Dail and Steffe, 1990) or short cooking times (Bagley and Christianson, 1982; Christianson and Bagley, 1983). However, granule deformability also depends on the magnitude of the shear stresses in the suspension (Bagley and Christianson, 1982) and a strongly pseudoplastic suspension is unlikely to turn dilatant. For the very high shear rates necessary to accomplish dilatancy, the shear stresses are so great that structural breakdown would be inevitable. The associated loss of viscosity is noticeable as an additional apparent pseudoplasticity.

If present, the role of amylase exuded into the continuous phase is considered to be minor (Evans and Haisman, 1979), though Doublier et al. (1987) reported a contribution by a weak structural network, which was reversibly broken under mild shear. Da Silva, Gonçalves, and Rao (1993) explained discrepancies between the steady and oscillatory shear measurements in terms of such a network, as it was not apparent in steady shear measurements. Nguyen et al. (1998) attributed thixotropy in gelatinised maize starch suspensions to the irreversible breakdown of the gel structure, after which the suspension exhibited regular pseudoplasticity.

Values of $\alpha$ and $\beta$ in the Yang model are close to one (Yang and Rao, 1998a; Liao et al., 1999), implying a strong pseudoplasticity ($n \to 0$). For low shear rates this may be explained from the destruction of the amyllose network in the continuous phase, or from the breaking of similar bonds between amyllopectine side chains of different granules (Nguyen et al., 1998). In high shear flow however, this network is absent and the rheology is governed
by the swollen granules. Under those circumstances, cross-linked starches of interest for UHT-sterilisation exhibit considerably less pseudoplasticity (Härröd, 1989; Dail and Steffe, 1990; Abdelrahim et al., 1995).

The power-law model was combined with the Yang model to yield a description of the rheology at all shear rates expected in a UHT-steriliser. Up to a critical shear rate $\gamma_c$ the modified Cox-Merz rule of Yang and Rao was employed, and beyond it the power-law model took over. To ensure a smooth transition, the consistency index $m$ was calculated from the apparent viscosity at the critical shear rate. The non-equilibrium apparent viscosity $\mu(t, \gamma, T)$ can then be calculated from equation 3.22.

$$\mu(t, \gamma, T) = \sqrt{\mu^2 \left( \frac{t, \gamma, T}{C} \right)_C \left( \frac{\omega_{ref}}{\gamma} \right)^{\beta}} \quad \forall \quad \gamma < \gamma_c$$

$$\mu(t, \gamma, T) = m (\gamma)^{n-1} = \sqrt{\mu^2 \left( \frac{t, \gamma, T}{C} \right)_C \left( \frac{\omega_{ref}}{\gamma_c} \right)^{\beta} \cdot \left( \frac{\gamma}{\gamma_c} \right)^{n-1}} \quad \forall \quad \gamma \geq \gamma_c$$  \hspace{1cm} (3.22)

As an example, figure 3.9 shows the apparent viscosity at the reference shear rate for equilibrium conditions (eq), for a high heating rate (tl), for significant shear destruction (sd) and for a combination of both (tl+sd).

### 3.3 Modelling gelatinised suspensions

When cooled to room temperature or below, amyllose exuded into the continuous phase may form a gel-like network, which slowly increases in crystallinity. On prolonged storage, the amyllopectine of the swollen granular ghosts recrystallises, and side chains of amyllopectine molecules in different granules may associate. These processes imply further rheological changes, particularly an increase in viscosity and the onset of solid-like behaviour.

Whereas amyllose gelation starts immediately following gelatinisation, amyllopectine gelation and recrystallisation occur over a period of days to weeks (Zobel, H.F., 1984; Durrani and Donald, 1995). Their kinetics can safely be ignored on the time scale of the cooling stage of the UHT-treatment, which is on the order of seconds to minutes.

It is difficult to distinguish between the separate effects on viscosity of the incipient amyllose gelling, and the instantaneously increasing rigidity of the swollen granules. Little quantitative results are available on the gelling kinetics of amyllose, as gelation is usually monitored as a function of time and
temperature simultaneously (Hanssen, Hoseney, and Faubion, 1990; Svegmark, Kidman, and Hermansson, 1993; Breton, Korulczuk, Doublier, and Maingonnat, 1995; Lii, Tsai, and Tseng, 1996; Vesterinen, Suortti, and Au-tio, 2001). Aguilera and Rojas (1997) presented the developing storage modulus \( G' \) for a 10% w/w gelatinised cassava starch suspension at constant temperature. Using oscillatory rheometry, \( G'' \) was shown to reach a constant value after 5 minutes on quenching from 90 to 10 °C, indicating completion of gelation.

Gelation of amylase probably requires minutes to have a significant impact on the rheological properties of gelatinised starch suspensions. However, cooling rates in industrial sterilisers are very high, and the amylase exudation into the continuous phase is often limited as a result of cross-linking in the starches employed. Moreover, the continuous application of shear is likely to suppress network formation, as any inter-granular bonds present in maize and waxy maize starch suspensions were broken by steady shearing (Nguyen et al., 1998). Launay, Doublier, and Cuveller (1986) state that gelation does not impact the structure of the suspension provided measurements are performed rapidly after pasting. An important implication of this is that in the current work visco-elasticity can be neglected.
Apparent viscosity of predominantly viscous, gelatinised starch suspensions can be modelled using the regular power-law model (Evans and Haisman, 1979; Härröd, 1989; Self et al., 1990; Dail and Steffe, 1990; Holdsworth, 1993; Abdelrahim et al., 1995; Nguyen et al., 1998; Rao and Tattiyakul, 1999), extended with the Arrhenius model (Launay et al., 1986; Dolan and Steffe, 1990; Heydon et al., 1996; Xu and Raphaelides, 1998; Okechukwu and Rao, 1998; Lagarrigue and Alvarez, 2001) to incorporate the immediate temperature dependency. Where appropriate, shear destruction can be included using equation 3.19. For rapid gelatinisation and an identical response of all classes to mechanical load, the apparent viscosity of a hot and cooled paste in steady flow is given by equation 3.23.

\[
\mu(\gamma, T, t) = m_0 \gamma^{n-1} e^{-\frac{E_a}{RT}} e^{-\int k_a \Phi dt}
\]

(3.23)

### 3.4 Conclusions

Gelatinisation and consecutive gelation cause dramatic changes in the rheological properties of heated dilute starch suspensions. The viscosity may rise several orders of magnitude, and upon cooling and storage solid-like gels are prone to form.

Two models describing the developing viscosity during gelatinisation were critically assessed. The Morgan model is the more comprehensive as it incorporates both instantaneous and history effects of the shear rate and temperature applied during pasting. Yang’s model is perhaps more practical, as the rheometry required is relatively simple. However, Yang’s model essentially regards gelatinisation to be at equilibrium, and kinetics of gelatinisation and shear destruction are omitted.

To develop a model of the shear-, temperature- and time dependent rheology of starch suspensions in industrial UHT-sterilisers, the Yang model was adapted for use under non-equilibrium conditions. Discrete gelatinisation classes were introduced to enable incorporation of gelatinisation-lag due to high heating rates, and shear destruction resulting from high shear stresses. Finally, the immediate shear dependency was made to conform to power-law rheology above a critical shear rate.
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Chapter 4

Starch gelatinisation in coiled heaters

A gelatinising model food derived from a 5% w/w cross-linked waxy maize suspension was simulated in coiled heaters to assess the impact of centrifugal forces on developing laminar flow and heat transfer. For four coil diameters ($D = 0.25, 1, 2.5$ and $\infty$ m) and three flow velocities ($\overline{V} = 0.5, 1$ and $2$ m/s), heat transfer, viscous development and the severity of channeling were evaluated.

Increasing curvature proved to suppress channeling as a result of more uniform heating and gelatinisation. The maximum attainable viscosity is also higher, implying a lower starch consumption for a target viscosity. Higher flow rates necessitate longer heaters, and the maximum viscosity decreases. Moderate product velocities are therefore recommended.

4.1 Introduction

Continuous thermal processing is extensively used in the food industry to preserve liquid foods. Compared to batch processing, continuous pasteurisation and sterilisation yield safe products of a higher nutritional quality (Holdsworth, 1992).

Viscous products such as sauces and custards may contain thermally activated thickeners. A commonly used agent is starch, which is extracted in granular form from a variety of plants. In excess water and heated above a

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1Submitted for publication to Biotechnology Progress.
threshold temperature \( T_g \), starch granules lose their semi-crystalline properties and absorb large amounts of water. Such melting and swelling, which is often accompanied by leaching of soluble material into the continuous phase, is termed gelatinisation (Lund, 1984).

Macroscopically, gelatinisation causes a dramatic increase in viscosity in the course of the production process, and it is of great importance in convective heat transfer. Large differences in viscosity between raw and cooked suspensions induce channeling phenomena (Liao, Rao, and Datta, 2000) which strongly widen residence time distributions. Different thermal histories of the fluid elements may imply the coexistence of ungelatinised and severely overcooked product at the exit of a heater.

Chapter 5 presents simulations of the sterility and thiamine retention for constant property viscous power-law foods in the high temperature \( T > 100^\circ \text{C} \) section of a continuous coiled Ultra High Temperature (UHT) steriliser. Contrary to previous authors (Simpson and Williams, 1974; Kumar and Bhattacharya, 1991; Bhamidipati and Singh, 1994; Jung and Fryer, 1999; Liao, Rao, and Datta, 2000), the impact of buoyant and centrifugal forces, where appropriate, was fully accounted for. Coiled heating and cooling tubes are shown to be potentially superior to straight heat exchangers, primarily as a result of higher heat transfer rates.

Though most authors include the instantaneous shear- and temperature dependence of viscosity, only Liao, Rao, and Datta (2000) attempted to model the dramatic viscous changes associated with gelatinisation. To this end, a thermo-rheological model developed by Yang and Rao (1998), was applied to continuous heating and cooling in a tubular UHT-steriliser.

However, the Yang model is essentially an equilibrium model, linking macroscopic apparent viscosity to the instantaneous temperature. As such it may be suitable for use at low flow and low heating rates, but it does not represent industrial plant conditions where high shear and high heating rates are common (Lagarriague and Alvarez, 2001).

At high heating rates, gelatinisation and the macroscopic viscosity at a given temperature \( T > T_g \) lag behind those of the equilibrium curve. Furthermore, swollen granules are vulnerable to rupture and fragmentation by shear stresses. In food processing equipment such as scraped surface heat exchangers, the loss of viscosity can be severe (Härröd, 1989). Though mechanical degradation in tubular heat exchangers is usually less (please refer to chapter 3), it cannot be totally discounted. In addition to an instantaneous dependence on shear rate and temperature, the viscosity of a starch suspension therefore also depends on temperature- and shear history (figure 4.3, Dolan and Steffe (1990)).
Finally, Yang’s model implies a quite strong pseudoplasticity, as shear rate and apparent viscosity are almost inversely related. However, Hårröd (1989) and Abdelrahim, Ramaswamy, and van de Voort (1995) reported considerably less pseudoplasticity, and (Dail and Steffe, 1990) even found dilatancy in gelatinised suspensions.

This chapter reports simulations of the impact of curvature on the flow and heat transfer of a gelatinising viscous starch suspension in coiled tubular heaters. The thermo-rheological equilibrium model of Yang and Rao was extended to include shear destruction and the lag in gelatinisation for high heating rates. At high shear rates, the instantaneous shear dependence was made to conform to the standard power-law model. A model food derived from the properties of a gelatinising 5% w/w cross-linked waxy maize suspension was heated for three average velocities in four different coils.

4.2 Problem analysis

A gelatinising viscous model food was simulated in straight and coiled tubular heaters (figure 4.1). The geometry, and the flow and temperature range were chosen such that gelatinisation would start some distance into the heater, but would most likely be complete before the exit.

![Figure 4.1: Set-up of the heater and the ultra high temperature section](image)

Product enters the heater \((d = 0.025 \text{ m}, L = 200 \text{ m})\) at \(T = 50^\circ\text{C}\) and is heated by a constant wall temperature \(T_w = 100^\circ\text{C}\). Downstream, the fluid passes into the UHT-section treated in chapter 5. Four coils having diameter \(D = 0.25, 1, 2.5\) and \(\infty\) m were tested, the latter corresponding to a straight tube. To assess the impact of flow rate, three average axial fluid velocities \((\overline{w} = 0.5, 1\) and \(2 \text{ m/s}\)) in the range employed in industrial heaters were adopted. Coil orientation was unimportant as buoyancy effects were not considered.
4.2.1 Governing equations and boundary conditions

The equations of continuity, motion and energy in cartesian coordinates for steady, incompressible power-law fluids are given by equations 4.1 to 4.3 (Bird, Stewart, and Lightfoot, 1960).

\[ \nabla \cdot \mathbf{u} = 0 \quad (4.1) \]

\[ \rho (\mathbf{u} \cdot \nabla) \mathbf{u} = -\nabla p + \mu \nabla^2 \mathbf{u} + \mathbf{F} \quad (4.2) \]

\[ \rho c_p \mathbf{u} \cdot \nabla T = \lambda \nabla^2 T + \mu \Phi \quad (4.3) \]

The progress of the gelatinisation and the subsequent shear destruction (the \( f' \) and \( g' \) functions respectively, please refer to section 4.2.2) are modelled using transport equations for scalars (equation 4.4).

\[ \rho (\mathbf{u} \cdot \nabla) \phi = \rho \mathbf{D} \nabla^2 \phi + S_\phi \quad (4.4) \]

In the case of loosely coiled pipes \((d/D < 0.1, \text{Baurmeister and Brauer (1979)})\), the effect of curvature on the flow and temperature field can be included in the term \( \mathbf{F} \) in the equations of motion. This centrifugal body force \( F_c \) (equation 4.5) acts in the x-direction as shown in figure 4.2.

\[ F_c = \rho \frac{w^2}{x} \quad (4.5) \]

Chapter 5 and appendix D show buoyant forces to be of minor importance in curved heaters. As the foods in this chapter are more viscous, and the fluid velocities are equal or higher, the magnitude of the buoyant relative to the centrifugal secondary flow further decreases (Sillekens, Rindt, and Van Steenhoven, 1998) and buoyant forces are omitted. Coil pitch is well below the coil diameter, rendering torsional effects unimportant (Mishra and Gupta, 1979; Xin and Ebadian, 1997).

Viscous dissipation enters the energy equation 4.3 through the term \( \mu \Phi \) (Bird, Stewart, and Lightfoot, 1960). In gelatinised viscous flows, dissipation may increase the fluid temperature up to several degrees.

As inlet boundaries, developed Newtonian velocity profiles and uniform temperature and scalar quantities were employed. At the wall, the no-slip condition and a constant temperature were imposed, and the wall was assumed impermeable to scalar species. At the outlets, static pressure as well as all axial gradients were set zero. All cases featured planar symmetry.
4.2.2 Fluid properties

Using small amplitude oscillatory rheometry, Yang and Rao (1998) obtained the complex viscosity $\mu^*(\omega, T)$ of gelatinising dilute corn starch suspensions for moderate heating rates as a function of frequency $\omega$. When plotted against temperature $T$ and scaled with an arbitrary reference frequency $\omega_{\text{ref}}$, these curves collapsed into a reduced complex viscosity 'master' curve $\mu^*_c(T)$, which was subsequently linked to the apparent viscosity $\mu(\gamma, T)$ using a modified Cox-Merz rule (Cox and Merz, 1958)(equation 4.6).

$$
\mu^*(\omega, T) = \mu^*_c(T) \left(\frac{\omega_{\text{ref}}}{\omega}\right)^{\beta} = C \left[\mu(\gamma, T)\right]^\alpha \bigg|_{\omega = \gamma} \tag{4.6}
$$

Following a similar procedure, Liao, Tattiyakul, and Rao (1999) obtained a functional model of the apparent viscosity of waxy rice starch which was used to simulate the continuous sterilisation of a gelatinising suspension in a continuous heater and cooler (Liao, Rao, and Datta, 2000).

We propose the concept of gelatinisation classes to extend Yang’s model to the heating and shear rates common in industrial heaters. First, the equilibrium master curve is divided into $n'$ classes each contributing $\Delta \mu^*_c$ to the maximum equilibrium reduced complex viscosity $\mu^*_{\text{max}}$ (figure 4.4). $T_{g1}$ marks the starting temperature of the first gelatinisation class, whereas $T_{g_{n'+1}}$ represents the end temperature of the last class. Secondly, the actual

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2 Readers familiar with section 3.2.3 may proceed to the end of this section.
contribution of each class to the non-equilibrium master curve is determined taking gelatinisation and shear destruction kinetics into account. Finally, the apparent viscosity is calculated from the non-equilibrium master curve using the modified Cox-Merz rule. For high shear rates, the apparent viscosity is forced to follow a power-law rheology.

Gelatinisation classes are required to model partial gelatinisation and shear destruction for temperatures within the gelatinisation interval. Classes do not necessarily coincide with sets of individual granules, so granules may participate in several classes, extending their gelatinisation interval beyond that of a particular class.

The contribution of a class to the master curve depends on its degree of gelatinisation, and the extent to which the paste structure has been modified by shear destruction. The kinetics of gelatinisation and shear destruction are modelled using the functions $f_i' = f'_i(t, T)$ and $g_i' = g'_i(t, \gamma, T)$ respectively (equation 4.7).

$$\Delta \mu_{r_i}'(t, \gamma, T) = f'_i(t, T) g'_i(t, \gamma, T) \Delta \mu_{r_i}'$$ (4.7)
In the course of gelatinisation, $f_i^t$ monotonically increases from 0 to 1. It represents the lag of gelatinisation relative to the equilibrium curve as a result of high heating rates. As soon a class starts gelatinising ($f_i^t > 0$), it is susceptible to shear degradation, and $g_i^t$ asymptotically decreases from 1 to $g_i^t \geq 0$. The terminal value of $g_i^t$ depends on the equilibrium viscosity $\mu_\infty$ at a given shear rate and temperature.

To estimate the gelatinisation kinetics, we assume a heating rate $\dot{T} = dT/dt$ beyond which gelatinisation can no longer be considered at equilibrium. Adopting zero order kinetics for the rate of increase of reduced complex viscosity $d\mu_r^*/dt = k_g$, and combining with the heating rate $\dot{T} = dT/dt$ yields equation 4.8.

$$\frac{d\mu_r^*}{dT} = \frac{k_g}{\dot{T}} \tag{4.8}$$

Next, substituting an Arrhenius dependence for the reaction rate $k_g$ and integrating over the temperature boundaries yields the rate constant $k_{g_i}$ for each class $i$ (equation 4.9).

$$\Delta \mu_r^* = \int_{T_{g_i}}^{T_{g_i+1}} \frac{k_{g_i}}{T} e^{-\frac{E_g}{RT}} dT \tag{4.9}$$

Figure 4.4: Contribution of gelatinisation class $i$ to the reduced complex viscosity master curve.
Integrating the gelatinisation kinetics and dividing by the maximum viscous contribution of a class, we obtain the gelatinisation functions $f'_i$ (equation 4.10). The rate constant $k_{g_0}$ is set zero for $T < T_{g_i}$ (gelatinisation temporarily ceases) and for $f'_i \geq 1$ (gelatinisation is complete).

$$f'_i(t, T) = \frac{1}{\Delta \mu} \int k_{g_0} e^{-\frac{k_{g_0}}{\Delta \mu} d t} \quad \forall \quad \frac{1}{\Delta \mu} \int k_{g_0} e^{-\frac{k_{g_0}}{\Delta \mu} d t} \leq 1$$

$$f'_i(t, T) = 1 \quad \forall \quad \frac{1}{\Delta \mu} \int k_{g_0} e^{-\frac{k_{g_0}}{\Delta \mu} d t} > 1$$

(4.10)

Analogous to models developed for the extrusion of doughs (Lai and Kokini, 1991), we propose to relate shear destruction to the dissipation (equation 4.11). This seems reasonable, as the highest dissipation is accompanied by the highest shear stress, which plays a key role in granule rupture.

$$\frac{d \mu}{d t} \propto -\mu \Phi$$

(4.11)

Integrating equation 4.11 and defining a rate constant of shear destruction $k_s$ yields an exponentially decaying function for the apparent viscosity (equation 4.12). The dissipation function $\Phi$ is quadratically linked to the shear rate, and $\mu$ depends on $\gamma$ in a non-linear fashion.

$$\mu(t, \gamma) = \mu_{init}(\gamma) e^{-\int k_s \Phi d t}$$

(4.12)

In first approximation, all gelatinisation classes are assumed to respond identically to mechanical load. The rate constant $k_s$ can be calculated from the time $\Delta \mu = \mu_{\infty}$ required at constant shear rate to lower the initial viscosity $\mu_{init}$ to the asymptotic equilibrium viscosity $\mu_{\infty}$. To ensure a uniform shear rate, such tests should be performed in a concentric or a cone and plate rheometer. At equilibrium, the structural strength of the gelatinisation classes can be estimated from $\tau_g = \mu_{\infty} \gamma$.

Structural breakdown of a class only occurs above its gelatinisation temperature $T_{g_i}$ and for sufficiently high shear stresses. Rate constant $k_s$ in equation 4.12 is set zero when $T < T_{g_i}$ and when the structural strength of the class $\tau_g$ exceeds the effective shear stress $\tau_e$ in the fluid. Combining equations 4.6, 4.7 and 4.12 yields the shear destruction function $g'_i$ (equation 4.13).

$$g'_i(t, \gamma, T) = \left( \frac{\mu}{\mu_{init}} \right) = \left( e^{-\int k_s \Phi d t} \right)^\alpha$$

(4.13)
Both \( f'_i \) and \( g'_i \) are calculated as transported scalars having a zero diffusion coefficient \( \mathbb{D} \). The source term \( S_0 \) in equation 4.4 is equal to \( k_g \exp(-E_g/RT) \) for the gelatinisation kinetics (equation 4.10), and to \( k_s \Phi \) for the shear destruction (equation 4.13).

Using the equilibrium reduced viscosity \( \mu^*_r(T) \), the contributions per class \( \Delta \mu^*_r \) and the functions \( f'_i \) and \( g'_i \), we can construct the non-equilibrium reduced complex viscosity \( \mu^*_r(t, \gamma, T) \) (equation 4.14).

\[
\begin{align*}
\mu^*_r(T) &= 0 \quad &\forall \quad T < T_{g1} \\
\mu^*_r(t, \gamma, T) &= f'_1 g'_1 \mu^*_r(T) \quad &\forall \quad T_{g1} \leq T < T_{g2} \\
\mu^*_r(t, \gamma, T) &= f'_1 g'_1 \Delta \mu^*_r + f'_2 g'_2 (\mu^*_r(T) - \Delta \mu^*_r) \quad &\forall \quad T_{g2} \leq T < T_{g3} \\
\mu^*_r(t, \gamma, T) &= \sum_{i=1}^{n'} (f'_i g'_i \Delta \mu^*_r) + f'_n g'_{n'} (\mu^*_r(T) - (n' - 1) \Delta \mu^*_r) \quad &\forall \quad T_{g_{n'}} \leq T < T_{g_{n'+1}} \\
\mu^*_r(t, \gamma, T) &= \frac{\mu^*_r(T)}{\sum_{i=1}^{n'} (f'_i g'_i \Delta \mu^*_r)} \cdot \sum_{i=1}^{n'} (f'_i g'_i \Delta \mu^*_r) \quad &\forall \quad T \geq T_{g_{n'+1}}
\end{align*}
\]  

(4.14)

Finally, the power-law rheology was combined with the non-equilibrium model to cover all shear rates expected in industrial heaters. Up to a critical shear rate \( \gamma_c \), the modified Cox-Merz rule (equation 4.6) was employed, and beyond it the power-law model took over. To ensure a smooth transition, the consistency index \( m \) was calculated from the apparent viscosity at the critical shear rate. The non-equilibrium model for the apparent viscosity \( \mu(t, \gamma, T) \) is then given by equation 4.15.

\[
\begin{align*}
\mu(t, \gamma, T) &= \mu_0 + \frac{\mu^*(t, \gamma, T)}{C} \left( \frac{\omega/\gamma_c}{\gamma} \right)^\beta \quad &\forall \quad \gamma < \gamma_c \\
\mu(t, \gamma, T) &= \mu_0 + m (\gamma)^{n-1} = \mu_0 + \frac{\mu^*(t, \gamma, T)}{C} \left( \frac{\omega/\gamma_c}{\gamma_c} \right)^\beta \cdot \left( \frac{\gamma}{\gamma_c} \right)^{n-1} \quad &\forall \quad \gamma \geq \gamma_c 
\end{align*}
\]  

(4.15)

For the current simulations, a model food was defined based on the properties of a 5% cross-linked waxy maize suspension. The complex viscosity master
curve presented by Tattiyakul and Rao (2000) was divided into ten gelatinisation classes, and the heating rate they employed in oscillatory rheometry ($T = 0.035 \degree C/s$) was assumed to be the highest for which the equilibrium assumption still held. After a review of the literature, the activation energy of the gelatinisation and viscous build-up was set to $E_g = 100$ kJ/mol. The kinetics and extent of shear destruction were estimated from the work of Nguyen, Jensen, and Kristensen (1998) on 6% w/w native waxy maize starch pastes, using $\mu_\infty = 0.5\mu_\text{init}$ and $\Delta t_{\mu=\mu_\infty} = 600$ s. The power-law index $n = 0.7$ above the critical shear rate $\gamma_c = 50$ l/s was taken from gelatinised 5% w/w cross-linked waxy maize starch suspensions investigated by Abdelrahim, Ramaswamy, and van de Voort (1995). To ensure laminar flow at all flow velocities, $\mu_0$ in equation 4.15 was set to 0.05 Pa s ($Re_{\text{max}} = 2000$). Application of the proposed non-equilibrium model for this gelatinising model food under constant heating- and shear rate yields the apparent viscosity curves shown in figure 4.3.

Though gelatinisation is an endothermic process (Lund, 1984) representing a sink term in the energy equation, the enthalpy of gelatinisation $\Delta H_g$ was neglected. Tentative calculations on a coarse mesh with $\Delta H_g = 20$ J/g confirmed its small impact on the current dilute suspensions.

Density $\rho = 1000$ kg/m³, heat capacity $c_p = 4180$ J/(kg °C), and the thermal conductivity $\lambda = 0.5$ W/(m °C), were assumed constant. All diffusion coefficients $\mathcal{D}$ in equation 4.4 were set zero.

### 4.3 Numerical considerations

Solutions to equations 4.1 - 4.3 and 4.4 have been approximated using the Star finite volume package³. The SIMPLE algorithm was employed to solve the coupled equations after discretisation, which was performed for all variables using the second order MARS scheme (CD Ltd., 2001). Solutions were considered converged when all variables in the cell at the location of maximum axial velocity had become constant at double machine precision.

For the current simulation, an $r \times \theta = 35 \times 80$ cross-sectional mesh was used, which was shown to be independent for developed power-law ($n = 0.2 - 2$) flow and heat transfer (please refer to appendix B). In the axial direction 100 cells were employed, with gradual cell refinement from the inlet (1/20 L, 30 cells) and towards the exit (1/20 L, 10 cells).

Due to gelatinisation and the associated rapid changes in apparent viscosity, gradients in velocities, temperatures and other scalars are generally

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³[www.cd.co.uk](http://www.cd.co.uk)
greater than in Newtonian flow. For a representative case ($\bar{w} = 1 \text{ m/s}, D = 1 \text{ m}$), an $r \times \theta \times z = 70 \times 160 \times 100$ and an $r \times \theta \times z = 35 \times 80 \times 200$ mesh were tested. The standard and the refined meshes yielded virtually identical results regarding developing bulk and minimum temperatures, maximum axial velocity and viscosity. The $r \times \theta \times z = 35 \times 80 \times 100$ mesh was therefore used for all simulations.

4.4 Validation of the simulations

Developed flow and heat transfer in mildly curved ($d/D < 0.1$) flow can be characterised by the Dean ($De$) and Prandtl ($Pr$) number (Shah and Joshi, 1987). Unequally distributed centrifugal forces induce a symmetric secondary flow (Dean vortices, figure 4.5 middle), which may distort the axial velocity and temperature field (figure 4.5 left & right). Curved Newtonian developed friction factors and Nusselt numbers agreed well with experimental correlations recommended in the literature (please refer to chapter 2).

However, it should be stressed that due to the gelatinisation and the consecutive shear degradation, the flow is continuously developing, both hydrodynamically and thermally. Validation of flow and temperature fields, and of integrated quantities such as friction factor and heat transfer coefficients is therefore difficult.

![Figure 4.5: Examples of the axial velocity (left), the secondary flow vectors (middle) and temperature (right) for a heated, Newtonian flow ($De = 100$).](image)

4.5 Results and discussion

For gelatinising suspensions, complex flow and temperature fields are found, as the apparent viscosity varies dramatically in the cross-section. Figure 4.6
shows the developing axial velocity, the strength of the secondary flow, and
the temperature for the model food in a straight and a curved heater.

In the inlet of a straight heater, the temperature near the wall rapidly
exceeds the gelatinisation temperature and the fluid becomes very viscous,
whereas the ungelatinised core remains thin and is strongly accelerated. This
phenomenon is referred to as channeling, and it results in slow heating, and a
wide residence time distribution of the food, implying simultaneous over- and
underprocessing. Due to the continuously changing axial velocity distribu-
tion, a small, radially directed secondary flow is present in the cross-section.

Curvature induces cross-sectional mixing, especially in the entrance region
where the suspension is still thin. Bulk temperature rises more rapidly, and
the temperature distribution in the cross-section is more uniform, resulting in
a more uniform gelatinisation and a smaller residence time distribution. As
the food gelatinises slowest in the low temperature regions, viscosity remains
low at the two temperature minima characteristic of curved tube heating.
Hence the initial velocity maximum splits in two, as soon as the temperature
field has sufficiently developed. Further down the heater, all product has
gelatinised, and the flow and temperature fields resemble those of constant
property pseudoplastic power-law fluids. However, shear degradation upon
gelatinisation slowly decreases the viscosity, and the strength of the secondary
flow is seen to increase near the end of the heater.

4.5.1 Impact of curvature

Curvature effects in loosely coiled tubes are commonly correlated using the
Dean number $De = Re \sqrt{d/D}$. In chapter 5 the coil diameter is fixed at 1
m, and the Dean number is set using the apparent viscosity in the Reynolds
number $Re$. In this chapter, the initial Dean number depends on the coil
diameter and the average fluid velocity. Due to gelatinisation and the chang-
ing apparent viscosity, $De$ continuously varies along the heater. Moreover,
the meaning of a mass averaged Dean number in the cross-section is unclear,
and it can therefore only be used in a qualitative sense.

Figure 4.7 shows a considerably more rapid increase in bulk temperature
for decreasing coil diameters, which is due to the greater strength of the
secondary flow. Raw suspensions start out thin ($\mu_{init} = 0.05$ Pa s), and
the initial Dean numbers are high ($De = 50, 79$ and $158$), but as a result
of gelatinisation, the viscosity quickly rises. This is apparent in the falling
Nusselt numbers beyond $L = 10$ m when the secondary circulation slows
down. Though the flow gelatinises and permanently develops in the straight
($D = \infty$) heater as well, both temperature and heat transfer are similar to
those in constant property power-law flow.

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Figure 4.6: Paired comparison of straight ($D = \infty$ m, upper) and curved ($D = 1$ m, lower) axial velocity (top), strength of the secondary flow $v_s = \sqrt{v_r^2 + v_\theta^2}$ m/s (middle), and temperature (bottom); $\overline{w} = 1$ m/s.
Figure 4.7: Bulk temperature (top) and heat transfer (bottom) for varying curvature and $\overline{w} = 1$ m/s.

The minimum temperature and the maximum normalised axial velocity in figure 4.8 underscore the channeling occurring during gelatinisation in straight ($D = \infty$ m) heater flow. The minimum temperature at the centerline remains below the gelatinisation temperature for the entire length of the heater. Hence, fluid at the centerline does not gelatinise, whereas fluid closer to the heated wall does. Viscous fluid near the wall slows down, and the thin core
Figure 4.8: Minimum fluid temperature (top) and maximum normalised axial velocity (bottom) for varying curvature and $\overline{w} = 1$ m/s.

flow is strongly accelerated. Near the exit of the heater, more fluid gelatinises and the viscosity and velocity differences decrease. Finally, most fluid has gelatinised and the velocity distribution resembles that of developed pseudo-plastic flow.

Curvature ($D = 2.5, 1, \& 0.25$ m) induces a strong cross-sectional mixing in the entrance region, and minimum temperatures rise much more rapidly. Differences in degree of gelatinisation and viscosity are smaller, and all fluid
has fully gelatinised well before the exit of the heater. Though the velocity maximum for straight and curved flow (except for the $D = 0.25$ m case) occurs at roughly the same axial location ($z = 25 - 30$ m), curved maxima are considerably lower and assume quasi-developed values more rapidly. In curved flow residence time distributions are therefore narrower.

Figure 4.9: Relative viscosity for varying curvature and $\overline{\nu} = 1$ m/s.

Apart from gelatinisation kinetics, shear destruction was incorporated in the current model. To separate the immediate from the cumulative impact of shear rate and temperature on viscosity, the increase in non-equilibrium apparent viscosity was divided by the increase in equilibrium apparent viscosity (no time-lag and shear destruction) at equal shear rate. This yields the mass averaged relative viscosity $\overline{\mu}_{\text{rel}}$ (equation 4.16).

$$\overline{\mu}_{\text{rel}} = \frac{\int_A (\mu(t, \gamma, T) - \mu_0)/{(\mu(T) - \mu_0)} w \rho \, dA}{\int_A w \rho \, dA} \cdot 100\% \quad (4.16)$$

The relative viscosity is a measure of the lag of gelatinisation relative to the equilibrium process before complete gelatinisation. It indicates the extent of shear destruction in the post-gelatinisation phase. When all classes have gelatinised, the relative viscosity peaks (figure 4.9), and an asymptotic decay resulting from shear destruction is apparent. Shear destruction occurs as soon as the suspensions starts gelatinising, but initially its detrimental effect is largely eclipsed by the rapid viscous increase. However, the peak relative viscosity is a measure of shear destruction during gelatinisation.
Peak relative viscosity is both higher and attained faster for increasing curvature. Friction factors are higher in curved than in straight tubes as a result of increased local shear rates on the outside of the coil that are not compensated for by decreases on the inside. The current model implies a quadratic relationship between shear rate and shear destruction (equation 4.11), and shear destruction per unit tube length should therefore increase for stronger curvature. Moreover, temperatures ramp faster and gelatinisation proceeds more rapidly, increasing viscosity and shear destruction. However, the detrimental effect of shear rates and viscosity is off-set by the shorter exposure to shear stresses while gelatinising. Retarded gelatinisation resulting from high heating rates in fact increases the eventual viscosity, as structural breakdown only starts after substantial gelatinisation and viscous increase. The higher maximum relative viscosities therefore imply a potentially lower starch consumption in curved heaters.

4.5.2 Impact of velocity

To assess the impact of velocity on curved gelatinising flow, two additional velocities ($\bar{u} = 0.5$ and $2$ m/s) were simulated at a fixed coil diameter $D = 1$ m. Curvature is now affected by $Re$ in the Dean number $De = Re\sqrt{d/D}$. Simultaneously the average residence time doubles or halves.

Bulk temperatures rise more slowly for increasing fluid velocities (figure 4.10). As the average temperature difference between the wall and the bulk is larger for higher velocities, and the local heat transfer is higher due to the increased curvature, this is a result of the shorter residence time only. Minimum temperatures (figure 4.11) are also lower, with the exception of the entrance region. Here the highest flow velocity ($\bar{u} = 2$ m/s) induces a secondary flow sufficiently strong to temporarily raise the minimum temperature above that of the other flow rates.

Figure 4.11 underscores the importance of curvature to reduce channeling, as the maximum normalised axial velocity in the gelatinising region in the entrance is lower for higher flow velocities. Further down the heater however, the highest flow rate exhibits the highest value of $u^+/\bar{u}$. This is due to the reduced pseudoplasticity of the fluid above the critical shear rate $\gamma_c$ as defined in equation 4.15. For higher flow rates, more fluid will be in this regime, and the velocity profile approaches the Newtonian parabolic profile. The simultaneous apparent increase in pseudoplasticity near the wall as a result of the greater local shear destruction, is of minor importance.

The developing relative viscosity is shown in figure 4.12. Higher flow velocities shift the maximum $\bar{\eta}_{gel}$ down the heater, and this maximum is also lower, despite the shorter exposure to shear stresses. This can be explained
Figure 4.10: Bulk temperature (top) and heat transfer (bottom) for varying velocity and $D = 1$ m.

from the quadratic dependence of $\Phi$ on the shear rate (equation 4.11), and from the generally greater shear stresses, exceeding $\tau_g$ of the gelatinisation classes in a greater part of the cross-section. If no other considerations are taken into account, low flow velocities are recommended to minimise starch consumption and heater length.
Figure 4.11: Minimum fluid temperature (top) and maximum normalised axial velocity (bottom) for varying velocity and $D = 1 \text{ m}$.

### 4.6 Conclusions

We simulated a model food derived from a 5\% w/w cross-linked waxy maize suspension as it gelatinised in a curved heater. An existing thermo-rheological model was adapted and extended to the high heating and shear rates common in industrial tubular heaters.
Figure 4.12: Relative viscosity for varying velocity and $D = 1$ m.

Channeling, or an extremely broad residence time distribution implying simultaneous over- and underprocessing of the product proved a serious problem in straight heaters. However, moderate curvature strongly reduces this phenomenon. Heating and gelatinisation rates are highest at a maximum degree of curvature, that is, in tightly wound coils. At minimum coil diameter, the increase in relative viscosity is also highest, implying a minimum starch consumption to reach a target viscosity.

At fixed coil diameter, higher flow velocities prompt longer heaters, and the maximum attainable relative viscosity decreases. To obtain low shear degradation and the most economical use of starch, low product velocities are recommended.
Bibliography


H-J. Liao, M.A. Rao, and A.K. Datta. Role of thermo-rheological behaviour in


Chapter 5

Power-law foods in continuous coiled sterilisers

The heating, holding and cooling sections of the high temperature region \((T > 100^\circ C)\) of coiled UHT-sterilisers were simulated 3-dimensionally to assess the impact of centrifugal and buoyant forces on viscous power-law food products. For a wide range of curvature \((De = 0 \text{ – 500})\), heat transfer, lethality development and thiamine concentration were analysed.

Curvature and pseudoplasticity substantially increase heat transfer and render the axial velocity profile more uniform. As a result, processing times in heater, holder and cooler can be shorter, thus greatly improving thiamine retention. Significant lethality accumulates in the heating section, but the cooling section contributes very little to product sterility. Buoyant forces were shown to be of minor importance in coiled steriliser flow at the current process conditions.

5.1 Introduction

Continuous UHT-sterilisers are widely used in the food industry to ensure microbial safety of liquid foods ranging from milk and fruit juices to sauces and custards. Continuous sterilisation holds a major advantage over batch sterilisation (e.g. cans) in its better uniformity of thermal treatment. Characteristic diameters are smaller, and internal heat transfer is higher due to forced

convection, as opposed to conduction and free convection only. For a given heating rate, temperature differences in the food are therefore smaller, and processing temperatures can be higher without risking unacceptable changes in colour and taste in the highest temperature regions. Ultra High Temperature (UHT) sterilisation strongly favours microbial and sporidical inactivation relative to degradation of quality factors such as colour and vitamin concentration. Combined with the application of short processing times, UHT-sterilisation yields safe products of a high quality (Holdsworth, 1992).

However, when UHT-sterilisation is applied to viscous products, the associated laminar flow and temperature profiles induce a strongly non-uniform heat treatment. Poor heat transfer and broad residence time distributions result in fluid elements with very different thermal histories. This reflects in a non-uniform distribution of lethality and quality factors.

Several studies investigate the heat treatment of viscous foods in tubular sections of continuous UHT-sterilisers. Simpson and Williams (1974) presented the first non-isothermal results for lethality and thiamine retention for a pseudoplastic fluid in a steriliser consisting of a heater and a cooler. Kumar and Bhattacharya (1991) analysed lethality and thiamine retention in the developing flow of a pseudoplastic food with a temperature dependent viscosity. Jung and Fryer (1999) simulated a pseudoplastic model food with a temperature dependent rheology in a heating-, holding- and cooling section of a steriliser. Liao, Rao, and Datta (2000) studied lethality and thiamine retention in a heater and a cooler by solving flow and temperature fields of a gelatinising starch suspension. Bhamidipati and Singh (1994) applied the concept of thermal time distribution to shear thinning tomato juice with a temperature dependent rheology in a tubular heater. All authors stress the importance of the uniformity of heat treatment, and for microbiologically safe products at maximum thiamine retention they recommend the smallest practical tube diameter.

Though some authors recognise the potential importance of 3 dimensional effects such as curvature (Kumar and Bhattacharya, 1991) and free convection (Bhamidipati and Singh, 1994; Jung and Fryer, 1999; Liao et al., 2000), these studies regard the flow to be 2-dimensional. Secondary flows result from curvature, as non-uniform centrifugal forces induce Dean-vortices (Dean, 1927) (figure 5.3, top). As a result, cross-sectional mixing occurs, decreasing residence time distributions (Saxena and Nigam, 1984) while substantially increasing heat transfer (Shah and Joshi, 1987). Density differences between the bulk and the wall induce Morton-vortices (Morton, 1959) (figure 5.3, middle) which promote cross-sectional mixing and increase heat transfer (Morcos and Bergles, 1975). When curvature and buoyancy forces are both significant, a mixed flow regime can be observed (figure 5.3, bottom).
Both Morton and Dean vortices are expected to render the UHT-treatment more uniform and effective. As a result of smaller residence time distributions and increased heat transfer, the thermal history will more closely resemble a square wave function, making optimal use of the UHT-strategy.

This chapter reports simulations of the 3-dimensional effects of curvature and buoyancy on the performance of a continuous UHT-steriliser. For viscous power-law foods, heat transfer, lethality development and nutrient retention will be evaluated for the heating, holding and cooling section of the high temperature part of the steriliser.

### 5.2 Problem analysis

#### 5.2.1 Physical model

Viscous products were simulated in the heating, holding and cooling section shown in figure 5.1. Having the greatest impact on sterility and product properties, the high temperature region \( T > 100 \, ^\circ C \) was chosen.

![Figure 5.1: Geometry of the heating, holding and cooling section.](image)

Product enters the heating section \( (L_{heat} = 200 \, m) \) at \( T = 100 \, ^\circ C \) and is heated indirectly by steam, resulting in a constant wall temperature \( T_w = 145 \, ^\circ C \). Immediately after, the fluid is assumed thoroughly mixed and led into an isothermal holding section \( (L_{hold} = 3 \, m) \) kept at \( 140 \, ^\circ C \). Next the fluid enters the cooling section \( (L_{cool} = 200 \, m) \) where it is cooled by a constant wall temperature of \( 95 \, ^\circ C \). For all sections a pipe diameter \( d = 0.025 \, m \) and an average axial fluid velocity \( \overline{\nu} = 0.5 \, m/s \) were adopted. For the curved or mixed flow cases the steriliser sections are coils with a diameter \( D = 1 \, m \), of which the axis is aligned with the vector of gravity.
5.2.2 Governing equations and boundary conditions

The equations of continuity, motion and energy in Cartesian coordinates for steady, incompressible power-law flow are given by equations 5.1 to 5.3.

\[ \nabla \cdot \mathbf{u} = 0 \]  \hspace{1cm} (5.1)

\[ \rho (\mathbf{u} \cdot \nabla) \mathbf{u} = -\nabla p + \mu \nabla^2 \mathbf{u} + \mathbf{F} \]  \hspace{1cm} (5.2)

\[ \rho c_p \mathbf{u} \cdot \nabla T = \lambda \nabla^2 T + \mu \Phi \]  \hspace{1cm} (5.3)

Thiamine concentrations are computed using the transport equation for passive scalars, given by equation 5.4.

\[ \rho (\mathbf{u} \cdot \nabla) \phi = \rho \nabla \cdot (\nabla^2 \phi) + S_\phi \]  \hspace{1cm} (5.4)

Centrifugal and buoyant body forces enter the model through terms \( \mathbf{F} \) in the equations of motion. In the case of loosely coiled pipes \((d/D \leq 0.1, \text{Baurmeister and Brauer (1979)})\) higher order effects are neglected and the impact of curvature on the flow and temperature field can be included by the term \( F_c \) (equation 5.5) acting in the x-direction as shown in figure 5.2.

\[ F_c = \rho \frac{u^2}{x} \]  \hspace{1cm} (5.5)

For isobaric density changes that have no significant effect on the flow field, buoyancy forces can be included in the y-momentum equation using the term \( F_b \) (equation 5.6).

\[ F_b = -g \cdot \rho \frac{1}{1 + \beta (T - T_0)} \]  \hspace{1cm} (5.6)

Current simulations do not account for coil pitch. Experimental work of Mishra and Gupta (1979) only attributes importance to torsional forces for very high values of curvature and for a coil pitch exceeding the coil diameter. In a recent experimental study, Xin and Ebadian (1997) could find no influence of pitch on the developed heat transfer.
Viscous dissipation enters the energy equation 5.3 through the term $\mu \Phi$ (Bird, Stewart, and Lightfoot, 1960). For the highest viscosity $\mu \Phi$ is important, as in this case the fluid temperature may rise several degrees due to dissipation. The source term $S_\theta$ in the scalar transport equation can be derived from the denaturation kinetics of thiamine which will be discussed in section 5.2.4.

![Coordinate systems and direction of forces](image)

**Figure 5.2:** Coordinate systems and direction of forces.

The inlet boundaries for heater, holder and cooler are fully developed power-law velocity profiles and uniform temperature and scalar quantities. At the wall the no-slip condition and a constant temperature are imposed, and the wall is assumed to be impermeable to scalar species. At the outlets static pressure as well as all axial gradients are set to zero. Except in cases involving buoyancy, planar symmetry has been employed.

### 5.2.3 Fluid properties

Many viscous liquid foods can be adequately described by an Ostwald-de Waele or power-law model (Holdsworth, 1993; Lagarrigue and Alvarez, 2001). For 3-dimensional flows the apparent viscosity $\mu$ depends on power-law index $n$, consistency index $m$, and the magnitude of the rate of strain tensor $\Delta_{ij}$ (Bird, Stewart, and Lightfoot, 1960).

$$
\mu = m \left| \frac{1}{2} \sum \sum \Delta_{ij} \Delta_{ij} \right|^{\frac{n-1}{2}}
$$

(5.7)

Simulations were carried out for pseudoplastic ($n = 0.5$), Newtonian ($n = 1$) and dilatant ($n = 1.5$) fluids. For each combination of $n$ and $De$, $m$ was
calculated from the apparent viscosity $\mu_a$ for straight pipe flow. Density $\rho = 1000$ kg/m$^3$, heat capacity $c_p = 4180$ J/(kg °C), thermal conductivity $\lambda = 0.5$ W/(m °C) and the volumetric expansion coefficient $\beta = 0.0009$ 1/°C were assumed constant.

### 5.2.4 Reaction kinetics

 Destruction of spores can be adequately described by first order kinetics, either through a Bigelow (Bigelow, 1921) or an Arrhenius model (Holdsworth, 1993). However, the Arrhenius model is considered to be more accurate in the very high (100 - 150°C) temperature range of the UHT-treatment (Hallström, B. and Skjödebrand, C. and Trägårdh, C., 1988) and will be used in this work. Kinetic parameters $k_0 = 2 \cdot 10^{10}$ 1/s and $E_a = 310.1$ kJ/mol for deactivation of spores of Clostridium Botulinum are those employed by Simpson and Williams (1974).

The concentration of spores $N$ for an initial concentration $N_{\text{init}}$ and a variable temperature is given by equation 5.8.

$$N = N_{\text{init}} e^{-\int_0^t k_0 e^{-\frac{E_a}{RT(t)}} dt}$$  \hspace{1cm} (5.8)

For a constant reference temperature $T_{\text{ref}}$ equation 5.8 simplifies to:

$$N = N_{\text{init}} e^{-\left( k_0 e^{-\frac{E_a}{T_{\text{ref}}}} \right) t}$$  \hspace{1cm} (5.9)

The equivalent lethality $F_0$ is the time at $T_{\text{ref}}$ having the same impact on spore concentration as the actual variable temperature heat treatment. Substitution of $F_0$ for $t$ in equation 5.9 and solving for $F_0$ from equation 5.8 and 5.9 yields:

$$F_0 = \int_0^t e^{\frac{E_a}{R T(t)}} \left( \frac{1}{T_{\text{ref}}} - \frac{1}{T(t)} \right) dt$$  \hspace{1cm} (5.10)

Conform practice $T_{\text{ref}}$ is set to 250 F or 121.1°C. In a continuous steriliser the axial coordinate $z$ and time $t$ are coupled by the axial velocity $w$.

$$d z = w \, dt$$  \hspace{1cm} (5.11)
Differentiation of equation 5.10 and substitution of $dt$ from equation 5.11 yields the lethal rate.

$$\frac{dF_0}{dz} = e^{\frac{E_p}{RT}} \left( \frac{1}{\tau_{\text{ref}}} - \frac{1}{\tau(z)} \right)$$

(5.12)

For all computations the steriliser coils are divided in axial computational sections. Both temperature and velocity vary in each cross-section and therefore $dF_0/dz$ will not be constant. As a conservative estimate of the lethality $\Delta F_0$ produced in each axial section, the minimum value of $dF_0/dz$ in the cross-section is used.

$$\Delta F_0 = \Delta z \frac{dF_0}{dz} \bigg|_{\text{min}}$$

(5.13)

Summation over the axial elements of interest yields the minimum axial $F_0$ value, which will be referred to as lethality.

$$F_0(z) = \sum \Delta F_0$$

(5.14)

In the absence of buoyant and centrifugal forces velocity and temperature profiles are axi-symmetric. Equations 5.12 to 5.14 then reduce to the concept of the cold centerline in the heater.

$F_0$ is a conservative estimate of lethality as the existence of a fluid element is assumed that receives a minimum heat treatment in every computational section of the steriliser. However, streamlines resemble corkscrews in curved and buoyant flow, and fluid elements experience a fluctuating axial velocity and temperature. To assess the true sterility, spore concentrations should be computed as transported scalars. Unfortunately, the very large change in spore concentration (at least 11.5 decimal reductions) proved beyond the accuracy of the CFD software.

Retention of quality factors is modelled by the vitamin thiamine, as its sensitivity to temperature is very similar to that of other vulnerable constituents (Holdsworth, 1992). Thiamine concentrations are calculated using the scalar transport equation 5.4 and the sink term $S_0$ given by equation 5.15. According to Horak and Kessler (1981), destruction of thiamine must be modelled as a 2nd order reaction for conversions exceeding 10%. This is clearly the case in the current simulations, where laminar heat transfer leads to long processing times and substantial thiamine losses.
\[ S_\phi = -\frac{k_0}{\phi_{in}} \cdot e^{-\frac{E_a}{RT} \phi^2} \] (5.15)

The initial thiamine concentration \( \phi_{in} \) is assumed to be 40 mg/l for heater, holder and cooler, the kinetic constants are given by \( k_0 = 1.02 \cdot 10^{10} \) 1/s and \( E_a = 101.4 \) kJ/mol (Horak and Kessler, 1981).

Spores and thiamine are assumed to undergo convective transport only (\( \mathcal{D} = 0 \)). According to the Wilke-Chang method (Reid, Prausnitz, and Poling, 1988) the diffusion coefficient \( \mathcal{D} \) of thiamine is on the order of \( 10^{-9} \) m\(^2\)s\(^{-1}\) or smaller, yielding Peclet numbers on the order of \( 10^4 \) or more. Diffusion of the larger spores is of course slower.

### 5.3 Numerical considerations

Solutions to equations 5.1 - 5.4 have been approximated using the Star finite volume package\(^2\). The SIMPLE algorithm was employed to solve the coupled equations after discretisation, which was performed for all convective fluxes using the third order QUICK scheme. Solutions were considered converged when all variables in a cell at the tube axis in the outlet of the section had become constant at double machine precision.

Mesh independence in the \( r \) and \( \theta \) direction was established for Newtonian flow by computing developed friction factors and Nusselt numbers for several increasingly refined meshes and for several values of the Dean number. A butterfly mesh with 35 unequally spaced radial cells and 80 equally spaced tangential cells was considered independent (within 1.5 \% of independent solution) in all cases.

Mesh independence in the \( z \) direction was established by varying the axial number of cells of the heater, and comparing the axial development of the two most sensitive quantities: average bulk temperature and the lethality \( F_0 \). In the axial direction 40, 80 and 160 cells were taken, using the cross-sectional discretisation established in the developed case. The simulations proved remarkably insensitive to the axial discretisation, as 40, 80 and 160 axial cells yielded virtually identical results.

Based on the refinement studies a \((r \times \theta \times z) = (35 \times 80 \times 80)\) mesh was selected for the heating and cooling sections. To resolve entrance effects, in the first 1/20 of the section (30 cells) the axial length increased according to a fixed ratio. In the remaining developed part of the section (50 cells), the

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\(^2\text{www.cd.co.uk}\)
axial length was constant. For the holding section an identical cross-sectional discretisation and 60 equidistant axial cells were employed.

5.4 Validation of the simulations

Developed flow and heat transfer in loosely coiled and purely buoyant flows can be characterised by a combination of the Dean ($De$) and Prandtl ($Pr$) number, and the Grashof ($Gr$) and Prandtl number respectively (Shah and Joshi, 1987). Examples of the laminar flow and temperature fields in a heated tube in the curved, buoyant and mixed regime are shown in figure 5.3. The curvature of the coil and the direction of the vector of gravity are as in figure 5.2. In figure 5.3 red indicates a high velocity or temperature, and blue a low.

Newtonian friction factors agree excellently with the experimental correlation of Mishra and Gupta (1979) as recommended by Gnielinski (1986). Curved heat transfer (figure 5.4) is in good agreement with the hybrid experimental numerical correlation of Manlapaz and Churchill (1981), as recommended by Ebadian and Dong (1998). Inclusion of buoyant body forces results in a slight increase in heat transfer, but does not affect friction factors.

A further quantity of interest is the maximum axial fluid velocity $w^+$, as it governs the minimum residence time in the steriliser. Figure 5.5 shows $w^+ / \bar{w}$ as a function of curvature and rheology. For straight pipe flow, total pseudoplasticity ($n = 0$) yields plug-flow ($w^+ / \bar{w} = 1$), whereas total dilatancy ($n = \infty$) yields a triangular velocity profile with $w^+ / \bar{w} = 3$. The simulations agree excellently with the analytical maximum velocities for straight pipe flow ($De \rightarrow 0.1$), and both curvature and pseudoplasticity are seen to make the axial velocity profile more uniform.

5.5 Results and discussion

5.5.1 Analysis of the heating section

Curved axial velocity fields are distorted by centrifugal forces when compared to straight flow. The velocity maximum is lower and it shifts to the outside of the coil (figure 5.3). Fluid properties are assumed independent of temperature so the axial velocities are constant once the flow has hydrodynamically developed. Hydrodynamic entrance length in curved flow is very short compared to the total length of the heater.

The secondary motion in the cross-section splits the single temperature minimum of straight flow into two minima for curved flow, which are located
away from the centerline (figure 5.3). This occurs even for the lowest simulated curvature. The single thiamine maximum of straight flow is also split into two maxima away from the tube center. Though the rate of thiamine destruction is a function of temperature only, these maxima do not coincide with the minima in temperature, as the thiamine concentration also depends on residence time, and hence axial and secondary velocity.
Figure 5.4: Developed heat transfer for curved and mixed flow.

Figure 5.5: Normalised maximum axial velocity for curved flow.

5.5.1.1 Impact of curvature

Bulk temperature, Nusselt number, lethality and thiamine concentration may serve to evaluate the quantitative impact of curvature on Newtonian flow. Figure 5.6 shows bulk temperatures rising much more rapidly when centrifugal forces are present, even for the lowest curvature $(De = 1)$. Axial
temperature in hydrodynamically developed but thermally developing flow in a straight ($De = 0$) pipe (the Graetz-Nusselt problem) is indistinguishable from the analytical results presented by Shah and London (1978). Curvature increases both local and asymptotic Nusselt numbers (figure 5.6) and accelerates thermal development. In the entrance region, developing flow and temperature fields cause the Nusselt number to 'oscillate', a phenomenon experimentally observed by e.g. Janssen and Hoogendoorn (1978).
For a straight heater, the minimum lethal rate (equation 5.12) is governed by the cold centerline where the axial velocity is highest and the temperature is lowest. Curvature increases heat transfer and depresses the maximum axial velocity (figure 5.5), rendering both temperature and velocity field more uniform. As a result lethality (figure 5.7) is higher in coiled flow, even for weak curvature ($De = 1$). For strong curvature ($De = 500$) and large axial distances ($L > 150$ m), the bulk temperature approaches the wall temperature, and lethality increases linearly. Though higher temperatures favour sterilisation, they induce greater losses of thiamine. As a result, the normalised thiamine concentrations are significantly lower for increasing degrees of curvature (figure 5.7).

5.5.1.2 Impact of rheology

The impact of rheology on straight and curved heaters is significant. Especially lethality (figure 5.8) increases as axial temperatures rise more rapidly in pseudoplastic flow. Pseudoplasticity also enhances lethality as it renders the axial velocity more uniform (figure 5.5). Thiamine concentrations are not very sensitive to rheology. For a given curvature, a pseudoplastic fluid exhibits the greater loss in thiamine, which is largely due to higher bulk temperatures.

5.5.1.3 Impact of buoyancy in mixed flow

Buoyant forces necessitate modelling of the entire tube cross-section. Though the flow is dominated by centrifugal forces and there is little qualitative change in the solution fields, buoyancy can have a subtle influence.

Buoyancy results in slightly higher heating rates (figure 5.9), which can be explained by an intermediate region in the heater where buoyant forces actually increase local heat transfer. However, except for the $De = 10$ case, lethality is lower (figure 5.10). As buoyancy does not significantly affect the axial velocity profile, this is due to asymmetry between the two temperature minima, one of which deepens. Since buoyant heating rates are higher and axial velocity profiles are unaffected, inclusion of buoyancy leads to a minor increase in thiamine destruction.

5.5.1.4 The performance of a coiled heater

Curvature increases heating rates and promotes sterilisation, but at the same time causes greater losses in thiamine. To evaluate the performance of straight and coiled heaters, lethality and thiamine concentrations are calculated when the bulk temperature has reached $140^\circ$C. In table 5.1 the required
heater lengths, and the resulting $F_0$ values and thiamine concentrations are given for all cases considered.

Because of improved heat transfer rates, curvature decreases the length required for the heating section. The resulting shorter process time in the heater enables the retention of more thiamine. Pseudoplasticity also decreases the heater length and increases thiamine retention through the combined action of a higher heating rate and a more uniform velocity profile.
Figure 5.8: Lethality in curved non-Newtonian heater flow.

Figure 5.9: Bulk temperature in mixed Newtonian heater flow.

Free convection (denoted by + in table 5.1) slightly reduces heater length, but at the same time decreases sterility and thiamine concentration. The lethality accumulated in the heater is quite significant, but for certification purposes only lethality produced in the holder is taken into account.
Figure 5.10: Lethality in mixed Newtonian heater flow.

Table 5.1: Impact of curvature, rheology and buoyancy on the heater ($\overline{T} = 140\, ^\circ C$).

\[
\begin{array}{cccccccccc}
De & n = 0.5 & & n = 1 & & n = 1.5 & \\
    & L & F_0 & \phi_n & L & F_0 & \phi_n & L & F_0 & \phi_n \\
0   & 311.1 & 1832 & 68.1 & 333.7 & 1692 & 67.4 & 343.6 & 1599 & 67.2 \\
1   & 153.0 & 1302 & 79.6 & 164.6 & 1256 & 78.5 & 168.3 & 1218 & 78.2 \\
& + 152.8 & 879 & 79.3 & 164.4 & 1082 & 78.3 & 168.2 & 1097 & 77.9 \\
10  & 136.6 & 1039 & 80.4 & 145.3 & 1004 & 79.1 & 145.9 & 924 & 78.9 \\
& + 135.6 & 975 & 80.1 & 141.6 & 933 & 79.1 & 141.8 & 880 & 78.8 \\
100 & 110.0 & 727 & 82.8 & 113.9 & 794 & 82.4 & 116.6 & 807 & 82.0 \\
& + 106.2 & 511 & 82.8 & 109.5 & 509 & 82.4 & 111.3 & 482 & 82.1 \\
500 & 54.1 & 453 & 90.6 & 61.7 & 745 & 89.8 & n.a. & n.a. & n.a. \\
& + 52.0 & 404 & 90.7 & 61.7 & 487 & 89.5 & n.a. & n.a. & n.a. \\
\end{array}
\]
5.5.2 Analysis of the holding section

Similarly to the heater, curvature distorts the axial velocity field in coiled holder flow. Due to its much shorter length however, the hydrodynamic entrance length can be a significant part of the holder. As the temperature is uniform and constant, buoyancy effects are absent and the location of minimum lethal rate coincides with the velocity maximum (equation 5.12). For curved flow, the thiamine concentration exhibits two symmetric maxima resulting from the interplay of secondary flow and the axial velocity.

5.5.2.1 Impact of curvature and rheology

In developed holder flow, the lethal rate is inversely related to the maximum axial velocity, and lethality (figure 5.11) increases linearly with axial distance. Hydrodynamic entrance effects are apparent in the lethal rate (not shown) for the highest degrees \((De \geq 100)\) of curvature, but largely vanish when integrating into the lethality. Curvature and pseudoplasticity both increase lethal rate and lethality. As the maximum axial velocities for \(De = 0\) and \(De = 10\) are approximately equal, lethality is the same for equal value of \(n\). Rheology clearly has the greater impact on lethality.

![Figure 5.11: Lethality in curved power-law holder flow.](image)

Differences in residence time distribution resulting from curvature or rheology have virtually no impact on the thiamine concentrations.
5.5.2.2 The performance of a coiled holder

Following Simpson and Williams (1974), 11.5 decimal reductions in spore concentration of Clostridium Botulinum are required, equivalent to a processing time $F_0 = 165$ s. As the temperature is constant, $F_0$ is only affected by the maximum axial velocity, which in turn is controlled by rheology and curvature. In table 5.2, the required lengths and resulting thiamine concentrations are given.

Table 5.2: Impact of curvature and rheology on the holder ($F_0 = 165$ s).

<table>
<thead>
<tr>
<th>$De$</th>
<th>$n = 0.5$</th>
<th>$n = 1$</th>
<th>$n = 1.5$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$L$ $\phi_n$</td>
<td>$L$ $\phi_n$</td>
<td>$L$ $\phi_n$</td>
</tr>
<tr>
<td>0</td>
<td>1.79 99.46</td>
<td>2.15 99.36</td>
<td>2.36 99.29</td>
</tr>
<tr>
<td>1</td>
<td>1.79 99.46</td>
<td>2.15 99.36</td>
<td>2.36 99.29</td>
</tr>
<tr>
<td>10</td>
<td>1.79 99.46</td>
<td>2.15 99.35</td>
<td>2.36 99.29</td>
</tr>
<tr>
<td>100</td>
<td>1.70 99.48</td>
<td>1.90 99.42</td>
<td>2.02 99.39</td>
</tr>
<tr>
<td>500</td>
<td>1.69 99.49</td>
<td>1.88 99.43</td>
<td>n.a. n.a.</td>
</tr>
</tbody>
</table>

Depression of the maximum axial velocity for curved and pseudoplastic flow shortens the holding section and improves thiamine retention. However, thiamine losses in the holder are already very small.

5.5.3 Analysis of the cooling section

In the absence of buoyancy, axial velocity fields in the cooler are identical to those in the heater for equal curvature and rheology. Furthermore, developing temperature and thiamine fields in the cooler are the inverse of those in the heater, with temperature minima replaced by maxima, and thiamine minima replaced by maxima. Only lethality is qualitatively different from its counterpart in heater flow: as fluid near the wall is cooled quite rapidly, the minimum lethal rate is now located close to the wall.

Centrifugal forces strongly promote heat transfer, and bulk temperatures drop more rapidly for increasing degrees of curvature (figure 5.12). Rapid cooling near the wall and the strong dependence on temperature of lethal rate result in minimal accumulation of lethality, even in straight tube flow (figure 5.13). Some authors (Kumar and Bhattacharya, 1991; Jung and Fryer,
attribute a further increase of lethality to the cooler, and regard it to be an additional safety factor. However, their conclusion is based on the assumption that in the cooler the centerline accumulates the least lethality. Though this is true for the heater, in the cooler the high temperature in the core is the overriding factor.

\[
\begin{align*}
\text{Figure 5.12: Bulk temperatures in curved Newtonian cooler flow.}
\end{align*}
\]

Of course, lower temperatures slow down reaction rates and increasing curvature better retains thiamine. Comparing figures 5.7 and 5.13 reveals the higher thiamine loss in the heater: in the heater the temperature asymptotically approaches a maximum, whereas in the cooler the fluid is at a relatively low temperature most of its residence time.

Rheology and buoyancy impact the cooler similarly to the heater, which is largely through their effect on heat transfer. Pseudoplastic fluids cool slightly faster and thiamine is a little better preserved. Nusselt numbers for the heater and the cooler are identical, and lethality is very small. At equal \( n \) and \( De \), buoyancy slightly promotes heat transfer and better retains thiamine. As a side effect, lethality production is further depressed.

### 5.5.3.1 The performance of a coiled cooler

Table 5.3 presents the required length of the cooler to reach an average exit temperature of 100 \(^\circ\text{C}\) as a function of curvature, rheology and buoyancy. Lethality and thiamine concentration are also given.
Figure 5.13: Lethality (top) and thiamine concentration (bottom) in curved Newtonian cooler flow.

Improved heat transfer resulting from curvature decreases the length required for the cooling section. Pseudoplasticity and buoyancy further enhance cooling, but to a smaller extent. Thiamine is better preserved but the lethality produced in the cooler is negligible.

Since cooler and heater have the same geometry and operate at identical process parameters (same difference between wall and inlet temperature, same required temperature change of the fluid), the length of heater and
Table 5.3: Impact of curvature, rheology and buoyancy (+) on the cooler ($T = 100^\circ C$).

<table>
<thead>
<tr>
<th>$De$</th>
<th>$n = 0.5$</th>
<th>$n = 1$</th>
<th>$n = 1.5$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$L$</td>
<td>$F_0$</td>
<td>$\phi_n$</td>
</tr>
<tr>
<td>0</td>
<td>350.4</td>
<td>6.0</td>
<td>87.3</td>
</tr>
<tr>
<td>1</td>
<td>170.7</td>
<td>2.0</td>
<td>92.3</td>
</tr>
<tr>
<td></td>
<td>+ 166.7</td>
<td>1.9</td>
<td>92.3</td>
</tr>
<tr>
<td>10</td>
<td>137.6</td>
<td>1.4</td>
<td>93.8</td>
</tr>
<tr>
<td></td>
<td>+ 136.3</td>
<td>1.3</td>
<td>93.9</td>
</tr>
<tr>
<td>100</td>
<td>110</td>
<td>1.1</td>
<td>95.0</td>
</tr>
<tr>
<td></td>
<td>+ 106.0</td>
<td>1.0</td>
<td>95.2</td>
</tr>
<tr>
<td>500</td>
<td>54.1</td>
<td>0.8</td>
<td>97.3</td>
</tr>
<tr>
<td></td>
<td>+ n.a.</td>
<td>n.a.</td>
<td>n.a.</td>
</tr>
</tbody>
</table>

Cooler should be the same for corresponding values of $De$ and $n$. From table 5.1 and 5.3 follows this is indeed so for $De \geq 10$, but not for the highest viscosity associated with the $De = 0$ and $De = 1$ cases. For this viscosity dissipation is significant, and internal heat generation causes a more rapid heating, or a slower cooling.

5.6 Conclusions

Simulations of the high temperature heater, holder and cooler of coiled UHT-sterilisers have clarified the 3-dimensional effects of centrifugal and buoyant forces on flow and heat transfer of viscous power-law foods.

In the heater, curvature and pseudoplasticity increase heat transfer and decrease the length of the section, resulting in a better preservation of thiamine. Depression of the maximum axial velocity and more uniform cross-sectional temperatures increase lethality, which exceeds the requirement for the holder. Including buoyant forces in curved flow slightly improves heat transfer, but due to asymmetry in the temperature field, lethality is reduced.
The length of the holder required to meet the specified lethality decreases for curved and pseudoplastic flow. Improvements in thiamine preservation are negligible however, as thiamine destruction is very small compared to the losses in heater and cooler.

In the cooler, curvature and pseudoplasticity increase heat transfer and decrease processing time, thus preserving more thiamine. Because temperatures asymptotically approach a minimum value in the cooler, thiamine destruction is much smaller than in the heater for corresponding rheology and curvature. Buoyancy slightly improves heat transfer, but lethality is negligible.
Bibliography


S. Lagarrigue and G. Alvarez. The rheology of starch dispersions at high temper-
Chapter 6

Further research

To assess the validity of the gelatinisation model, product runs are to be performed on the Stork pilot plant (Midi-Sterideal). Products and their gelatinisation and degradation properties can be characterised using the procedure outlined in chapter 3 and appendix E. As a start, tests are being prepared to validate the numerical flow and heat transfer of viscous sugar solutions in a counter current glycol cooler of the Midi-Sterideal.

The impact of curvature and rheology on lethality and nutrient degradation in the high temperature section of the steriliser can be validated using thermal time integrators (Hendrickx, Maesmans, De Cordt, Noronha, Van Loey, and Tobbback, 1995). However, such measurements are not very accurate and cumbersome to perform (Van Loey, Hendrickx, De Cordt, Haentjens, and Tobbback, 1996). A simpler check providing information on the flow conditions inside the coil is a comparison of experimental and numerical residence time distributions.

Irrespective of the accuracy of the numerical model, fluid properties of a food have to be determined by experiment. This is especially difficult for the complex and changing rheology of starch suspensions at temperatures exceeding the boiling point of water. To reduce the experimental effort on gelatinised foods, the method of apparent concentration may be employed (Härröd, 1989). Using this method a reference food is characterised for a range of temperatures, shear rates and starch concentrations. For minor changes in composition and preparation procedure, a one-point measurement on a similar food then suffices to designate this food an apparent concentration. The food exhibits the same behaviour as the reference product at this apparent concentration.
An interesting application of the 3-dimensional CFD model may be in predicting local fouling rates during heat treatments of dairy products. At low temperatures (70 $\leq T \leq$ 110 $^\circ$C) fouling of heat transfer surfaces consists mainly of a soft and spongy protein deposit. For temperatures exceeding 110$^\circ$C precipitation of hard, inversely soluble calcium phosphates dominates (Jeurnink, Walstra, and de Kruijf, 1996; Visser and Jeurnink, 1997). The polymerisation model for the aggregation of $\beta$-lactoglobulin including the activity of the casein micelles (Roefs and de Kruijf, 1994; De Jong and van der Linden, 1998), was combined with the experimental rate of deposition of activated mono- and polymer radicals (De Jong, Bouman, and van der Linden, 1992). Tentative simulations on coarse grids revealed the expected impact of curvature on the protein fouling rates in heater coils (Heijthuisen, 2002). Curvature enhances mass transfer, thus increasing protein fouling rates. However, higher temperatures also promote the deactivation of the radicals, reducing fouling rates. These preliminary results seem to justify a further exploration of the 3-dimensional flow, heat transfer and protein-fouling model of laminar, milk-based products. An extension to the quasi-steady modelling of the growing fouling layer during operation could be considered.

Finally, to cover all aspects of thermally processed single-phase gelatinising foods, the turbulent-laminar transition needs modelling. As raw starch suspensions approximately display the viscosity of water, the flow is turbulent. On gelatinising however, the viscosity strongly increases, ensuring laminar flow. Further heating lowers the viscosity, both as a result of the direct temperature dependency and of structural degradation, and the flow may return to the turbulent regime. Finally, cooling below room temperature brings the food in a very viscous laminar state. As yet, no commercially available flow and heat transfer package incorporates these transitions.

Re-laminarisation is a well known phenomenon in strongly heated and accelerated gas flows (Torri, Shimizu, Hasegawa, and Higasa, 1990; Torii and Yang, 1997; Ezato, Shehata, Kunugi, and McEligot, 1999), in polymerising flows (Grishin and Nemirovskii, 1984), in curved flows (Kalb and Seader, 1983) and in some buoyant flows (Tanaka, Maruyama, and Hatano, 1987). Several authors adapted the low Reynolds number k-$\varepsilon$ model (Jones and Launder, 1972), to predict straight tube friction factors and heat transfer, both in the laminar and the turbulent regime (Torii et al., 1990; Huiren and Songling, 1991; Torii and Yang, 1997; Ezato et al., 1999).
Preliminary simulations showed that flow and heat transfer in the laminar and low turbulent range can indeed be predicted correctly applying small modifications to the low Reynolds number models native to the Star package. However, application to the turbulent-laminar transition as a result of (a combination of) curvature and gelatinisation, and a possible further transition to the turbulent state at high temperatures, still remains a considerable challenge.
Bibliography


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List of symbols

\(a\) \hspace{2em} m \hspace{2em} \text{radius of the tube}

\(A\) \hspace{2em} m^2 \hspace{2em} \text{cross-sectional area of the tube}

\(A'\) \hspace{2em} - \hspace{2em} \text{ratio gelatinised/ungelatinised viscosity (Morgan’s model)}

\(b\) \hspace{2em} - \hspace{2em} \text{constant (Morgan’s model)}

\(B\) \hspace{2em} - \hspace{2em} \text{relative viscous reduction (Morgan’s model)}

\(c\) \hspace{2em} \% \text{ w/w} \hspace{2em} \text{starch concentration}

\(C\) \hspace{2em} - \hspace{2em} \text{constant in modified Cox-Merz rule}

\(c_p\) \hspace{2em} \text{J/kg}^\circ\text{C} \hspace{2em} \text{heat capacity}

\(d\) \hspace{2em} m \hspace{2em} \text{tube diameter}

\(d'\) \hspace{2em} - \hspace{2em} \text{constant (Morgan’s model)}

\(D\) \hspace{2em} m \hspace{2em} \text{coil diameter}

\(D_{ec} = Re \sqrt{d/D}\) \hspace{2em} - \hspace{2em} \text{generalised Dean number}

\(E_a\) \hspace{2em} \text{kJ/mol} \hspace{2em} \text{activation energy of viscous temperature dependency}

\(E_g\) \hspace{2em} \text{kJ/mol} \hspace{2em} \text{activation energy of gelatinisation}

\(f = 2\Delta pd/\rho \omega^2\) \hspace{2em} - \hspace{2em} \text{circumferentially averaged friction factor}

\(f_n = f Re/64\) \hspace{2em} - \hspace{2em} \text{circumferentially averaged normalised friction factor}

\(f'\) \hspace{2em} - \hspace{2em} \text{degree of gelatination}

\(F\) \hspace{2em} \text{m}^3/\text{s} \hspace{2em} \text{volume flow rate}

\(F_0\) \hspace{2em} \text{s} \hspace{2em} \text{equivalent lethality}

\(F_b\) \hspace{2em} \text{N/m}^3 \hspace{2em} \text{buoyant body force}

\(F_c\) \hspace{2em} \text{N/m}^3 \hspace{2em} \text{centrifugal body force}

\(F\) \hspace{2em} \text{N/m}^3 \hspace{2em} \text{vector of body forces}

\(g\) \hspace{2em} \text{m/s}^2 \hspace{2em} \text{gravitational acceleration}

\(g\) \hspace{2em} \text{m/s}^2 \hspace{2em} \text{vector of gravity}

\(g'\) \hspace{2em} - \hspace{2em} \text{extent of shear destruction}
\[ q \quad \text{m/s}^2 \quad \text{vector of gravity} \]
\[ G^* \quad \text{Pa} \quad \text{loss modulus} \]
\[ G_r = \frac{\beta g \Delta T d^3 \rho^2 / \mu_s^2}{-} \quad \text{generalised Grashof number} \]
\[ h \quad \text{W/m}^2 \text{°C} \quad \text{heat transfer coefficient} \]
\[ H_e = \frac{Re \sqrt{d/D(1 + (p'/\pi D)^2)}}{-} \quad \text{generalised Helical number} \]
\[ k_0 \quad \text{1/s} \quad \text{rate constant of Clostridium Botulinum or thiamine destruction} \]
\[ k_g \quad \text{Pa s/s} \quad \text{rate of reaction of gelatinisation} \]
\[ k_{g0} \quad \text{Pa s/s} \quad \text{rate constant of gelatinisation} \]
\[ k_{g0}' \quad \text{1/s °C} \quad \text{rate constant of gelatinisation (Morgan’s model)} \]
\[ k_s \quad \text{s} \quad \text{rate constant of shear destruction} \]
\[ L \quad \text{m} \quad \text{length of section} \]
\[ m \quad \text{Pa s}^n \quad \text{consistency index} \]
\[ m_0 \quad \text{Pa s}^n \quad \text{pre-exponential consistency index} \]
\[ M_\phi \quad * \quad \text{inflow of } \phi \text{ (*unit depends on scalar)} \]
\[ n \quad - \quad \text{power-law index} \]
\[ n' \quad - \quad \text{number of gelatinisation classes} \]
\[ N \quad - \quad \text{number of spores} \]
\[ Nu = h d / \lambda \quad - \quad \text{circumferentially averaged Nusselt number} \]
\[ P \quad \text{Pa} \quad \text{pressure} \]
\[ p' \quad \text{m} \quad \text{coil pitch} \]
\[ P_e = \bar{\rho} d / \mathcal{D} \quad - \quad \text{Péclet number} \]
\[ Pr = c_p \mu_s / \lambda \quad - \quad \text{generalised Prandtl number} \]
\[ q \quad \text{W/m}^2 \quad \text{heat flux} \]
\[ r \quad \text{m} \quad \text{radial coordinate} \]
\[ r_\phi \quad * \quad \text{local residual (*unit depends on scalar)} \]
\[ R \quad \text{J/mol °C} \quad \text{universal gas constant} \]
\[ R' \quad \text{m} \quad \text{radius of the coil} \]
\[ R_\phi \quad - \quad \text{sum of normalised residuals} \]
\[ Re = \rho \bar{\mu} d / \mu_s \quad - \quad \text{generalised Reynolds number} \]
\[ S_\phi \quad * \quad \text{scalar source term (*unit depends on scalar)} \]
\[ t \quad \text{s} \quad \text{time} \]
\[ T \quad ^\circ C \quad \text{temperature} \]
\[ \dot{T} \quad ^\circ C/s \quad \text{heating rate} \]
\[ T_g \quad ^\circ C \quad \text{gelatinisation temperature} \]
\[ u, v, w \quad \text{m/s} \quad \text{cartesian velocities} \]
\[ \textbf{v} \quad \text{m/s} \quad \text{velocity vector} \]
\[ v_s = \sqrt{v_x^2 + v_y^2} \quad \text{m/s} \quad \text{strength of the secondary flow} \]
\[ \overline{w} \quad \text{m/s} \quad \text{average axial velocity} \]
\[ x, y, z \quad \text{m} \quad \text{cartesian coordinates} \]
\[ \ddot{z} = \frac{\mu z}{\rho d^2 \overline{w}} \quad - \quad \text{dimensionless hydrodynamic length} \]
\[ z^* = \frac{\lambda z}{\rho c_p d^2 \overline{w}} \quad - \quad \text{dimensionless thermal length} \]
\[ \alpha \quad - \quad \text{coefficient in modified Cox-Merz rule} \]
\[ \alpha' \quad - \quad \text{constant (Morgan’s model)} \]
\[ \beta \quad 1/\circ C \quad \text{volumetric expansion coefficient} \]
\[ \beta' \quad - \quad \text{exponent of frequency scale factor} \]
\[ \delta \quad \text{rad} \quad \text{phase angle} \]
\[ \delta' = \frac{a}{R} = \frac{a}{D} \quad - \quad \text{radius of curvature} \]
\[ \Delta_{i,j} = \quad \frac{\partial v_i / \partial x_j + \partial v_j / \partial x_i}{1/s} \quad \text{rate of strain tensor} \]
\[ \Delta H_g \quad \text{J/g} \quad \text{enthalpy of gelatinisation} \]
\[ \Delta p \quad \text{Pa/m} \quad \text{pressure drop} \]
\[ \Delta t_{\mu-\mu_{\infty}} \quad \text{s} \quad \text{time to sheared asymptotic viscosity} \]
\[ \Delta \mu_r \quad \text{Pa s} \quad \text{contribution of each class to the complex reduced viscosity} \]
\[ \gamma \quad 1/s \quad \text{shear rate} \]
\[ \gamma' \quad - \quad \text{oscillatory strain} \]
\[ \gamma_0 \quad - \quad \text{oscillatory strain amplitude} \]
\[ \overline{\gamma} \quad 1/s \quad \text{average shear rate (Morgan’s model)} \]
\[ \lambda \quad \text{W/m}^{\circ C} \quad \text{conductivity} \]
\[ \mu \quad \text{Pa s} \quad \text{apparent dynamic viscosity} \]
\[ \mu_s = m \left( \frac{x_n}{m} \right)^n \left( \frac{\overline{w}^2}{3} \right)^{-1} \quad \text{Pa s} \quad \text{apparent dynamic viscosity for straight tube flow} \]
\[ \mu^* \quad \text{Pa s} \quad \text{complex dynamic viscosity} \]
\[ \mu_r \quad \text{Pa s} \quad \text{reduced complex dynamic viscosity} \]
\[ \overline{\mu}_{rel} \quad - \quad \text{mass averaged relative viscosity} \]
\begin{align*}
\omega & \quad \text{rad/s} \quad \text{frequency} \\
\rho & \quad \text{kg/m}^3 \quad \text{density} \\
\phi & \quad \ast \quad \text{scalar concentration (unit depends on scalar)} \\
\phi_n = \phi / \phi_{in} \cdot 100\% & \quad \% \quad \text{normalised thiamine concentration} \\
\Phi & \quad 1 / \text{s}^2 \quad \text{dissipation function} \\
\Phi' & \quad - \quad \text{integral strain history (Morgan’s model)} \\
\psi & \quad \circ \quad \text{axial coordinate in curved flow} \\
\Psi & \quad - \quad \text{integral temperature history (Morgan’s model)} \\
\tau & \quad \text{Pa} \quad \text{shear stress} \\
\tau_e & = \mu \left[ \frac{1}{2} \sum \sum \Delta ij \Delta ij \right]^{\frac{1}{2}} \quad \text{Pa} \quad \text{effective shear stress} \\
\tau_g & \quad \text{Pa} \quad \text{structural strength of gelatinisation class} \\
\tau' & \quad \text{Pa} \quad \text{oscillatory stress response} \\
\tau_0' & \quad \text{Pa} \quad \text{oscillatory stress amplitude} \\
\mathcal{D} & \quad \text{m}^2 / \text{s} \quad \text{diffusion coefficient}
\end{align*}

Sub- and superscripts

\begin{align*}
c & \quad \text{critical} \\
i & \quad \text{class number} \\
in & \quad \text{inlet} \\
out & \quad \text{outlet} \\
init & \quad \text{initial} \\
min & \quad \text{minimum} \\
max & \quad \text{maximum} \\
ref & \quad \text{reference} \\
r & \quad \text{radial} \\
w & \quad \text{wall} \\
\theta & \quad \text{tangential} \\
+ & \quad \text{maximum} \\
\infty & \quad \text{asymptotic}
\end{align*}
Appendix A

The toroidal coordinate system

In this section the equivalence of the equations of motions in a weakly curved toroidal coordinate system and a Cartesian system including a centrifugal body force will be demonstrated. According to Berger et al. (1983), the non-dimensional and scaled equations of motion for a toroidal coordinate system (figure 2.4) of arbitrary curvature ratio \( \delta' = d/D \) are given by equation A.1 to A.3. For convenience, the pressure related and viscous terms on the right hand side have been abbreviated by \( P_{r,\theta,\psi} \) and \( V_{r,\theta,\psi} \) respectively.

\[
v_r \frac{\partial v_r}{\partial r} + \frac{v_\theta}{r} \frac{\partial v_r}{\partial \theta} + \frac{v_\psi}{r} \frac{\partial v_r}{\partial z} - \frac{v_\theta^2 \cos \theta}{1 + \delta' r \cos \theta} = P_r + V_r \quad (A.1)
\]

\[
v_r \frac{\partial v_\theta}{\partial r} + \frac{v_\theta}{r} \frac{\partial v_\theta}{\partial \theta} + \frac{v_\psi}{r} \frac{\partial v_\theta}{\partial z} + \frac{v_r v_\theta}{r} + \frac{v_\psi^2 \sin \theta}{1 + \delta' r \cos \theta} = P_\theta + V_\theta \quad (A.2)
\]

\[
v_r \frac{\partial v_\psi}{\partial r} + \frac{\delta' v_r v_\psi \cos \theta}{1 + \delta' r \cos \theta} + \frac{v_\theta}{r} \frac{\partial v_\psi}{\partial \theta} - \frac{\delta' \sin \theta}{1 + \delta' r \cos \theta} v_r v_\psi + \frac{v_\psi}{r} \frac{\partial v_\psi}{\partial z} = P_\psi + V_\psi \quad (A.3)
\]

Application to equations A.1 and A.2 of the weakly curved assumption (\( \delta' \ll 1 \)) for all terms except the fifth, and to all terms of equation A.3 yields equations A.4 to A.6.

\[
v_r \frac{\partial v_r}{\partial r} + \frac{v_\theta}{r} \frac{\partial v_r}{\partial \theta} + v_\psi \frac{\partial v_r}{\partial z} - \frac{v_\theta^2 \cos \theta}{1 + \delta' r \cos \theta} = P_r + V_r \quad (A.4)
\]

\[
v_r \frac{\partial v_\theta}{\partial r} + \frac{v_\theta}{r} \frac{\partial v_\theta}{\partial \theta} + v_\psi \frac{\partial v_\theta}{\partial z} + \frac{v_r v_\theta}{r} + \frac{v_\psi^2 \sin \theta}{1 + \delta' r \cos \theta} = P_\theta + V_\theta \quad (A.5)
\]
\[ v_r \frac{\partial v_r}{\partial r} + v_\theta \frac{\partial v_r}{\partial \theta} + v_\psi \frac{\partial v_r}{\partial z} = P_r + V_r \]  

(A.6)

Equations A.4 to A.6 can be transformed to their dimensional form by the substitutions given in equation A.7.

\[ r = \frac{r'}{a} \quad z = \frac{R' \sqrt{\delta'}}{a} = \frac{z'}{\delta'} \quad \delta' = \frac{a}{R'} \]  

(A.7)

\[ v_r = \frac{v'}{\sqrt{\delta'}} \quad v_\theta = \frac{v'_\theta}{\sqrt{\delta'}} \quad v_\psi = \frac{v'_\psi}{\delta'} \]

On substitution, omitting the accent in \( r, z \) and the velocities, and multiplication by the density \( \rho \), we obtain equations A.8 to A.10.

\[ \rho \left( v_r \frac{\partial v_r}{\partial r} + \frac{v_\theta}{r} \frac{\partial v_r}{\partial \theta} + v_\psi \frac{\partial v_r}{\partial z} - \frac{v_\psi'^2 \cos \theta}{r'} - \frac{v_\theta'^2 \cos \theta}{r'} \right) = P'_r + V'_r \]  

(A.8)

\[ \rho \left( v_r \frac{\partial v_\theta}{\partial r} + \frac{v_\theta}{r} \frac{\partial v_\theta}{\partial \theta} + v_\psi \frac{\partial v_\theta}{\partial z} + \frac{v_r v_\theta}{r} + \frac{v_\psi'^2 \sin \theta}{r'} \right) = P'_\theta + V'_\theta \]  

(A.9)

\[ \rho \left( v_r \frac{\partial v_\psi}{\partial r} + \frac{v_\theta}{r} \frac{\partial v_\psi}{\partial \theta} + v_\psi \frac{\partial v_\psi}{\partial z} \right) = P'_\psi + V'_\psi \]  

(A.10)

For large values of the radius of curvature \( R' = D/2 \) equations A.8 to A.10 revert to the cylindrical coordinate system, and the axial velocity \( v_\psi \) equals \( v_z = w \). A weakly curved pipe can therefore be described by a cylindrical coordinate system and two extra terms representing the centrifugal force. Transformation of the cylindrical coordinate system may yield the Cartesian coordinate system. Vectorial addition of the additional centrifugal \( r \) and \( \theta \) component yields the total centrifugal force \( F_c \) (equation A.11), which points in the x-direction of the Cartesian coordinate system.

\[ F_c = \rho \sqrt{\left( \frac{v_\psi'^2 \cos \theta}{R' + r \cos \theta} \right)^2 + \left( \frac{v_\psi'^2 \sin \theta}{R' + r \cos \theta} \right)^2} = \rho \frac{w^2}{x} \]  

(A.11)
Appendix B

Mesh and scheme dependence

To establish independence of the developed solutions, a large number of cross-sectional meshes was tested. To approximate the circular shape of the tube by essentially square computational cells, two grid arrangements termed butterfly and cartwheel, were employed (figure B.1).

Although cartwheel meshes are easily built and yield satisfactory results in axi-symmetric flows, they perform poorly in curved flows, probably as a result of the small internal angle of the computational cells neighbouring the centerline. Further independence testing was therefore carried out using the butterfly type only. After establishing a satisfying radial location of the triple points (four points where only three cells neighbour) and the ratio of the number of tangential to radial cells, five increasingly refined meshes were constructed.

Figure B.1: Butterfly (left) and cartwheel (right) mesh.
For developed flow, the impact of discretisation practice on the convective terms was assessed by comparing a first, a second and a third order accurate scheme. As first order scheme, upwind differencing (UD) was employed (Versteegh and Malalasekera, 1995). The second order discretisation consisted of a MUSCL (Hirsch, 1988) type scheme with a TVD limiter (Van Leer, 1977), termed mono advection and reconstruction scheme (MARS) (CD Ltd., 2001). For third order accuracy, the quadratic upstream interpolation for convective kinetics (QUICK) was employed (Versteegh and Malalasekera, 1995).

Mesh and scheme dependence were studied over the entire range of curvature ($De = 0 - 1000$) and rheology ($n = 0.2 - 2$). For the boundary conditions and fluid properties used, the reader is referred to section 2.2.1. The normalised friction factor $f_n$ and the Nusselt number $Nu$ as a function of discretisation scheme, rheology, curvature and mesh refinement are presented in tables B.1 to B.9.

Regarding the friction factor, MARS and QUICK performed equally well, and the results obtained using UD are acceptable. Independence was obtained quickly, and for all schemes the $r \times \theta = 27 \times 64$ mesh seems sufficient. For heat transfer, the MARS and the QUICK scheme yielded very similar results, with the QUICK scheme (as expected) approaching mesh independence slightly faster. The UD scheme seems inappropriate for thermal computations, as it predicts extremely high values of the $Nu$ number for curved flow, and fails to reproduce the analytical values for straight pipe flow. Inspection of the temperature field reveals that the UD scheme does not yield a radially symmetric solution for straight pipe flow ($De = 0$).

It is noteworthy that for the increasing number of computational cells, heat transfer does not approach a clear asymptotic value, which is due to the sensitivity of the solution to the location of the triple-points and the ratio of the number of radial to tangential cells. This seems to be a feature of high Prandtl number flows: for smaller tube diameters (and hence lower viscosities and Prandtl numbers at equal Dean number) unambiguous mesh independence could be obtained. However, for engineering purposes the $r \times \theta = 35 \times 80$ mesh combined with either MARS or QUICK is deemed sufficient.

To establish axial mesh independence for developing flows, three meshes having 50, 100 and 200 axial cells were tested. In the inlet region (1/10 of the tube length) axial cell size increased according to a fixed ratio to resolve the entrance effects of the developing flow. For reasons of computational stability, in the outlet region (1/20 of the tube length) cells decreased in axial size towards the exit. The $r \times \theta = 35 \times 80$ mesh was employed for
the cross-sectional discretisation. Because of the poor performance of the UD scheme in developed flow, only the second order MARS and third order QUICK scheme were tested. Mesh and scheme dependence were studied over the range of curvature of $De = 0 - 500$ and for a Newtonian rheology. The boundary conditions and fluid properties can be found in sections 2.2.1 and 2.2.2 respectively.

In figures B.2 to B.5 the normalised developing friction factor $f_n(\hat{z})$ and Nusselt number $Nu(z^*)$ are presented as a function of discretisation scheme, curvature and axial mesh refinement. A mesh having 100 axial cells seems sufficient to resolve the details of the oscillatory behaviour of both the friction factor and the Nusselt number. Comparing friction factor and the Nusselt number at 100 cells for the MARS and the QUICK scheme in figures B.6 and B.7, reveals only a slightly more conservative Nusselt number for the QUICK scheme. This agrees with the trends of the developed simulations of tables B.5 and B.8.
<table>
<thead>
<tr>
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</tr>
<tr>
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<td>1.000</td>
</tr>
<tr>
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Table B.1: Dependence of $f_n$ and $Nu$ on the mesh for UD and $n = 0.2$.

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Table B.2: Dependence of $f_n$ and $Nu$ on the mesh for UD and $n = 1$.

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Table B.3: Dependence of $f_n$ and $Nu$ on the mesh for UD and $n = 2$. 

128
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Table B.4: Dependence of $f_n$ and $Nu$ on the mesh for MARS and $n = 0.2$. 

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Table B.5: Dependence of $f_n$ and $Nu$ on the mesh for MARS and $n = 1$. 

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</tr>
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Table B.6: Dependence of $f_n$ and $Nu$ on the mesh for MARS and $n = 2$. 

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Table B.7: Dependence of \( f_n \) and \( Nu \) on the mesh for QUICK and \( n = 0.2 \).

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Table B.8: Dependence of \( f_n \) and \( Nu \) on the mesh for QUICK and \( n = 1 \).

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Table B.9: Dependence of \( f_n \) and \( Nu \) on the mesh for QUICK and \( n = 2 \).
Figure B.2: Dependence of $f_n$ on the axial mesh for MARS.

Figure B.3: Dependence of $N_u$ on the axial mesh for MARS.
Figure B.4: Dependence of $f_n$ on the axial mesh for QUICK.

Figure B.5: Dependence of $Nu$ on the axial mesh for QUICK.
Figure B.6: MARS versus QUICK: $f_n$ for 100 axial cells.

Figure B.7: MARS versus QUICK: $Nu$ for 100 axial cells.
Bibliography

Appendix C

Instationary curved pipe flow

No steady solutions could be found for developed dilatant flow and $De > 250$. To assess whether the flow is physically unsteady, simulations were carried out on the full cross-section of the pipe using Issa’s transient PISO (Pressure Implicit with Splitting of Operators) algorithm to decouple the discretised equations 2.5 - 2.7 including all time-dependent terms. Fully implicit first order accurate temporal discretisation was employed, and a time-step $\Delta t = 0.002$ s amounting to about 200 time-steps per oscillation. All convective terms were discretised using the standard MARS scheme. The resulting algebraic equations were solved using a conjugate gradient method. Completion of the computations was judged from the global rates of change of mass, momentum and energy.

From a developed steady straight pipe flow, all cases quickly ’converged’ to oscillating, symmetrical solutions with frequencies of 2.2 - 2.3 Hz. Figure C.1 shows the instationary axial velocity, the magnitude of the secondary velocity, and the temperature fields for ten instances during one period for $De = 500$ and $n = 2$.

The unsteadiness may be explained from the dilatancy of the flow. If the axial velocity maximum moves to the outside of the coil, local shear rates increase and the apparent viscosity near the wall rises. This in turn slows down the secondary flow which is confined to the boundary layer at these high Dean numbers. As the secondary circulation slows down, the axial flow maximum returns towards the pipe axis, decreasing the velocity gradients and the apparent viscosity at the outer wall. This additional coupling between axial and secondary flow may induce a self-sustaining oscillation. It must be stressed however that the temporal discretisation used was only first order and that no experimental data is available to verify these numerical results.

As the fluctuations in the friction factor and Nusselt number proved at most 2% and 1.9% respectively, $f_s$ and $Nu_s$ shown in figures 2.9 and 2.10 are time-averages of ten instances uniformly distributed over one oscillation.
Figure C.1: From top to bottom: oscillatory axial and secondary velocity ($v_{a}$), and temperature ($De = 500$ and $n = 2$).
Appendix D

Mixed convective flow

Buoyancy forces are usually incorporated in curved pipe flow and heat transfer simulations by the Boussinesq approximation (Lee, Simon, and Chow, 1985; Yang and Chang, 1994; Goering, Humphrey, and Greif, 1997). Only the forces arising from the density differences of the fluid are accounted for and for all other purposes density is assumed constant. This approximation is justifiable for isobaric density changes that have no significant volumetric effects on the flow field, implying moderate temperature differences across the fluid (Lee et al., 1985). Two additional elements of the Boussinesq approximation, Newtonian flow and the absence of viscous dissipation (Sillekens, 1995), have been released in this study.

The effect of density variations can be added as $F_b$ (equation D.1) to $E$ in the $y$-momentum equation 2.6.

$$F_b = -g \cdot \rho \frac{1}{1 + \beta (T - T_0)}$$  \hspace{1cm} (D.1)

Estimates of the magnitude of the buoyancy relative to the centrifugal force were given by Sillekens, Rindt, and Van Steenhoven (1998), and flow regime maps to determine whether the flow is in the buoyant, mixed or centrifugal region were provided by Prusa and Yao (1981), Lee et al. (1985), Futugami and Aoyama (1988) and Goering et al. (1997). Though these authors do not quantitatively agree (Goering et al., 1997), based on the process conditions in laminar steriliser flow (high velocities and high viscosity), flow and heat transfer are expected to be dominated by centrifugal and viscous forces.

It has to be borne in mind however that buoyancy and centrifugal forces exert their influence in essentially different locations in the flow. Centrifugal forces prevail in the core of the flow where axial velocities are highest, whereas
buoyancy dominates close to the wall, especially in high Prandtl number flow (Lee et al., 1985). Moreover, velocity and temperature fields are coupled in mixed flows. Normalised developed friction factors $f_n$ and developed Nusselt numbers $Nu$ were therefore simulated with and without buoyancy (figures D.1 and 5.4).

![Graph showing developed friction factor for curved and mixed flow.](image)

Figure D.1: Developed friction factor for curved and mixed flow.

For the current simulations, developed $f_n$ and $Nu$ do not depend on buoyant effects. To assess the importance of buoyancy in developing flow and at a maximum driving force, simulation of the high temperature heater and cooler of a steriliser were performed with and without buoyancy. In section 5.5.1.3 the impact of buoyancy is shown to be minimal.
Bibliography


Appendix E

Rheometry of starch suspensions

Over the course of an UHT-sterilisation, the temperature of a starch containing food recipe varies between the storage (5°C) the sterilisation (140°C), and the filling (5°C) temperature. During heating, the food gelatinises and the rheology of the eventual product is quite different from the initial formulation. Extensive measurements are required to describe fluids whose rheological properties depend on temperature, shear rate, temperature history, and shear rate history (Lagarrigue and Alvarez, 2001). Two distinct techniques, oscillatory and steady shear rheometry, are briefly treated.

E.1 Oscillatory shear rheometry

Small amplitude oscillatory rheometry can be employed to characterise a sample while keeping structural modification minimal. In chapter 3, the apparent viscosity $\mu$ was derived from the complex viscosity $\mu^*$ using a modified Cox-Merz rule.

When a gelatinising suspension is subject to a small sinusoidal strain $\gamma'(t) = \gamma_0 \sin(\omega t)$, its stress response $\tau'(t)$ may be partly elastic and partly viscous (equation E.1).

$$\tau'(t) = G'\gamma_0 \sin(\omega t) + G''\gamma_0 \cos(\omega t)$$  \hspace{1cm} (E.1)

The elastic stress is in phase with the strain and is linked to it by the storage modulus $G'$. The viscous stresses are 90 degrees out of phase and are proportional to the loss modulus $G''$. Visco-elastic suspensions exhibit partly
viscous and partly elastic behaviour, reflected in a phase lag $\delta$ in their stress response (equation E.2).

$$\tau'(t) = \tau_0' \sin(\omega t + \delta)$$  \hspace{1cm} (E.2)

From equations E.1 and E.2 expressions for the storage and the loss modulus can be derived (equation E.3).

$$G' = \left(\tau_0'/\gamma_0'\right) \cos(\delta)$$  \hspace{1cm} (E.3)

$$G'' = \left(\tau_0'/\gamma_0'\right) \sin(\delta)$$

The complex modulus $|G^*| = \sqrt{G'^2 + G''^2}$ can be divided by the oscillatory frequency to yield the complex viscosity $|\mu^*| = |G^*|/\omega$ (Ferry, 1980; Okechukwu and Rao, 1998).

### E.2 Steady shear rheometry

Steady shear rheometry may be employed to obtain the power-law coefficients of gelatinised suspensions. Similarly, the equilibrium viscosity of gelatinising shear resistant starch suspensions can be determined without using any form of the Cox-Merz rule (Tattiyakul, Rao, and Datta, 2002). For suspensions that are susceptible to shear, it provides the kinetics of shear degradation (Nguyen, Jensen, and Kristensen, 1998).

For stable, gelatinised suspensions following the Arrhenius temperature dependency, the power-law relationship is given by equation E.4. At constant temperature, the pre-exponential consistency index $m_0$ and the power-law index $n$ can be calculated from the shear stress $\tau$ as a function of shear rate $\dot{\gamma}$. Provided $n$ is not a function of temperature, the activation energy $E_a$ can then be obtained from the shear stress as a function of temperature.

$$\tau = m_0 e^{E_a/k_B} \dot{\gamma}^n$$  \hspace{1cm} (E.4)

The rate constant of shear destruction $k_s$ is found using equation 3.19. For constant shear rate $\dot{\gamma}$, integrating over the time interval $t_{\infty}$ from initial $(\mu_{\text{init}})$ to equilibrium $(\mu_{\infty})$ apparent viscosity yields equation E.5.

$$\frac{\mu_{\infty}}{\mu_{\text{init}}} = e^{-k_s \dot{\gamma} t_{\infty}}$$  \hspace{1cm} (E.5)

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For a concentric rheometer $\Phi$ equals $\gamma_r^2$. If sufficient data is available, $k_s$ can be made a function of temperature. Next, the granular shear strength $\tau_g$ can be calculated from equation E.6. Given the underlying progressive swelling and softening of the granules at higher temperatures, $\tau_g$ is likely to be a function of temperature as well.

$$\tau_g(T) = \mu_\infty(T) \gamma_r$$

(E.6)

By virtue of their well defined shear conditions, concentric or cone and plate rheometers are preferred (Lagarrigue and Alvarez, 2001). However, when temperatures exceed 95 °C, the rheometer must be pressurised to prevent excessive vapourisation and boiling. Though Abdelrahim, Ramaswamy, and van de Voort (1995) reported data on cross-linked waxy maize starch in a pressurised concentric rheometer, tube viscometry is more common at elevated pressures (Dail and Steffe, 1990; Self, Wilkins, Morley, and Bailey, 1990; Heydon, Scott, and Tucker, 1996; Xu and Raphaelides, 1998).

Tube viscometry is limited to simple power-law flow at equilibrium, as the shear conditions in gelatinising flow are essentially unknown. For power-law flow, the relationship between pressure drop across the instrument and the volume flow rate $F$ is given by equation E.7 (Skelland, 1967; Heydon, Scott, and Tucker, 1996).

$$\frac{d \Delta p}{4 L} = m \left( \frac{32 F}{\pi d^3} \right)^n \left( \frac{3n + 1}{4n} \right)^n$$

(E.7)

Performing differential pressure measurements over a tube section $L$ for several flow rates yield the power law index $n$ and the consistency index $m$. Repeating the procedure at different temperatures enables description of the temperature dependency of the consistency index in terms of the Arrhenius model. An in-line viscometer was built to use in conjunction with the Stork UHT test set-up (Kelder, 1999).
Bibliography


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Curriculum Vitae

Jasper Kelder was born on March 25, 1972 in Groningen, the Netherlands. After graduation from secondary school at the Rijks Scholengemeenschap in Heerenveen, he studied mechanical engineering at the Twente University. In August 1996 he graduated on a comparison of measurements and simulations of the flow in the volute of a quasi 2-dimensional centrifugal pump. This work was carried out at the Laboratory of Thermal Engineering of Prof. Dr. Ir. J.J.H. Brouwers. In January 1997 he started a two-year post-graduate course in chemical engineering at the Stan Ackermans Institute of the Eindhoven University of Technology. During the concluding year of this course, the numerical modelling of spiral sterilisers was initiated. In a joint effort of Stork Food & Dairy Systems B.V. and the Eindhoven University of Technology, this work was extended to a PhD project, which was supervised by Dr. Ir. K.J. Ptasinski and Prof. Dr. Ir. P.J.A.M. Kerkhof of the Laboratory of Separation Technology. In November 2002 Jasper Kelder has joined the Agro & Industrial Production Chains department of ATO B.V. in Wageningen.