

Performance measures of packed beds for decontamination of industrial emissions

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PERFORMANCE MEASURES OF PACKED BEDS FOR DECONTAMINATION OF INDUSTRIAL EMISSIONS.

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ABSTRACT

A theoretical model for the flow of a gas mixture at relatively high speed through a packed bed with steep temperature gradients and with exothermic reactions taking place [1] is used to study the performance of packed beds. The model is based on analytical solutions for each bed element, making the computational algorithm fast and facilitating the performance study of the bed.

A so-called enthalpy transport length is introduced as a convenient optimization tool. The thermal efficiency is found to be less valuable in this respect. Particle size dispersion and inhomogeneous packing of bed material is examined with the aid of the model and the enthalpy transport length.

1. INTRODUCTION

Many industrial processes discharge airflows containing concentrations of volatile hydrocarbons into the atmosphere. Especially for low concentrations packed bed regenerators are an efficient means to overcome this type of air pollution by thermal oxidation of the hydrocarbons to CO_2 and H_2O .

In the packed bed decontamination or deodorization occurs at a high temperature level in a matrix of randomly dumped ceramic solid particles. After entering the bed the airflow is heated up to a high temperature level of approximately 1000°C about half way into the bed, in a region we call the mid bed zone. In this zone, the thermal oxidation rate is sufficiently high for complete destruction of the contaminants within the residence time. Enthalpy released by the oxidation process, as well as the enthalpy taken up in the first bed half is released again in the second bed half, downstream of the mid bed zone. At an average temperature of 40 to 70°C above inlet temperature (dependent on the reaction enthalpy) the cleaned airflow leaves the regenerator. By the continuous removal (upstream) and returning

(downstream) of heat, the bed temperature profile gradually shifts in flow direction. In order to limit this shifting the air flow direction is periodically alternated.

Optimization of the design and energy efficiency of this type of regenerator preferably requires efficient performance measures.

An enthalpy transport length is introduced as a convenient optimization tool. Employing this performance measure, as well as the profile shift velocity, the influence on regenerator performance of inhomogeneous packing of the bed is theoretically investigated. The use and applicability of both enthalpy transport length and profile shift velocity are studied with the aid of selected test cases and with a numerical model. The model predicts temperature distributions by Laplace transformations of the transport equations valid in a bed element [1].

The theoretical model, computational method and results are presented in this paper. As compared to other models available in literature [2,3,4] our model takes full account of large temperature gradients, by solving for an extended set of initial and boundary conditions and for internal heat sources. The Schumann model [3,4], for example, only employs solutions with uniform initial bed temperature, constant inlet temperature and no heat sources. The variation of physical properties, such as mass densities and heat capacities, is accounted for by applying the analytical solutions to bed segments only.

2. MODELING

The one-dimensional model neglects heat diffusion in radial direction and computes the time-dependent gas mixture temperature, T , and the temperature at the surface of solid particles, θ .

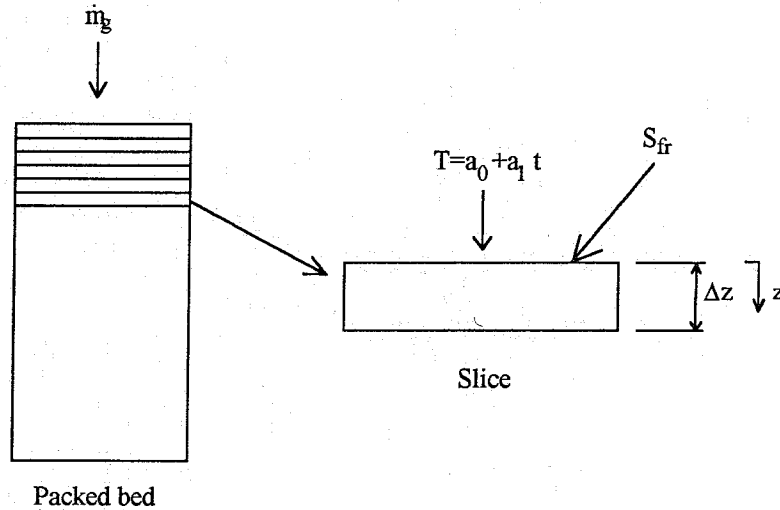


FIGURE 1. SCHEMATIC OF PACKED BED AND BED SLICE. S_{fr} DENOTES THE AREA OF ONE SIDE OF THE CYLINDRICAL SLICE, I.E. THE FRONTAL AREA THROUGH WHICH THE GAS ENTERS WITH A MASS FLOW RATE \dot{m}_g .

For predicting temperature distributions in the entire bed, first only a finite cylinder of packed bed is considered, with axis z in flow direction and very small height Δz , such that the parameters that determine flow and heat transfer are approximately constant in this slice, see figure 1.

Similarly, a finite time interval Δt is considered, in which time-dependence of flow and transfer parameters is negligible. The flow and transfer parameters encompass physical properties, e.g. mass density of the gas mixture and particles, ρ_g and ρ_p , the heat capacities c_g and c_p and the convective transfer coefficient for heat transport between both phases, h . Each parameter is evaluated at the known thermodynamical conditions at the beginning of the timestep, at $t=0$. It is noted that these parameters are only kept constant within the slice Δz and time interval Δt .

Temperature T of the gasflow at inlet of a slice, at $z=0$ and initial bed temperature θ , at $t=0$, are described by functions of the form:

$$T = a_0 + a_1 \cdot t \quad (1)$$

$$\theta = b_0 + b_1 \cdot z + b_2 \cdot z^2 \quad (2)$$

The bed temperature, θ , refers to the temperature at the surface of the particles and is considered as not affected by internal conduction effects.

For each slice the superficial gas velocity, u_g , equal to $\frac{\dot{m}_g}{\rho_g \cdot S_{fr}}$, is assumed to be high enough to justify the neglect of the rate of

enthalpy accumulation in the gas, $c_g \cdot \rho_g \cdot \frac{dT}{dt}$, with respect to

enthalpy convection, $c_g \cdot \rho_g \cdot u_g \cdot \frac{dT}{dz}$. In practice, transport

agencies such as conduction inside the bed particles, mixing of gas volumes at a different temperature, convective transport and radiation occur which are partially interacting [5]. Hence, observed heat transfer rates in packed beds are the result of local heat transport agencies at the particle surfaces and flow patterns in the voids among the particles. The apparent contribution of these effects is lumped into a single parameter: the convective transfer coefficient, h . Note that the velocity u_g is quite high and not constant in the bed due to density changes. Usually the flow regime is turbulent.

These assumptions permit to reduce the enthalpy balances for gas mixture and packing material to

$$\rho_g \cdot c_g \cdot u_g \cdot \frac{dT}{dz} = h \cdot a_{spec} \cdot (\theta - T) + \dot{q}_g \quad (3)$$

$$(1 - \epsilon) \cdot \rho_p \cdot c_p \cdot \frac{d\theta}{dt} = h \cdot a_{spec} \cdot (T - \theta) + \dot{q}_p \quad (4)$$

where a_{spec} denotes the heat exchanging bed surface area per unit volume of packed bed, ϵ the void fraction of the packed bed and \dot{q}_g , \dot{q}_p heat sources [W/m^3] due to internal heating of gas and bed particles respectively. In catalytic reactors \dot{q}_p is well defined, but in this study we focus attention on thermal reactors where $\dot{q}_p = 0$. Reaction heat released in the gas is attributed to \dot{q}_g . This is further outlined in [5].

The set of equations (1-4) reduces to the Schumann model [3,4] for a time-independent inlet temperature of the gas mixture at $z=0$ ($a_1=0$), constant initial bed temperature ($b_1=b_2=0$) and no heat sources in either phase ($\dot{q}_g = \dot{q}_p = 0$).

Equations 3 and 4 are simplified by defining the dimensionless temperatures T_g and T_p for gas and particles respectively

$$T_g = \frac{^{def} T - b_0 - a_1 \cdot t}{a_0 - b_0} \quad T_p = \frac{^{def} \theta - b_0 - a_1 \cdot t}{a_0 - b_0} \quad (5)$$

and the dimensionless length ξ and time η by

$$\xi = \frac{^{def} h \cdot a_{spec} \cdot z}{\rho_g \cdot c_g \cdot u_g} \quad \eta = \frac{^{def} h \cdot a_{spec} \cdot t}{(1-\varepsilon) \cdot \rho_p \cdot c_p} \quad (6)$$

After substitution of (5) and (6), the governing equations (3,4) and boundary conditions (1,2) are Laplace transformed. After inverse transformation solutions for the dimensionless temperatures T_g and T_p yield

$$\begin{aligned} T_g(\eta, \xi) = & (1 - Q_g + k - 2 \cdot l) \cdot e^{-\xi} \cdot e^{-\eta} \cdot I_0(2\sqrt{\xi \cdot \eta}) + \\ & + (1 - 2 \cdot Q_g - Q_p + 2 \cdot k - 6 \cdot l) \cdot e^{-\xi} \cdot \int_0^\eta e^{-u} \cdot I_0(2\sqrt{\xi \cdot u}) du + \\ & - (Q_g + Q_p - k - 6 \cdot l) \cdot e^{-\xi} \cdot \int_0^\eta \int_0^m e^{-u} \cdot I_0(2\sqrt{\xi \cdot u}) du dm + \\ & - 2 \cdot l \cdot e^{-\xi} \cdot \int_0^\eta \int_0^m \int_0^n e^{-u} \cdot I_0(2\sqrt{\xi \cdot u}) du dn dm + \\ & + l \cdot \xi^2 + \xi \cdot (k - 2 \cdot l) - 2 \cdot \xi \cdot \eta \cdot l + (Q_g - k + 2 \cdot l) + \\ & + \eta \cdot (Q_g + Q_p - k + 4 \cdot l) + \eta^2 \cdot l \end{aligned} \quad (7)$$

$$\begin{aligned} T_p(\eta, \xi) = & (1 - Q_g + k - 2 \cdot l) \cdot e^{-\xi} \cdot \int_0^\eta e^{-u} \cdot I_0(2\sqrt{\xi \cdot u}) du + \\ & - (Q_g + Q_p - k + 4 \cdot l) \cdot e^{-\xi} \cdot \int_0^\eta \int_0^m e^{-u} \cdot I_0(2\sqrt{\xi \cdot u}) du dm + \\ & - 2 \cdot l \cdot e^{-\xi} \cdot \int_0^\eta \int_0^m \int_0^n e^{-u} \cdot I_0(2\sqrt{\xi \cdot u}) du dn dm + \\ & + l \cdot (\xi^2 + \eta^2) + k \cdot \xi - 2 \cdot \xi \cdot \eta \cdot l + \eta \cdot (Q_g + Q_p - k + 2 \cdot l) \end{aligned} \quad (8)$$

Here I_0 denotes the modified Bessel function of the first kind.

These analytical results for a bedslice Δz are employed for computing temperatures in the entire bed. With an adaptive step algorithm each stepsize Δz is tuned to the gradient variation $\frac{d^2\theta}{dz^2}$, which is proportional to b_2 . Note that a varying inlet temperature of a slice and large temperature gradients are explicitly accounted for by the boundary conditions 1 and 2.

Equations 7 and 8 yield temperatures at the end of each slice at the next time step. Time step size Δt is small enough to have material properties constant *within the time step*.

The model has been validated both theoretically and experimentally in [1]. In this paper, the model is used to evaluate the usefulness (in section 4) of the performance measures introduced in the next section.

3. PERFORMANCE MEASURES

The regenerator design described in the introduction guarantees high thermal efficiencies if packing characteristics and bed height are appropriately chosen. In this section, the thermal efficiency is evaluated as a means of comparing different regenerator designs in the process of optimizing the packed bed regenerator.

Another parameter, the so-called enthalpy transport length, is introduced. Its usefulness as an optimizing tool is examined and compared with the thermal efficiency.

After some starting-up period the bed always reaches a quasi-stationary working situation. Thermal efficiency, η_{th} , of the packed bed is then defined by:

$$\eta_{th} = \frac{T_{max} - T_{out}}{T_{max} - T_{in}} \quad (9)$$

Maximum gas temperature, T_{max} , near the mid bed zone depends on the concentration of hydrocarbons in the airflow, their reaction kinetics as well as packing- and flow characteristics [1,2,6].

In the autothermal mode, time-averaged outlet temperature T_{out} exceeds inlet temperature T_{in} by an amount ΔT , being intimately related to the heat released in the center of the bed by exothermic oxidation [1]. Temperature rise ΔT therefore hardly depends on flow and heat transfer characteristics during autothermal operation¹.

So η_{th} , typically about 97%, merely depends on process parameters and less on bed characteristics. Thermal efficiency therefore is a difficult measure for assessing the effect of bed characteristics such as void fraction maldistribution on regenerator performance. In the following a parameter is introduced that will be shown (section 4) more convenient for this.

¹Matros [2] has found a dependence of T_{max} on ΔT close to a logarithmic one. Results of our own experiments for natural gas mixtures through a packed bed of 1" saddles are correlated by [5]

$$T_{max} = 217,694 + 169,164 \cdot \ln(\Delta T)$$

This relation was also satisfied by the theoretical predictions of the model described in section 2.

The discretized energy balance for the gas in a section of the bed where no heat sources occur reads

$$\rho_g \cdot c_g \cdot u_g \cdot \frac{\Delta T}{\Delta z} = h \cdot a_{spec} \cdot (\theta - T) \quad (10)$$

An estimate for the distance, Δz , traveled by a small volume of gas to loose $n\%$ of its enthalpy content is derived directly from this equation:

$$\Delta z = n\% \cdot \frac{\rho_g \cdot c_g \cdot u_g \cdot T}{h \cdot a_{spec} \cdot |(T - \theta)|} \quad (11)$$

Constant n is an arbitrary integer of order 1.

Let us examine a certain volume of gas crossing the bed downstream of the temperature peak in the mid bed zone. The gas gradually returns the enthalpy taken up in the upstream bed part, as well as the reaction enthalpy, to the particles. If the enthalpy transport length is short, it takes only a small traveling distance for the gas to return a certain amount of its enthalpy to the bed. The total length available for returning enthalpy is equal to the traveling distance through the downstream part of the packed bed. The shorter the enthalpy transport length, the more enthalpy is returned to the packing material before the gas leaves the bed and the lower are enthalpy losses from the bed.

The continuous returning of enthalpy in the downstream bed part results in increasing bed temperatures there. This implies a continuous shifting of the temperature profile in flow direction. The shifting of the temperature profile should not happen too fast, since this would necessitate a short cycle time in order to limit enthalpy losses from the bed. Each alternation of flow direction causes gas remainders to be exhausted without being deodorized or detoxified [1]. Optimal performance of a packed bed regenerator therefore corresponds to long cycle times, if it is possible to arrange a slow shifting of the bed temperature profile.

From the governing equations 3 and 4 it is possible to construct a wave equation [7]. From this equation and assuming locally constant physical properties the profile shift velocity is seen to be:

$$V_g = \frac{\rho_g \cdot c_g \cdot u_g}{\rho_p \cdot c_p \cdot (1 - \varepsilon)} \quad (12)$$

The enthalpy transport length in conjunction with the shift velocity offers a direct way to study bed performance dependencies. For example, the enthalpy transport length of a bed filled with 25 mm torus saddles and a superficial air velocity of 1 m/s is about three times bigger than that of a bed of 5 mm chamotte particles and a typical superficial velocity of 0,3 m/s. For equal thermal performance the height of the saddle bed therefore has to be about a factor three larger than that of

chamotte bed. Similarly, since the profile shift velocity of the saddle bed is a factor five higher than for the chamotte bed its cycle time has to be about a factor five shorter.

For the estimation of the performance of different beds both the enthalpy transport length and the shift velocity are useful tools. Employing these tools, the effect of particle size and void fraction maldistribution is investigated in the next section.

4. MALDISTRIBUTION OF PARTICLE SIZE OR VOID FRACTION

In small scale packed beds (bed dimensions to particle size <20 [8]) the wall region behaves distinctively different than the core region. Due to ordering of particles near the wall, void fraction near the wall is slightly higher than in the core, resulting in a lower flow resistance near the wall. Effects of subsequent bypass flows through the wall region on heat transfer have been evaluated by Schlünder [9]. In the following we focus on large scale packed beds consisting of a mixture of particles of different sizes. Temperatures are assumed equal in each plane perpendicular to the flow direction of the gas by perfect mixing of heat in this direction. Performances of beds consisting of volumetric clusters with a composition which differs from the average bed composition, are compared to the performances of perfectly mixed beds for two generic cases.

Particle size.

In literature on packed beds, different definitions are in use for the average particle size, quantified as an effective diameter. The average particle diameter, $\langle d \rangle$, is here defined by

$$\langle d \rangle = \frac{\sum_i M_i}{\sum_i M_i / d_i} \quad (13)$$

where M_i denotes the total mass of spherical particles with diameter d_i and void fraction ε_i in a part of the packed bed with volume V_i :

$$M_i = \rho_p \cdot (1 - \varepsilon_i) \cdot V_i \quad (14)$$

If heat transfer rates are computed with the model of section 2 in a packed bed filled with perfectly mixed particles of two different diameters d_1 and d_2 , the same heat transfer rates are computed in the bed with particle size $\langle d \rangle$. Since heat transfer is of prime importance, definition (13) is the proper diameter average to be used in packed bed studies.

We now consider a bed B with one half filled with particles of 3 mm diameter and the other half filled with particles of 6 mm diameter. The gas meets only particles of one kind, see figure 2a.

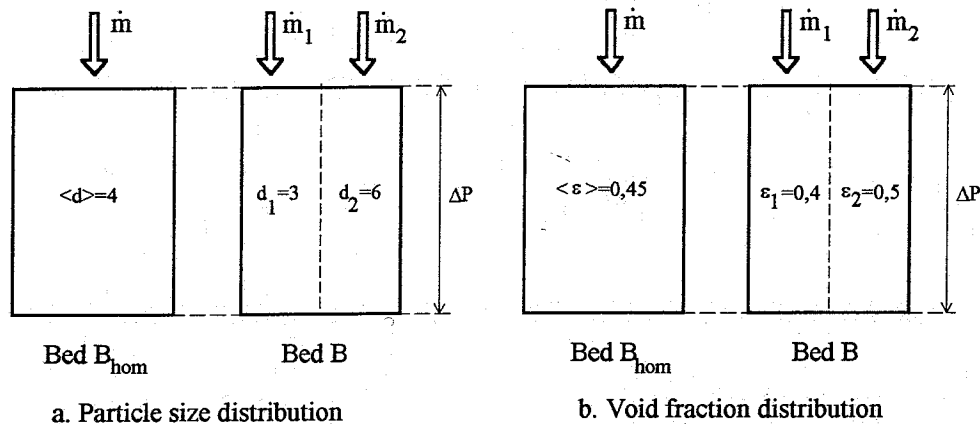


FIGURE 2. INHOMOGENEOUS PACKED BEDS AND THEIR HOMOGENEOUS EQUIVALENT FOR PARTICLE SIZE AND VOID FRACTION DISTRIBUTIONS.

The value of $\langle d \rangle$ is according to definition (13) 4 mm, so we also consider a bed B_{hom} homogeneously made up of only particles with size 4 mm. The two beds B and B_{hom} are operated with the same pressure drop, equal void fractions and equal temperature profiles. We are to compare the performances of bed B and B_{hom} .

With the aid of the model of section two the temperature profiles are computed, as well as temperature dependent properties such as the gas mass density, that enable to compute a pressure drop with the well-established Ergun correlation [10]. For the homogeneous bed with $d=4$ mm and superficial velocity $u_g=0,3$ m/s the pressure drop is calculated to be 1,745 kPa.

For the other two particle sizes the inlet mass flow rate is now varied until the same pressure drop of 1,745 kPa was reached. The resulting inlet velocity is 0,208 m/s for the 3 mm particles and 0,452 m/s for the 6 mm particles.

Temperature profiles being equal because of perfect mixing perpendicular to the flow direction of the gas, cause the term $h_{\text{a,spec}} \cdot (T-\theta)$ of the enthalpy transport length to be proportional to \dot{m} in each part of bed B, with the same proportionality constant². We therefore derive that the enthalpy transport lengths in both parts are the same, and identical to the one of bed B_{hom} .

The sole difference between bed B and bed B_{hom} is the fact that the total mass flow rates are different. The total mass flow rate of bed B is 10% higher than the one of bed B_{hom} . With the same cycle frequency more air is cleaned in the unmixed bed B. On the other hand, a higher mass flow rate with the same void fraction corresponds to a higher profile shift velocity in bed B, see equation (12). The high temperature region therefore reaches

sooner the outlet or, to put it differently, the time-averaged outlet temperature of the gas is higher.

For the same cycle time and equal mass flow rate ($u_g=0,33$ m/s), the homogeneous bed B_{hom} is computed to have 16% lower average convective heat losses than the inhomogeneous bed B, that needs 13% lower fan power. Since the thermal efficiency is very high, the extra fan power required in many cases is more significant than the extra losses, that can still be reduced by lowering the cycle time.

Whether bed B or bed B_{hom} performs better depends on the operation conditions required.

Void fraction.

For studying the effect of void fraction maldistribution on performance, the same line of reasoning is employed. The particle diameter is kept constant at 4 mm while the void fraction is varied: 0,4 and 0,5 with average 0,45, see figure 2b. Again, the enthalpy transport lengths are the same and for equal pressure drop the inlet velocities are computed to be different, i.e. 0,3 m/s for ε equal to 0,4 and 0,421 and 0,565 for void fractions of 0,45 and 0,5 respectively. The result for equal pressure drop is 3% higher mass flow rate for the unmixed bed B³. For equal velocity (u_g in B_{hom} increased from 0,421 to 0,43 m/s), convective heat losses from bed B_{hom} are computed to be 13% lower than from bed B. So maldistribution of void fraction reduces the performance of a regenerator.

In these two examples the enthalpy transport length and the profile shift velocity have successfully been considered to directly analyze and compare the performances of different packed beds.

² This is easily seen from the differential equation (3) representing the energy balance of the gas phase.

³ Since $\{1/2 \cdot (0,3 + 0,565) - 0,421\} / 0,421 \approx 3\%$

5. CONCLUSIONS

A theoretical model for the flow of a gas mixture at relatively high speed through a packed bed with steep temperature gradients and with exothermic reactions taking place is used to study the performance of packed beds. The model is based on new analytical solutions for complex boundary conditions in bed slices, making the computational model fast and facilitating the performance study of the bed.

Many parameters govern flow and heat transfer processes in a packed bed regenerator, which often hampers optimization. An enthalpy transport length and the shift velocity are presented as convenient optimizing tools. The thermal efficiency is of less value in this respect.

The effect of particle size dispersion is such that less fan power is required for the same gas mass flow rate if particles are inhomogeneously packed. Convective heat losses in this case slightly increase if cycle time is not reduced. Inhomogeneous packing of the bed, corresponding to notable void fraction variations, has a similar effect on regenerator performance.

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NOMENCLATURE

a_{spec}	Heat exchanging area per unit bed volume	$[\text{m}^{-1}]$
c	Heat capacity	$[\text{J}/(\text{kg}\cdot\text{K})]$
d	Diameter	$[\text{m}]$
h	Convective heat transfer coefficient	$[\text{W}/(\text{m}^2\cdot\text{K})]$
I_0	Modified Bessel function of the first kind	$[-]$
k	Coefficient in dimensionless boundary condition for solid phase	$[-]$
l	Coefficient in dimensionless boundary condition for solid phase	$[-]$
\dot{m}_g	Mass flow rate of gas through packed bed	$[\text{kg}/\text{s}]$
M_i	Mass of solid particles with diameter d_i	$[\text{kg}]$
Q	Dimensionless heat source	$[-]$
\dot{q}	Heat source	$[\text{W}/\text{m}^3]$
S_{fr}	Cross sectional area of packed bed, perpendicular to flow direction	$[\text{m}^2]$
T	Temperature of gas phase	$[\text{K}]$
T_g	Dimensionless gas temperature	$[-]$
T_p	Dimensionless surface temperature of packed bed	$[-]$
t	Time	$[\text{s}]$
u_g	Superficial gas velocity	$[\text{m}/\text{s}]$
V	Volume	$[\text{m}^3]$
V_g	Shift velocity	$[\text{m}/\text{s}]$
Δx	Enthalpy transport length	$[\text{m}]$
z	Axial coordinate	$[\text{m}]$
Δz	Thickness of slice of packed bed	$[\text{m}]$

Greek:

ε	Void fraction: Volume of gas per unit volume of packed bed	[-]
η	Dimensionless time	[-]
η_{th}	Thermal efficiency, equation 9	[-]
ρ	Mass density	[kg/m ³]
θ	Temperature at surface of solid packing material	[K]
ξ	Dimensionless length	[-]

Subscripts:

g	Gas phase
p	Particles