MODELLING OF ROTARY KILNS

PROEFSCHRIFT

ter verkrijging van
de graad van doctor aan de Universiteit Twente,
op gezag van de rector magnificus,
Prof.dr. F.A. van Vught,
volgens besluit van het College voor Promosies
in het openbaar te verdedigen
op vrijdag 26 januari 2001 te 16:45 uur

door

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geboren op 13 juni 1959
te Pietersburg, Zuid-Afrika
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Dr.ir. A.B.M. Heesink
to my father
SUMMARY

This work primarily covers mass transfer in rotary kilns. The most important contribution is the identification of the dimensionless numbers that contribute to mass transfer in rotary kilns, and their relation to each other. In this respect, this work is also applicable to heat transfer in rotary kilns, where analogous dimensionless numbers can be identified. Another important contribution is the recognition of the important contribution of the passive layer in mass transfer.

Fig. 1 shows the nomenclature used in this summary, and the various zones found within a rotary kiln. In the active layer, particles slide over each other in granular flow. They are returned to the top part of the bed in the passive layer, where particles move as a solid mass concentrically around the axis of the kiln.

Figure 1. Radial cross section of a kiln, showing the nomenclature.

The first part of the work considers the reaction of the gas as it moves in plug flow through the passive layer within the interparticle voids; the active
layer is considered to be infinitely thin. The effectiveness factor $\eta$ for the bed is found to be dependent on the bed fill (represented by $h/R$) and the ratio of the rate constant to the rotation speed ($k/\omega$), shown in Fig. 2. Comparison against experimental data shows that this relationship is only applicable to slower reactions ($k/\omega < 1$).

![Figure 2. Effectiveness factor increases with decreasing bed fill.](image)

The effect of diffusion within the passive layer was investigated based on a two-dimensional continuity equation for the radial cross section, again with the assumption of an infinitely thin active layer. This requires the use of an additional dimensionless constant: either a Peclet number ($\epsilon \omega R^2/D_e$) or a Thiele modulus ($R \sqrt{k/D_e}$). It was shown that for practical values of Peclet number, diffusion within the passive layer can be neglected, but at higher values of $k/\omega$, active layer modelling is necessary to correctly predict mass transfer.

By making the (justified) assumption that the velocity gradient of particles ($m$) within the active layer is constant at any point, the active layer depth can be estimated at any distance $x$ along the bed surface using
The action of particles sliding over each other promotes movement of gas through the interparticle voids. A correlation for an active layer dispersion coefficient has been derived:

\[
D_{al} = 0.246f_m d_p^2 + \varepsilon^2 D_a
\]

where \( f \) is a directionality factor; with equal dispersion in all directions, \( f = 1/6 \). With fast reactions mass transfer can be estimated in terms of a mass transfer coefficient on the bed surface using:

\[
k_s = \sqrt{D_{al} \varepsilon k}
\]

This does not fully predict the dependence of mass transfer on rotation speed. It is also necessary to take into account the periodic renewal of gas in the voids between the particles within the active layer. This is done by adapting the value of the kinetic constant using

\[
k' = m(1 - e^{-k/m})
\]

where \( k' \) is the effective rate constant applicable to the active layer region. Using \( m = 70 \omega \), and the above assumptions, a good prediction of mass transfer could be obtained for fast reactions.

Finally, a model was developed that built on the principles established for both the active and passive layers. This combined model assumes continuity of concentration at the interface of the active and passive layers, where gas is entrained into the bed through rotation (Fig. 3).
Figure 3. A typical concentration profile using the combined model (clockwise rotation).

This model predicts that it is seldom possible to use either the passive layer model alone, or the active layer model alone to accurately predict mass transfer, and that usually both regions in a rotary kiln contribute significantly to mass transfer. We also find that when dispersion effects dominate over molecular diffusion effects in the active layer (expected for particle size > 3 mm), then scale-up effects can be represented by the ratio of kiln radius to particle diameter ($R/d_p$) instead of the active layer Thiele modulus.

An example of the predictions of the combined model is given in Fig. 4 for a bed fill of 5.2%. For such a shallow bed, the effectiveness factor is strongly dependent on $R/d_p$. It is also clear from the shape of the curves that the passive layer model alone is inadequate at predicting the effectiveness factor, even for low $k/\omega$. 
Figure 4. Effectiveness factors for a rotary kiln with bed fill 5.2%. The thick line shows the prediction of the passive layer model (Chapter 2).

The value of the work lies in two aspects: firstly, the recognition of the contribution of the passive layer towards mass transfer, and secondly, the relative simplification of the modelling approach by identifying and using appropriate dimensionless numbers to characterise the system. In this way, at a given bed depth, the effectiveness factor is characterized by a single graph such as that of Fig. 4. For heat transfer, a similar analogy should be possible.
SAMENVATTING


Figuur 1. Dwarsdoorsnee van een roterende oven

Figuur 1 laat de verschillende zones zien, zoals die in een roterende oven worden onderscheiden. In de actieve laag bewegen de deeltjes langs en over elkaar heen naar beneden. Door de roterende beweging van de oven
worden de deeltjes die het eind van de actieve laag hebben bereikt, via een roterende beweging door de onderliggende passieve laag weer naar het bovenste gedeelte van de actieve laag getransporteerd. De passieve laag roteert hierbij als een star lichaam om de as van de oven. Zowel in de actieve als in de passieve laag kunnen de deeltjes reageren met het gas dat vanuit het “lege” deel van de oven (het zogenaamde freeboard) continu wordt aangevoerd.

Na een inleiding over de relevantie van roterende ovens voor de industrie, wordt in het tweede hoofdstuk van dit proefschrift het transport en de reactie van het gas in de passieve laag beschouwd. Het gas, opgesloten tussen de deeltjes, beweegt daarbij als het ware in propstroom concentrisch door de passieve laag. De actieve laag wordt daarbij oneindig dun verondersteld. Er wordt een effectiviteitsfactor \( \eta \) gedefinieerd als de verhouding tussen de werkelijke omzettingssnelheid in het bed en de snelheid die zou worden bereikt wanneer overal in het bed de concentratie van de gasvormige reactant gelijk zou zijn aan de concentratie in het freeboard boven het bed. Deze blijkt sterk afhankelijk te zijn van de vulhoogte van de oven (1\( - h/R \)) en de verhouding tussen de reactiesnelheids-

![Effectiviteitsfactor bij verschillende vulhoogtes.](image)

\( \eta \) als \( k/\omega \)

Fill %

- 50%
- 37%
- 25%
- 14%
- 5.2%
- 1.9%
- 0.7%
- 0.17%

**Figuur 2.** Effectiviteitsfactor bij verschillende vulhoogtes.
constante en de rotatiesnelheid \((k/\omega)\), zoals weergegeven in Figuur 2. Een vergelijking met experimentele data (afkomstig uit de literatuur en eigen data) laat zien dat de gevonden afhankelijkheid alleen toepasbaar voor relatief langzame reacties \((k/\omega < 1)\).

In het derde hoofdstuk wordt het effect van diffusie binnen de passieve zone bestudeerd met behulp van de continuïteitsvergelijking voor een dwarsdoorsnee van de roterende oven. Hierbij is wederom aangenomen dat de actieve laag oneindig dun is. Dit model resulteert in een extra dimensieloos kental: ofwel een Peclet-kental \((\varepsilon \omega R^2/D_e)\) ofwel een Thiele modulus \((R \sqrt{k/D_e})\). In de praktijk blijkt diffusie in de passieve zone een verwaarloosbaar kleine bijdrage aan het stoftransport te leveren. Voor hoge waarden van \(k/\omega (k/\omega > 1)\) blijkt ook dit aangepaste passieve-zone model niet in staat om de experimentele resultaten te beschrijven. Geconcludeerd wordt dan ook dat een nadere beschouwing van de processen die optreden in de actieve laag noodzakelijk is.

In hoofdstuk 4 worden de hydrodynamica van en de stofoverdrachtsprocessen in de actieve laag nader onder de loep genomen. Door de (gerechtvaardigde) aannemer dat de snelheidsgradiënt van de deeltjes \((m)\) binnen de actieve laag constant is op elke positie binnen die laag, kan de dikte van de actieve laag worden afgeschat voor elke positie \(x\) (zie Figuur 1) langs het grensvlak volgens:

\[
d = \sqrt{\frac{R^2 - x^2 - h^2}{m/\omega - 1}}
\]

De beweging van de deeltjes over en langs elkaar heen veroorzaakt gastransport door het steeds weer creëren en opbreken van interstitiële ruimtes. Op basis hiervan is een overall dispersiecoëfficiënt voor de gasfase afgeleid:

xii
\[ D_{al} = 0.246 fm \, d_p^2 + \varepsilon^2 D_a \]

In deze vergelijking verdisconteert \( f \) de richting van het transport. Voor gelijke dispersie in elk van de orthogonale richtingen geldt \( f = 1/6 \). De symbolen \( d_p \) en \( D_a \) staan respectievelijk voor de deeltjesdiameter en de moleculaire diffusiecoëfficiënt van het reagerende gas, terwijl \( \varepsilon \) de (gemiddelde) porositeit van de actieve laag voorstelt. In het geval van snelle gas-vast reacties kan de stofoverdracht worden geïnterpreteerd in termen van een stofoverdrachtscoëfficiënt \( k_s \), betrokken op het totale bedoppervlak:

\[ k_s = \sqrt[D_a]{\varepsilon k} \]

Hierbij wordt echter het effect van de rotatie-snelheid op het stoftransport niet volledig verdisconteerd. Het is noodzakelijk om ook het effect van de periodieke verversing van gas in de holtes tussen de deeltjes te verdisconteren. Dit kan worden gedaan door aanpassing van de kinetiekconstante, volgens:

\[ k' = m\left(1 - e^{-k/m}\right) \]

waarin \( k' \) de effectieve reactiesnelheidsconstante voor de actieve zone voorstelt. Voor \( m=70\omega \) en bovengenoemde aannames wordt een goede beschrijving van de in de literatuur gerapporteerde experimentele data gevonden.

Tot slot wordt in hoofdstuk 5 een model ontwikkeld dat gebaseerd is op de eerder genoemde beginselen voor de actieve en passieve zones. Dit gecombineerde model gaat uit van een continu concentratie-profiel aan het grensvlak tussen de actieve en de passieve zones. Het gecombineerde
model voorspelt dat slechts bij hoge uitzondering alleen het passieve zone model of alleen het actieve zone model voldoet. Meestal zijn beide zones belangrijk in het beschrijven van het stoftransport in een roterende oven. Wanneer in de actieve zone moleculaire diffusie te verwaarlozen is ten opzichte van gasfase dispersie (en dit is aannemelijk voor deeltjes groter dan 3 mm), kunnen opschaaleffecten worden verdisconteerd via de verhouding van ovenstraal ten opzichte van deeltjesdiameter ($R/d_p$).

In Figuur 4 is een voorbeeld van een berekeningsresultaat van het gecombineerde model gegeven voor een kleine vulhoogte. De effectiviteits-

![Diagram](image)

**Figuur 3. Een typisch concentratieprofiel volgens het gecombineerde model (klokgewijze rotatie)**

**Figuur 4. Effectiviteitsfactor voor een roterende oven met een vulgraad van 5.2%. Dikke lijn geeft de voorspelling van het passieve-zone model.**
factor blijkt sterk af te hangen van \((R/d_p)\). Ook blijkt duidelijk dat het passieve-zone model alleen niet in staat is de effectiviteitsfactor goed te beschrijven, zelfs niet voor lage waarden van \(k/\omega\).

De resultaten van dit proefschrift tonen het belang aan van zowel de actieve als de passieve laag voor het totale stoftransport. De term “passief” is zeker niet altijd op zijn plaats. Door enkele vereenvoudigende aannamen en het gebruik van de juiste dimensieloze kentallen kan de stofoverdracht in roterende ovens goed worden gekarakteriseerd. Op deze wijze kan, voor een gegeven vulgraad van de oven, de effectiviteitsfactor als functie van de procesparameters in een enkele grafiek worden weergegeven. Voor warmtetransport is een analoge analyse mogelijk.
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**Acknowledgements**

**Curriculum Vitae**
Chapter 1 - Introduction

Rotary kilns are found in many processes that involve solids processing. These include drying, incineration, mixing, heating, cooling, humidification, calcination, reducing, sintering and gas-solid reactions (Jauhari et. al. 1998). The most common and industrially important application of rotary kilns is in cement production; all major producers use the rotary kiln as their equipment of choice. Cement kilns can be very large. Although this is not a cement kiln, Fig. 1 shows a 500 tpd lime kiln from Greer lime.

Another important application of rotary kilns is for the incineration of waste materials. Rotary kilns are popular for this role because of their flexibility. They can handle a large variety of feed materials, with variable calorific value, and removal of waste solids at the exit presents no problems. Typically, hazardous waste incinerators operate with relatively deep beds, and have a secondary combustion chamber after the rotary kiln to improve the heterogeneous combustion of waste (Rovaglio et. al. 1998). An example of such a hazardous waste incinerator is shown in Fig. 2.
In the mineral processing industry, there are many applications of rotary kilns: Some of the applications that have been published are magnetite oxidation (Davis, 1996), induration of iron ore pellets (Young et. al. 1979), coke calcining, (Bui et. al. 1995) and drying (Friedman & Marshall, 1949).

Rotary kilns are amongst the most well-established unit operations in the process industry, yet are amongst the least understood. They can be used for 3 purposes: heating, reacting and drying of solid material, and in many cases, they are used to achieve a combination of these aims. In the design of kilns, there are four important aspects to consider from a process engineering point of view, and these are heat transfer, flow of material through the rotary kiln, gas-solid mass transfer and reaction.

Heat transfer rates amongst the most important of these aspects, because in many cases, it is the heat transfer that limits the performance of the rotary kiln. This is reflected in the number of papers published on the modelling of heat transfer in rotary kilns. However, the first aspect of rotary kilns that was modelled was the movement of material through a rotary kiln. The first such modelling was done by Saeman (1951), based on data measured by Sullivan et. al. (1927).
The subject of mass transfer in rotary kilns has been neglected until Jauhari et. al. (1998) published an article on gas-solid mass transfer in a rotating drum. The purpose of this thesis is to model mass transfer and reaction in rotating drums, but the novel approach adopted by the author is firmly based on the foundations of chemical engineering science, can equally well be applied to the modelling of heat transfer.

**The radial plane vs axial direction**

In the literature on rotary kilns, we typically find diagrams of a radial cross-section through a rotary kiln, such as that shown in Fig. 3. This shows the movement of particles within the bed of a rotary kiln: particles move concentrically with the wall of the rotary kiln in the passive layer until they reach the surface of the bed, where they slide downwards in the active layer. Considering a single particle, this repeated process can cause the particle to move in the axial direction every time it moves down the active layer, as long as the rotary kiln is tilted in the axial direction. This is the basis for models that predict the movement of particles through a rotary kiln in an axial direction. These models can predict the bed fill (fractional area of the bed in a radial cross section) along the length of a rotary kiln as a function of solids feed rate.
In operation, solids heat/cool/react/dry progressively as they move through the kiln. In this thesis, we will refer to the operation as reaction. (This approach encompasses drying as a subset of reaction.) Simultaneously with the movement of the particles, gas may be flowing through the freeboard of the kiln, either co-currently or counter-currently. Therefore, at any point along the length of a rotary kiln, we can expect a different set of conditions to apply to both the solid and the gas. We will assume (as most researchers in the field do) that if we can understand and model the processes that occur in the radial cross-section, then we can use that radial cross-section as a differential element to model the rotary kiln as a whole in the axial direction using established engineering practices.
The flow regimes in rotary kilns

The nature of the solids flow in rotary kilns can change significantly with particle characteristics, kiln size, rotation speed and bed fill. Heinen et. al. (1983a,b) did useful work on this subject, introducing the concept of a Bed Behaviour Diagram (Fig. 4).

![Bed Behaviour Diagram](image)

*Figure 4. An example of a bed behaviour diagram; each material has its own characteristic bed behaviour diagram.*

For rotary kilns with shallow beds in particular, the bed is prone to slippage between the wall of the kiln and the bed. For deeper beds, the motion is either rolling, as described in Fig. 3, or at lower rotation speeds, slumping. In slumping mode, the angle of repose of the material increases as the static bed rotates, until a segment of material shears off and comes to rest in the bottom half of the kiln. This process occurs repeatedly, and is less desirable than rolling mode for heat and mass transfer. In rolling mode, the bed surface is continuously renewed, while in slumping mode, the renewal occurs at fixed intervals.
At higher bed fills and rotation speeds, the momentum of the particles as they reach the surface of the bed becomes important, and they fall down the surface of the bed in a trajectory. This causes a distortion in the surface of the bed (which is normally flat). Further increasing the rotation speed finally results in centrifugation, where particles are flung to the wall of the kiln. This is also an undesirable regime of operation, because the particles do not mix, resulting in poor mass and heat transfer.

Each of these modes of operation produces different conditions for particles within the rotary kiln, and therefore should be modelled differently. In this thesis, and in most literature articles, modelling is applicable to the rolling mode – the preferred mode of operation industrially.

**The need for the modelling of rotary kilns**

The need for a deep understanding of kilns and the need for effective fundamental models has been expressed very well by Barr *et. al.* (1989):

“Rotary kilns are ubiquitous fixtures of the metallurgical and chemical process industries. Despite challenges from newer and more specialised gas-solids reactors, they continue to find applications in the drying, heating (or cooling), calcining, reducing, roasting and sintering of a variety of materials. Rotary kilns can handle feed stocks with broad particle size distributions or whose physical properties change significantly during processing, while the long residence time of the material within the kiln promotes uniform product quality. In addition, dirty fuels often are utilised without serious product contamination, and multiple fuel capability is possible. Paradoxically, this versatility, which has in the past ensured the survival of the rotary kiln, now threatens its future. Because a thorough understanding of the processes occurring within rotary kilns has not been a prerequisite for their apparently satisfactory operation, research has not
progressed apace with competing, less tolerant, reactors. Until all the internal processes are understood and become predictable, rotary kilns will remain in the position of operating below their optimal performance in an increasingly sophisticated marketplace.”

An overview of this work

This work considers mass transfer in rotary kilns, by starting in Chapter 2 with a passive layer model.

In the passive layer model, we consider the movement of gas through that part of the bed that moves as a concentrically around the axis of the kiln. The gas is entrained in the interstices between the particles, reacting as it is transported through the bed. The model was based on chemical reactor theory, because mass transfer can be considered as a subset of reactor theory – mass transfer becomes limiting for very fast reactions. In this first model we show that the effectiveness factor of the bed is dependent on just two variables: the ratio of the rate constant to the angular velocity \((k/\omega)\), and the bed fill.

In the next chapter we consider passive layer diffusion, which involves solving a partial differential equation in the two dimensions of the radial cross section. This model is not flexible enough to be used over a wide range of \(k/\omega\) because of the need to consider the active layer. However, it does provide useful insight into the dimensional groups that are required to describe scale-up effects in a rotary kiln: either a Peclet number \((\varepsilon \omega R^2 / D_e)\) or a Thiele modulus \((R \sqrt{k / D_e})\) should be used in addition to \(k/\omega\) and bed fill.
In Chapter 4, some fundamental models for the *active layer* are presented: a hydrodynamic model that predicts the depth and shape of the active layer, and a dispersion model for predicting mass transfer within the active layer.

In the final chapter, a *combined model* is presented that expands on the theory established in Chapter 4 for the active layer, and uses the principles of Chapter 2 for modelling the passive layer. We find that it is seldom possible to use either the passive layer model alone or active layer model alone to accurately predict mass transfer, and that usually both regions in a rotary kiln contribute significantly to mass transfer. We also find that when dispersion effects dominate over molecular diffusion effects in the active layer (expected for particle size > 3 mm), then scale-up effects can be be represented by the ratio of kiln radius to particle diameter \( (R/d_p) \) instead of the Thiele modulus.
REFERENCES


Chapter 2 - Mass transfer in rolling rotary kilns: a novel approach

ABSTRACT

A novel approach to modeling mass transfer in rotary kilns is explored. The movement of gas in the interparticle voids in the bed of the kiln is considered, where particles move concentrically with the geometry of the kiln and gas is entrained by these particles. A reactor modeling approach has been used to derive effectiveness factors for the bed as a function of bed fill, reaction kinetics and rotation speed. In many cases, the entrained gas becomes depleted within the bed, leading to a simplified model for the bed effectiveness factor. Experimental data confirms the validity of this model for slower rates. At faster rates, mass transfer can be much higher than the model predicts, indicating that other mechanisms, such as dispersion or diffusion are also important in these conditions.

1. INTRODUCTION

Rotary kilns are used industrially in many applications such as drying, incineration, mixing, pre-heating, humidification, calcining, reducing, sintering and gas-solid reactions (Barr, 1989; Jauhari, 1998).

For many rotary kilns, heat transfer is the limiting factor, both in the heating section of the rotary kiln, and in the reaction zone (Barr, 1989). Consequently, most focus in the literature has been directed at understanding the heat transfer processes in rotary kilns. It is important
though to understand all of the processes that occur in rotary kilns on a fundamental level before rotary kilns can be designed and operated optimally.

With physical processes like drying and humidification for example, mass transfer is also important, as well as in gas-solid reacting systems with high specific reaction rates such as incineration. In this work we will examine the mass transfer in rolling rotary kilns and propose a novel approach to describe the phenomena that determine the rate of mass transfer.

2. MODELLING STUDIES

The first published experimental studies on rotary kilns recorded the relationship of rotation speed and kiln inclination on bed depth and solids residence time (Sullivan et al, 1927). A model was later developed based on the assumption that particles in a rolling bed move in a circular motion with the rotation of the kiln, and then fall down the surface of the bed in a thin layer (Saeman et al, 1951). The time taken to fall down the surface was assumed to be small compared to the time for a particle to move with the kiln from the bottom half to the top half of the bed. Using the geometry of an inclined rotary kiln, the angle of inclination necessary to maintain a constant bed height over the length of the rotary kiln could be determined for a given rotation speed. This basic model predicted the original data (Sullivan et al, 1927) well, and the model was further refined to predict axial movement of particles with different bed fills, taking into account the time for particles to fall down the surface of the bed (Kramers and Crookewit 1952). In later work that specifically measured the movement of particles at the surface of the kiln, the validity of the model of Kramers and Crookewit (1952), with minor exceptions, was confirmed (Lebas et al, 1995). More recently, the same fundamental model of the path of particles
was shown to be correct in a study of particle motion where there is no nett axial flow of particles in a rotating drum (Spurling et al, 2000).

The conditions that determine the mode in which a rotary kiln operates - slipping, slumping, rolling, cascading, cataracting, centrifuging - have been extensively examined, and can be represented using a Bed Behavior Diagram (Heinen et al, 1983a). An example of a Bed Behavior Diagram is shown in Fig. 1. In this paper, we will focus on kilns operating in the preferred mode for most circumstances - rolling.

![Bed Behavior Diagram](image)

*Figure 1. An example of a Bed Behavior Diagram (Heinen et al 1983a).*

The time for a colored particle to move down the surface of the bed was measured with video photography (Lebas et al, 1995), and this typically takes 15 - 30% of a full cycle. However, this ratio does not change with rotation speed, and the velocity of the particles moving down the surface of the bed was interpreted to be directly proportional to rotation speed (Lebas et al, 1995).
The layer of particles moving down the face of the bed leads to improved mass transfer, therefore the surface layer is often referred to as the “active layer” (See Fig. 2). Heinen et al (1983a) measured particle velocities inside this layer as well as the thickness of it, and found that it is relatively thin for beds with a depth that is much larger than the diameter of the particles. In most cases, the active layer is typically less than 8 particle diameters deep, except for deep beds with small particles, where the depth of the active layer is less than 12% of the bed depth, falling to as low as 4% for slowly rotating kilns (Heinen et al, 1983a).

![Diagram of rotary kiln zones](image)

*Figure 2. The various zones within a rotary kiln.*

### 3. MODEL DEVELOPMENT

For most of the volume of the bed of a rotary kiln (the passive layer), particles move concentrically with the wall of the bed, with no slippage between particles. As the particles approach the bed surface due to the kiln rotation, they slip down the face of the bed, and come to rest near the
bottom of the surface of the bed. We will attempt to quantify the volumetric flow rate and reaction rate of the gas that is trapped in the interparticle voids in the passive layer.

3.1 Gas volumetric flow rate

The volume of entrained gas can be determined for a single rotation of the kiln (for convenience, the bed has been represented horizontally):

\[
dx = (\sin \theta) R \, d\theta = (c/R) R \, d\theta = cd\theta
\]

\[
dA_x = \frac{1}{2}c \, dx = \frac{1}{2}c^2 \, d\theta
\]
Chapter 2 – Passive layer model

The area useful for the entrainment of gas is proportional to the void fraction of gas in the bed:

$$dA_{\text{sig}} = \frac{\varepsilon}{2}(R^2 - h^2)d\theta$$  \hspace{1cm} (3)

Integrating this expression over 1 radian, and converting to angular velocity, the volume flow rate of gas is:

$$Q = \frac{1}{2}\varepsilon\omega L(R^2 - h^2)$$  \hspace{1cm} (4)

This gives the volume flow rate of entrained gas through the bed of a rotary kiln. By estimating the concentration of the gas as it leaves the bed, the overall reaction rate can be determined.

To understand how the active layer influences the movement of gas, consider a line extending from the radial centre of the kiln, passing perpendicularly through the bed surface to the deepest part of the bed. All entrained gas will pass this line. If the active layer is infinitely thin, the flow rate $Q$ can easily be shown to be as in Eq. (4). A mass balance of the particles also means that the particles will move infinitely fast over the surface of the bed with the assumption of an infinitely thin active layer. In practice, the particles fill move down at a finite velocity, with a finite active layer thickness. However, for the purposes of this paper, the admittedly unrealistic assumption of an infinitely thin layer, not affecting the entrainment of gas perpendicular to the bed surface, will be taken through to its conclusion.
3.2 Nature of concentration profiles within the bed

The movement of the gas in the voids is assumed to be a circular arc, following the geometry of the kiln, until the gas reaches the surface of the bed again (Saeman et al., 1951). For a half-filled kiln, the retention time of such gas will be the same, regardless of the radius of the arc. Furthermore, this retention time for the half-full kiln will be the time it takes for half of a revolution: \( \tau = \pi/\omega \). If we assume that the gas following this path will move in plug flow, we can expect a concentration profile fitting an exponential decay. Therefore, we can expect the concentration profile within the bed of a rotary kiln to be as follows:

Diffusion and dispersion will no doubt play a role in the kiln, especially in the centre of the kiln, where several concentrations converge. In this part, the effect of the active layer will be important, and dispersion effects will predominate. The inclusion of dispersion will require a more complex model, not discussed here.

**Figure. 4.** For a half-filled kiln, the residence time at any radius is equal, yielding concentration profiles shown here for a first-order reaction with \( k/\omega = 1 \).
For bed fills of less than 50%, the residence time at each radius will be different, and the expected concentration profiles can be calculated.

\[
\tau(\theta, R) = \frac{\theta - \theta_0}{\omega} = \frac{\theta - \sin^{-1}(h/r)}{\omega}
\]

\[
\tau = \frac{\theta - \sin^{-1}(h/r)}{\omega}
\]

\[
\frac{h}{r} = \sin(\theta - \omega\tau)
\]

\[
= \frac{y}{r} \cos \omega\tau - \frac{x}{r} \sin \omega\tau
\]

\[
y = x \tan(\omega\tau) + \frac{h}{\cos(\omega\tau)}
\]

This applies to any reaction kinetics, and different reaction kinetics will simply determine the spacing of the lines. As an example, let us consider a 1st order reaction.
-\( P_A = ekC_A \) \hspace{1cm} (10)

where \( P_A \) is the reaction rate of \( A \) per unit of bed volume and \( k \) is the reaction rate constant with respect to the gas phase. Then

\[
k_\tau = -\ln \left( \frac{C_A}{C_{A0}} \right) \hspace{1cm} (11)
\]

Substitute \( \tau \) into Eq. (4) for 1st order iso-concentration lines, shown in Fig. 6.

![Figure 6. Concentration profiles for \( k/\omega = 1 \) at \( C_A/C_{A0} \) intervals of 0.1.](image)

3.3 Effectiveness factors

For convenience, such concentration profiles can rather be expressed by an effectiveness factor. This factor is defined as the ratio of the actual rate of the reaction over the bed to the rate that would be obtained if the whole bed were exposed to gas at the highest gas concentration - the concentration above the bed.
In the general case of kilns with a given fill, actual rate is the average rate over the entire cross-sectional area. (See Fig. 6.)

\[
\int_V P_A(r, \theta, z) dV_{\text{bed}} = \left[ \int_{\theta_1}^{\theta_2} \int_0^{\theta_{\text{in}}} e^{\frac{k}{\theta} \sin^{-1}(h/r)} r dr d\theta \right] dz \quad (12)
\]

\[\theta_1 = \sin^{-1}(h/R) \quad \theta_2 = \pi - \sin^{-1}(h/R) \quad r_1 = h / \sin \theta \quad (13)\]

From the definition of the effectiveness factor and realising that the cross-sectional area of the bed equals \( \frac{1}{2} R^2 (\theta_2 - \theta_1 - \sin(\theta_2 - \theta_1)) \),

\[
\eta = \frac{\int_V P_A(r, \theta, z) dV_{\text{bed}}}{\varepsilon k C_{\text{av}} V_{\text{bed}}} \quad (14)
\]

\[
\eta = \frac{2}{R^2 (\theta_2 - \theta_1 - \sin(\theta_2 - \theta_1))} \int_{\theta_1}^{\theta_2} \int_0^{\theta_{\text{in}}} e^{\frac{k}{\theta} \sin^{-1}(h/r)} r dr d\theta \quad (15)
\]

This function is represented graphically in Fig. 7 for different bed fillings as a function of the \( k / \omega \) ratio. It is interesting to note that the size of the rotary kiln is irrelevant using this model. The only variables in this model are the bed fill and \( k / \omega \). Later in the paper, we will discuss the role of other parameters that are dependent on the scale of the rotary kiln.
Figure 7. Effectiveness factors increase as bed fill decreases, and are independent of bed radius $R$. Fill fractions shown are 0.5 (solid line), 0.37, 0.25, 0.14, 0.052, 0.019, 0.007 and 0.0017 (thin line).

As expected, for a given $k/\omega$, the effectiveness increases for shallower beds. For the special case of a half-filled cylinder, $h = 0$:

$$\eta = \frac{2}{\pi R^2} \int_0^{\pi} R^2 e^{-\frac{k}{\omega} \theta} d\theta$$

(16)

$$= \frac{2}{\pi R^2} \int_0^{\pi} R^2 e^{-\frac{k}{\omega} \theta} d\theta$$

(17)

$$= \frac{\omega}{k\pi} \left( 1 - e^{-\frac{k}{\omega}} \right)$$

(18)

This is also easily derived from first principles. Taking a mole balance for a given volume of bed, where $C_A$ is the concentration of gas leaving the bed,
Chapter 2 – Passive layer model

\[-P_A dV_{\text{bed}} = Q(C_{A0} - C_A)\]  \hspace{1cm} (19)

\[\eta \kappa C_{A0} V_{\text{bed}} = Q(C_{A0} - C_A)\]  \hspace{1cm} (20)

\[\eta \kappa A_{\text{bed}} L = Q \left(1 - \frac{C_A}{C_{A0}}\right)\]  \hspace{1cm} (21)

\[\frac{\eta \kappa \pi R^2 L}{2} = \frac{\varepsilon \omega R^2 L}{2} \left(1 - e^{-\frac{k \tau}{\omega}}\right)\]  \hspace{1cm} (22)

\[\eta = \frac{\omega}{k \pi} \left(1 - e^{-\frac{k \tau}{\omega}}\right)\]  \hspace{1cm} (23)

4. TOTAL DEPLETION MODEL

The double integral of equation (15) is cumbersome to work with; a simplified approach for estimating \(\eta\) would be more useful. For high \(k/\omega\) values such a simple approach is quite possible.

For high \(k/\omega\), the gas leaving the system at the surface of the bed can be assumed to be fully depleted (or saturated, in the case of drying). In this case, we assume complete reaction and use Eq. (4) to estimate the volume flow rate of entrained gas.

Choosing 95% conversion as the criterion, \(C_A/C_{A0} = 0.05\).

\[k \tau = -\ln \left(\frac{C_A}{C_{A0}}\right) = 3\]  \hspace{1cm} (24)
For a half-filled kiln, all paths through the kiln have the same residence time, and $\tau = \pi/\omega$. For lesser fill fractions, we will choose $\frac{1}{2}$ of the maximum angle. The angle subtended by this path is:

$$\theta = \frac{1}{2} \left( 2\cos^{-1}(h/R) \right)$$  \hspace{1cm} (25)$$

$$\tau = \theta / \omega = \frac{\cos^{-1}(h/R)}{\omega}$$  \hspace{1cm} (26)$$

The criterion for using the simplified model is therefore

$$\frac{k}{\omega} > \frac{3}{\cos^{-1}(h/R)}$$  \hspace{1cm} (27)$$

Using an effectiveness factor and assuming total conversion,

$$- P_A V_{bed} = QC_{x0}$$  \hspace{1cm} (28)$$

$$\eta k C_{x0} V_{bed} = \frac{\epsilon \omega}{2} (R^2 - h^2) L C_{x0}$$  \hspace{1cm} (29)$$

$$\eta k = \frac{\omega}{2 A_x} \left( R^2 - h^2 \right)$$  \hspace{1cm} (30)$$
Table 1. The error caused by using the simplified model at the criterion values is less than 6% in all relevant cases.

<table>
<thead>
<tr>
<th>h/R Fill fraction</th>
<th>k/\omega \quad (Eq. 15)</th>
<th>η \quad (m^2, basis R = 1m)</th>
<th>η, total conversion model (Eq. 30)</th>
<th>Error (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0 0.5</td>
<td>1.91</td>
<td>0.164</td>
<td>1.571</td>
<td>0.1667</td>
</tr>
<tr>
<td>0.2 0.374</td>
<td>2.19</td>
<td>0.1849</td>
<td>1.173</td>
<td>0.1867</td>
</tr>
<tr>
<td>0.4 0.252</td>
<td>2.59</td>
<td>0.2006</td>
<td>0.793</td>
<td>0.2047</td>
</tr>
<tr>
<td>0.6 0.142</td>
<td>3.24</td>
<td>0.214</td>
<td>0.447</td>
<td>0.2211</td>
</tr>
<tr>
<td>0.8 0.052</td>
<td>4.66</td>
<td>0.2259</td>
<td>0.164</td>
<td>0.2361</td>
</tr>
<tr>
<td>0.9 0.0187</td>
<td>6.65</td>
<td>0.2313</td>
<td>0.0587</td>
<td>0.2432</td>
</tr>
<tr>
<td>0.95 0.00666</td>
<td>9.45</td>
<td>0.2338</td>
<td>0.0209</td>
<td>0.2466</td>
</tr>
<tr>
<td>0.98 0.00169</td>
<td>14.97</td>
<td>0.2354</td>
<td>0.00532</td>
<td>0.2487</td>
</tr>
</tbody>
</table>

5. MODEL VALIDATION

5.1 Literature data

Jauhari et al. (1998) measured mass transfer in a rotating drum by measuring the evaporation rate of decane from impregnated alumina particles. Most of the measurements were done using shallow beds with baffles (flights) fitted to the rotary kiln. This gives rise to a geometry that is not well described by the model in this paper. However, one set of data was reported for a shallow rolling bed (4.3% fill), and only this set of data will be used here.
Mass transfer rates were measured for various kiln conditions, assuming an ideally mixed gas phase. The results were expressed in the form of a volumetric mass transfer coefficient \( k_s A/V_{\text{bed}} \), defined as:

\[
QC_{\text{decane,out}} = \frac{k_s A}{V_{\text{bed}}} \left( C_{\text{decane, sat}} - C_{\text{decane, out}} \right) V_{\text{bed}}
\]  

(31)

We can make use of the \( k_s A/V_{\text{bed}} \) data by comparing those with the equivalent data predicted by our model.

\[
QC_{\text{decane,out}} = \int P_{\text{decane}} dV_{\text{bed}} = \eta k (C_{\text{decane, sat}} - C_{\text{decane, out}}) V_{\text{bed}}
\]  

(32)

As mass transfer from the surface of the alumina particles is the rate determining step (as long as the pores are sufficiently filled), the first order reaction rate constant (with respect to the gas phase), follows from

\[
k = \frac{k_{gs} a}{\varepsilon} = \frac{6 k_{gs} (1 - \varepsilon)}{d_p \varepsilon}
\]  

(33)

with \( k_{gs} \) being the mass transfer coefficient at the surface of the alumina particles. We should thus compare the \( k_s A/V_{\text{bed}} \) data of Jauhari et al with the \( 6\eta k_{gs} (1 - \varepsilon) / d_p \) data that we can produce with our model using eq. (15) or (30). Before doing so, we must find an appropriate value for \( k_{gs} \). For that purpose, we use the findings of Sørensen and Stewart (1974) who studied mass transfer in packed beds. For a stagnant fluid they derived:

\[
k_{gs} = \frac{Sh \cdot D}{d_p}
\]  

with \( Sh = 3.8 \)

(34)
As the gas entrained into the bed of a kiln is regarded to have no slip velocity with regard to the particles in the bed, and the bed porosity is comparable to that of a packed bed, eq. (34) is assumed to be appropriate.

If we now apply the precise conditions at which Jauhari et al. have produced their $k_s A / V_{bed}$ data, we are able to calculate the corresponding $6 \eta k_s (1 - \epsilon) / d_p$ data with our model. These conditions are summarised in Table 2:

**Table 2: Conditions used in our model.**

<table>
<thead>
<tr>
<th>Total volume :</th>
<th>20.3 liters</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bed volume:</td>
<td>0.875 liters</td>
</tr>
<tr>
<td>$h/R$ :</td>
<td>0.824</td>
</tr>
<tr>
<td>$R$:</td>
<td>0.145 m</td>
</tr>
<tr>
<td>$h$:</td>
<td>0.1195 m</td>
</tr>
<tr>
<td>Cross-sectional bed area ($A_x$):</td>
<td>0.00284 m²</td>
</tr>
<tr>
<td>Particle diameter ($d_p$):</td>
<td>$3 \times 10^{-3}$ m</td>
</tr>
<tr>
<td>Binary diffusion coefficient of decane in nitrogen at 20°C ($D_{ab}$):</td>
<td>$6 \times 10^{-6}$ m² s⁻¹</td>
</tr>
<tr>
<td>Interparticle voidage ($\epsilon$):</td>
<td>0.45 (assumed)</td>
</tr>
<tr>
<td>Sherwood number ($Sh$):</td>
<td>3.8</td>
</tr>
<tr>
<td>Gas-solid mass transfer coeff. ($k_{gs}$):</td>
<td>$7.6 \times 10^{-3}$ m s⁻¹</td>
</tr>
</tbody>
</table>

\[
k = \left( \frac{6 k_s (1 - \epsilon)}{\epsilon d_p} \right) = 18.6 \text{ s}^{-1}
\]

Below we compare our results with the data measured by Jauhari et al.
Table 3. Comparison of measured data and model predictions.

<table>
<thead>
<tr>
<th>Rotation speed N (rpm)</th>
<th>Angular velocity ( \omega ) (s(^{-1}))</th>
<th>Measured data ( k_s A/V_{bed} ) (s(^{-1}))</th>
<th>( k ) (s(^{-1}))</th>
<th>Total depletion model: ( \eta \varepsilon )k (s(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.29</td>
<td>0.031</td>
<td>0.121</td>
<td>612</td>
<td>0.0164</td>
</tr>
<tr>
<td>0.59</td>
<td>0.062</td>
<td>0.198</td>
<td>301</td>
<td>0.0329</td>
</tr>
<tr>
<td>1.23</td>
<td>0.128</td>
<td>0.411</td>
<td>144</td>
<td>0.0685</td>
</tr>
<tr>
<td>1.67</td>
<td>0.175</td>
<td>0.502</td>
<td>106</td>
<td>0.0933</td>
</tr>
<tr>
<td>1.97</td>
<td>0.206</td>
<td>0.604</td>
<td>90</td>
<td>0.1101</td>
</tr>
</tbody>
</table>

In all cases, \( k/\omega \) is much higher than the value of the criterion given by Eqn. (29) (i.e. 0.09), meaning that saturation is easily reached within the bed, and that the mass transfer contribution by the convective model is practically insensitive to the calculated rate constant \( k \).

Fig. 8. Mass transfer data measured by Jauhari et al is significantly higher than that predicted by our model. —— Present model (total depletion) o Data for 0.043 fill (Jauhari et al. 1998)
5.2 Measured data

In order to test our model at conditions where kinetics are much slower, we performed similar experiments to Jauhari et al (1998), but using larger particles, allowing the evaporation to continue until the particles were dry. In this way, mass transfer within the pores of the particles became the rate limiting process in the final drying stage.

![Flow diagram of experimental apparatus](image)

*Figure 9. Flow diagram of experimental apparatus*

The rotating drum used has a diameter of 0.15 m and a length of just 0.041 m (Fig. 9). It is made of steel with a polycarbonate observation window. It was only operated in the rolling mode at two speeds: 0.052 radians/s and 0.266 radians/s. Air was introduced through a rotameter and blown through a 1 mm jet directly away from the bed towards the circular wall of the rotating drum, in order to ensure good mixing of the gas above the bed and to prevent any bypassing of gas to the central exit. Like Jauhari et al. (1998), hydrocarbon concentration was measured
continuously using a sensor. The sensor was calibrated so that the sensor reading could be transformed to a linear hydrocarbon concentration reading. Because the hydrocarbon concentration and air flow rate were known at all times during a run, it was possible to calculate what fraction of hydrocarbon liquid had evaporated at any time. The fraction of liquid remaining in the particle (liquid loading, \( z \)) has been used as a parameter to characterise the particle at a given set of conditions.

The experimental technique to measure mass transfer rates differs from that used by Jauhari et al. (1998), because the sensor was placed within the rotating drum, near the gas exit. This allowed us to directly measure the build-up of concentration within the freeboard gas after the gas flow had been stopped.

For the experiments reported here, we used spherical tab-alumina particles of average diameter 16 mm ± 1 mm soaked in 1-pentanol. The 44 pellets that were used gave an average fill fraction of 0.27. The particles were wiped dry before being loaded in the rotating drum. During the experiment, air flowed through the rotating drum. At suitable intervals, the gas flow was stopped, and the rate of increase of hydrocarbon concentration was measured until it came close to saturation. The characteristic rate of the system (\( \eta \xi k \)) was calculated from the exponential rise in the hydrocarbon concentration as a function of time:

\[
\eta \xi k (C_{\text{sat}} - C) V_{\text{bed}} = V_f \frac{dC}{dt}
\]  

which can be integrated to give:

\[
- \ln \left( \frac{C_{\text{sat}} - C}{C_{\text{sat}} - C_0} \right) = V_{\text{bed}} \frac{V_f}{\eta \xi k} t
\]  

which can be integrated to give:

\[
- \ln \left( \frac{C_{\text{sat}} - C}{C_{\text{sat}} - C_0} \right) = V_{\text{bed}} \frac{V_f}{\eta \xi k} t
\]  

(36)
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$C_{sat}$ could be accurately estimated by visually noting how the end points of the graph of $-\ln\left(\frac{C_{sat} - C}{C_{sat} - C_0}\right)$ vs $t$ fitted the straight line formed by the other points. An example of such a graph is given in Fig. 10.

![Graph showing mass transfer from bed to freeboard](image)

**Figure 10.** Mass transfer from bed to freeboard was calculated from the rate at which concentrations increased in the freeboard after gas flow was stopped. (Here, liquid loading was 0.57 and $\omega = 0.266$ s$^{-1}$)

Several such measurements were performed during a run (with liquid loading of the particles decreasing continuously), using both fast and slow rotation speeds. The saturation concentrations inferred with this technique did not vary substantially during the run, as can sometimes happen with small pores, or when desorption within the particles becomes rate limiting. The saturation concentrations varied by less than 1% throughout a run.

We used a similar technique to model the characteristic rate of the system ($\eta \varepsilon k$) as with Jauhari’s data, except that we now added a shrinking core
model to the resistance term. The term \( L \) is liquid loading, and \( \varepsilon_{\text{part}} \) is the intraparticle porosity:

\[
k = \frac{6(1-\varepsilon)}{\varepsilon} \frac{D}{d_p^2} \left[ \frac{1}{Sh} + \frac{1}{\varepsilon^2_{\text{part}}} \left( \frac{1}{z^{1/3}} - 1 \right) \right]
\]  

(37)

The effective dispersion coefficient within the particle was estimated as \( \varepsilon_{\text{part}}^2 D \), and the value of \( \varepsilon_{\text{part}} \) (0.15) was inferred from the experimental data taken at the slowest drying rates. The parameters used in the model are given in Table 4.

Table 4. Parameters used for the modelling of our data.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Binary diffusion coefficient, pentanol-air (D)</td>
<td>8.25 x 10^{-6} m²/s</td>
</tr>
<tr>
<td>Bed porosity (( \varepsilon ))</td>
<td>0.636</td>
</tr>
<tr>
<td>Particle porosity (( \varepsilon_{\text{part}} ))</td>
<td>0.15</td>
</tr>
<tr>
<td>Particle diameter (( d_p ))</td>
<td>0.016 m</td>
</tr>
<tr>
<td>Sherwood number (( Sh ))</td>
<td>3.8</td>
</tr>
</tbody>
</table>

The total depletion model was not applicable at these experimental conditions, so \( \eta \) was estimated using Eq. (15). This enabled us to plot our model \( \eta \eta_k \) values with those that we obtained by experiment:
Figure 11. Characteristic rate is well predicted by the model for both fast rotation —×— (0.266 s\(^{-1}\)), and slow rotation —○— (0.052 s\(^{-1}\)).

The predicted mass transfer rates follow the trend of the experimental data quite well. By using the model prediction of \(k\) at the various liquid loadings (Eq. 37), it is possible to calculate the effectiveness factor \(\eta\) and \(k/w\) at each of the experimental data points. These points (Fig. 12) follow the expected trend quite well, but show some scatter. The experimental data for \(0.6 < k/w < 2\) were measured with wet pellets, and the values measured for \(0.02 < k/w < 0.1\) using externally dry pellets are higher than expected, possibly due to uneven drying on the surface of the pellets.
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6. CONCLUSION

A novel model to predict mass transfer inside a rolling rotary kiln was presented. This model only considers convection inside the voids between the particles inside the bed, which are assumed to be filled with entrapped gas from the freeboard.

The model was found to underestimate mass transfer rates for fast reacting systems ($k/\omega > 1$), showing that other mechanisms like dispersion and/or surface phenomena are also important for such systems. Active layer models are to be preferred in these cases. However, for slower reactions ($k/\omega < 1$) the presented model appears to perform much better. It then provides a useful alternative to active layer models, which are clearly inappropriate if the whole bed contributes to mass transfer. For more general applicability, it would be useful to include diffusion/dispersion effects, which will be done in a future paper.
SYMBOLS

a \quad \text{pellet outer surface area per bed volume [m}^{-1}\text{]}
A \quad \text{area [m}^2\text{]}
A_x \quad \text{radial cross-sectional area of the bed [m}^{-2}\text{]}
C \quad \text{concentration [mol m}^{-3}\text{]}
D \quad \text{binary diffusion coefficient [m}^2\text{s}^{-1}\text{]}
d_p \quad \text{particle diameter [m]}
F \quad \text{solid fill fraction: bed volume/kiln volume}
g \quad \text{gravitational acceleration [m s}^{-2}\text{]}
h \quad \text{perpendicular distance from radial center of the kiln to the bed surface [m]}
k \quad \text{reaction rate constant [s}^{-1}\text{]}
k_{gs} \quad \text{mass transfer coefficient on the surface of particles [m s}^{-1}\text{]}
k_s \quad \text{mass transfer coefficient of active layer [m s}^{-1}\text{]}
L \quad \text{length of the rotary kiln in axial direction [m]}
P_A \quad \text{reaction rate of A [moles m}^3\text{(bed) s}^{-1}\text{]}
Q \quad \text{volumetric gas flow rate [m}^3\text{s}^{-1}\text{]}
R \quad \text{radius of rotary kiln [m]}
r \quad \text{distance from radial center of bed to the surface of the bed at angle 2 [m]}
s \quad \text{half of the chord length formed by the bed surface [m]}
V \quad \text{volume [m}^3\text{]}
V_{\text{bed}} \quad \text{volume of bed [m}^3\text{]}
V_f \quad \text{volume of freeboard [m}^3\text{]}
z \quad \text{liquid loading in particles: volume fraction of pores filled by liquid}
Greek symbols

\( \varepsilon \) bed voidage \([\text{m}^3(\text{gas}) \text{m}^{-3}(\text{bed})]\)

\( \eta \) effectiveness factor of reaction

\( \tau \) residence time of gas in the bed \([\text{s}^{-1}]\)

\( \theta \) angle \([\text{rad}]\)

\( \phi \) gas flow rate \([\text{m}^3(\text{gas}) \text{s}^{-1}]\)

\( \omega \) angular velocity \([\text{s}^{-1}]\)

REFERENCES


Chapter 3 - Diffusion effects in rotating rotary kilns

ABSTRACT

A novel approach to the modeling of mass transfer in rotary kilns has been described (Heydenrych et al, 2001). It considers the mass transfer to occur by the inclusion of gas in the interparticle voids in between the particles that move concentrically with the kiln. By doing so, the rate of mass transfer was found to be dependent on bed fill and the ratio of reaction rate constant to angular velocity \((k/\omega)\). The model was found to be valid at slow to medium fast reactions. For fast reactions it under-predicted mass transfer. Therefore in this paper, the model will be extended to include diffusion effects. An additional dimensionless number is necessary then to describe the system. This can either be a Peclet number \((\omega R^2/D_e)\) or a Thiele modulus \((kR^2/D_e)^{1/2}\). The solution of the 2-dimensional partial differential equations that describe the extended model gives a handle on the effect of scale-up in rotary kilns. For industrial-scale kilns, the Peclet number is large, which means that diffusion within the lower (passive) layer of the bed is unimportant for slower rates. With high reaction rates, iso-concentration lines are closely stacked near the surface of the bed, implying that it is important to model the active layer rather than the bed as a whole in these circumstances. However, the stiff differential equations are not easily solved then, and other methods of solution are advisable.
1. **INTRODUCTION**

Traditionally, mass transfer in rotary kilns has been modelled as a mass transfer coefficient at the surface of the bed of a rotary kiln (Jauhari *et al.* 1998). Recently, a novel approach to the modelling of mass transfer has been suggested though (Heydenrych *et al.*, 2001). This approach takes into account the inclusion of gas in the interparticle voids in between the concentrically moving particles in the passive layer of the bed. In that work, the role of the active layer where particles move along the surface of the bed was ignored by assuming that the thickness of the active layer region is infinitely thin. They concluded that their concentric movement model has merit, but that the effect of diffusion and dispersion within the bed must also be taken into account in the case of fast reactions. In this paper we will explore the effect of diffusion within the concentric flow region, and how it affects the overall mass transfer, again under the assumption of an infinitely thin active layer.

*Figure 1. Shows the concentric movement and active layer regions*
2. MATHEMATICAL MODEL

To take diffusion into account, consider the mass balance for a differential element within the bed (i.e. concentric flow region):

\[
\frac{\partial C_A}{\partial t} = -\nabla \cdot \mathbf{N}_A + r_A
\]  

(1)

After substituting equations for the transport of gaseous A (Fick’s diffusion plus convection) into the above equation, the concentration profile of A follows from

\[
\frac{\partial C_A}{\partial t} = \nabla \cdot (D_e \nabla C_A - c_A \varepsilon \mathbf{V}) + r_A
\]  

(2)

\[
\frac{\partial C_A}{\partial t} + \varepsilon \mathbf{V} \cdot \nabla C_A = D_e \nabla^2 C_A + r_A
\]  

(3)

Sharp concentration gradients are expected in the radial cross-section of kilns [Heydenrych et al, 2000]. In the axial direction, relatively low concentration gradients can be expected due to the high length to diameter ratio found in typical rotary kilns. This makes it a reasonable assumption to simplify the governing equation to the two dimensions that represent the radial cross-section. Although polar coordinates may be a more natural choice of coordinate system, we have chosen rectangular coordinates because of software restrictions. Assuming steady-state operation:

\[
\varepsilon V_x \frac{\partial C_A}{\partial x} + \varepsilon V_y \frac{\partial C_A}{\partial y} + D_e \frac{\partial^2 C_A}{\partial x^2} + D_e \frac{\partial^2 C_A}{\partial y^2} + r_A = 0
\]  

(4)
Here, $D_e$ is the effective diffusivity and $e$ is interparticle voidage. Both are assumed to be constant. $V_x$ and $V_y$ are the x and y components respectively of the velocity of particles (and gas) in the bed, and are described by the concentric motion of the bed. Zero flux at the solid walls gives a simple boundary condition: $\bar{n} \cdot \nabla C_\lambda = 0$ where $\bar{n}$ denotes the normal vector at the kiln wall. For the bed surface, various boundary conditions can be used as appropriate, discussed later in the paper.

Rewriting Eqn. 4 in terms of dimensionless variables, for an $n^{th}$ order reaction with $-r_A = \varepsilon k C_A^n$:

$$\frac{1}{Pe} \frac{\partial^2 \Psi}{\partial \lambda_x^2} + \frac{1}{Pe} \frac{\partial^2 \Psi}{\partial \lambda_y^2} + \nu_x \frac{\partial \Psi}{\partial \lambda_x} + \nu_y \frac{\partial \Psi}{\partial \lambda_y} + \frac{\varepsilon k C_A^{n-1}}{\omega} \Psi^n = 0 \quad (5)$$

where

$$\Psi = \frac{C_A}{C_{A,s}} \text{ dimensionless concentration,}$$

$$\lambda_x = \frac{x}{R}, \lambda_y = \frac{y}{R}, \nu_x = \frac{V_x}{\omega R}, \nu_y = \frac{V_y}{\omega R},$$

$$Pe = \frac{\varepsilon \omega R^2}{D_e}, \text{ a Peclet number modified for rotary kilns.}$$

The reaction term $\frac{k C_A^{n-1}}{\omega}$ is similar to the $k/\omega$ term used by Heydenrych et. al. (2001) to describe concentration terms within the bed of a rotary kiln in which a first order reaction takes place.

Noting that $\nu_x$ is simply the x-component of the velocity divided by the velocity at the wall, it follows from the concentric movement geometry that $\nu_x = \lambda_x$ and $\nu_y = -\lambda_y$ using the coordinate system given in Fig. 2. See the Appendix for more detail.
Figure 2: Applied coordinate system. We assumed that the active layer was infinitely thin.

In addition to the no-flux condition at the wall, the Danckwerts boundary condition for non-ideal flow in a plug flow reactor was used as the boundary condition at the bed surface:

\[ C_{A,x} = C_A - \frac{D_e}{\varepsilon V_x} \left( \frac{\partial C_A}{\partial x} + \frac{\partial C_A}{\partial y} \right) \]  \hspace{1cm} (6)

The condition may also be written in dimensionless form as

\[ \frac{\partial \psi}{\partial \lambda_x} + \frac{\partial \psi}{\partial \lambda_y} - Pe \nu_x \psi = -Pe \nu_x \]  \hspace{1cm} (7)

The no-flux condition at the wall is given in dimensionless form by

\[ n_x \frac{\partial \psi}{\partial \lambda_x} \bigg|_{\lambda=1} + n_y \frac{\partial \psi}{\partial \lambda_y} = 0 \]  \hspace{1cm} (8)
where \( n_x \) and \( n_y \) are the components normal to the wall confining the particulate solids.

The model described here (Eqns. 5, 7 and 8) is dependent on two dimensionless variables: \( Pe \) and \( \frac{kC_{A,s}^{n-1}}{\omega} \). Note that the square of the Thiele modulus \( \phi^2 = Pe \cdot \frac{kC_{A,s}^{n-1}}{\omega} \). This means that the system can be described by any two of the dimensionless groups \( Pe \), \( \phi \) or \( \frac{kC_{A,s}^{n-1}}{\omega} \). In this paper we have chosen to use \( \frac{kC_{A,s}^{n-1}}{\omega} \) because it is the only variable that is required to describe the no-diffusion case, and \( Pe \). The rate constant \( k \) typically changes significantly along the length of the kiln, as the solid phase approaches full conversion (or total drying). \( Pe \) only needs to be calculated once for a given kiln because it does not typically change along the length of a rotary kiln.

Fraction fill is the third independent dimensionless variable that is necessary to describe this model. It is also interesting to note that the fraction fill does not affect the governing equations (Eqn. 5), but only the geometry of the boundary conditions.

**Solution of the model**

This model was solved using Matlab’s partial differential equation toolbox. Although it would be possible to model \( n^{th} \) order reactions using the same toolbox, in this case, we have restricted our analysis to \( 1^{st} \) order reactions. For future reference, we will refer to the second dimensionless variable in its form for first-order reaction, \( k/\omega \).
The solution gives the concentration profile within the bed of the kiln, and
the effectiveness factor of the bed is calculated from that concentration
profile. The effectiveness factor of a rotary kiln is defined analogously to
the effectiveness factor of a catalyst pellet.

\[
\eta = \frac{- \int P_A dV}{- \int P_{A,s} dV} = \frac{\int \psi dA}{\int \phi dA} = \frac{\int C^n dA}{\int C^n dA_{bed}} = \frac{\int \Psi^n dA}{\int \Psi^n dA_{bed}} \quad (6)
\]

where \(-P_A\) is the local reaction rate in the bed and \(-P_{A,s}\) is the reaction rate
that would occur if the entire cross section of the bed was exposed to the
conditions at the surface of the bed.

For 1\textsuperscript{st} order reactions, \(\eta = \frac{\int \psi dA}{\int \phi dA_{bed}}\),

where area A is calculated in terms of \(\lambda_x\) and \(\lambda_y\). \quad (7)

Given that the solution of the partial differential equation (Eq. 5) is the
concentration profile over the bed, the effectiveness factor can be
determined from the average dimensionless concentration over the bed
cross section.

\section{3. RESULTS}

The effectiveness factor curves for a 50\%, 20\% and 10\% filled kiln that
were generated using the model are shown in figures 3 to 6. The curve for
an infinite Peclet number represents the no-diffusion case reported earlier
by Heydenrych \textit{et. al.} (2001).
As expected, the inclusion of diffusion effects in the model increases the effectiveness factor in all conditions. It should also be noted that for a given Peclet number (i.e. kiln size and rotation speed), the effect of diffusion as a fraction of the contribution by gas inclusion, is far higher at high values of $k/\omega$, where high concentration gradients occur in the bed (predominantly near the surface of the bed). For commercial scale rotary kilns, Peclet numbers are typically greater than 1000, discussed later in the paper.

At higher Peclet numbers ($Pe > 10$), the numerical method was unable to converge on a solution. The dashed lines on Figure 3 are extrapolations, where the model failed to solve. A possible reason for the poor convergence of the model at large Peclet numbers, is that the numerical methods used to solve the model were based on techniques suited for an elliptical (second-order) partial differential equation, with the first-order term simply being handled as a source term. At high Peclet numbers, the second-order terms of the partial differential equation become less important, and the numerical method is unsuited to the solution of what effectively becomes a first-order hyperbolic pde that dominates the model.
Similar solutions have been found for kilns with bed fills of 20% and 10% (Figs. 4 and 5 respectively).

**Figure 4.** Effectiveness factors as a function of $k/\omega$ for a 20% filled kiln; 1st order reaction.

![Figure 4](image1.png)

**Figure 5.** Effectiveness factors as a function of $k/\omega$ for a 10% filled kiln; 1st order reaction.

![Figure 5](image2.png)

As the fill fraction decreases, the effect of diffusion appears to become more important. The concentration profiles generated at certain Pe and $k/\omega$ values are interesting, and are shown in Fig. 6 for a half-filled kiln:
Figure 6: Concentration profiles at $Pe = 1$; 1st order reaction.

The straight iso-concentration lines predicted for the no-diffusion case (Heydenrych et al., 2001), Fig. 7, are only readily evident for the diagram...
where $k/\omega = 0.1$. For the other cases, diffusion effects dominate the shape of the concentration profile, and the rotation of the kiln does not distort the profile significantly.

Figure 7. If diffusion is not taken into account, straight iso-concentration lines are expected.

4. DISCUSSION

Scale-up effects

The models have shown that for Peclet numbers less than 100, diffusion effects are important. But under what circumstances can we expect such low Peclet numbers, and what Peclet numbers are typical for rotary kilns?

The diffusivities of a gas at atmospheric pressure vary between $10^{-4} - 10^{-6}$ m$^2$/s (Knudsen et. al., 1997). Values of $2 \times 10^{-4}$ m$^2$/s, $2 \times 10^{-5}$ m$^2$/s and $2 \times 10^{-6}$ m$^2$/s were used to for the diffusivity of the reactant gas $D_A$.

The effective diffusivity in the bed of the kiln can be estimated as follows:

$$D_e = \frac{D_f E}{\tau} \quad \text{(Knudsen et. al., 1997)}$$
Using a tortuosity (τ) of 2 and voidage (ε) of 0.5, we can expect effective diffusivities within the bed of $5 \times 10^{-5} \, \text{m}^2/\text{s}$ (max.), $5 \times 10^{-6} \, \text{m}^2/\text{s}$ and $5 \times 10^{-7} \, \text{m}^2/\text{s}$ (min.).

Kiln rotation speeds vary less, because at lower rotation speeds, slumping of the bed can occur. At higher rotation speeds, centrifugation begins. For this reason, only two angular velocities are considered, 0.5 rad/s and 0.1 rad/s.

The effect of scale is largely dependent on the radius, because $Pe \propto R^2$. Table 1 shows Peclet numbers for selected $D_e$ and $\omega$ values as a function of radius.

<table>
<thead>
<tr>
<th>$D_e$, m²/s</th>
<th>$\omega$, rad/s</th>
<th>$5 \times 10^{-5}$</th>
<th>$5 \times 10^{-6}$</th>
<th>$5 \times 10^{-7}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>0.1</td>
<td>0.5</td>
<td>0.1</td>
</tr>
<tr>
<td>0.05</td>
<td>0.1</td>
<td>5</td>
<td>25</td>
<td>50</td>
</tr>
<tr>
<td>0.1</td>
<td>0.1</td>
<td>20</td>
<td>100</td>
<td>200</td>
</tr>
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<td>0.5</td>
<td>0.5</td>
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<td>2500</td>
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<tr>
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<td>1</td>
<td>2000</td>
<td>10000</td>
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<tr>
<td>5</td>
<td>5</td>
<td>50000</td>
<td>250000</td>
<td>500000</td>
</tr>
</tbody>
</table>

As an example, the Peclet numbers for the rotating drum used by Jauhari et al. (1998), with a rotating drum of radius 0.145 m, varies from 530 to 3600, depending on rotation speed. It is obvious that diffusion as a possible extra transport mechanism to gas entrapment can safely be neglected for practical applications as well as for most lab experiments.
5. CONCLUSIONS

A mathematical model has been presented that improves on the previous model (Heydenrych et al., 2001) by adding diffusion effects to gas entrapment in the concentric flow region of a rotary kiln. The technique, but not the model, is limited to moderate Pe numbers. For practical values of Pe, a modified solution technique is required. Heydenrych et al., 2001 have shown that $k/\omega$ is the parameter that describes mass transfer at low to moderate reaction rates. In this paper, Peclet number was implemented (in addition to $k/\omega$) to model diffusion effects. This number is defined as $\omega R^2/D_e$. The Peclet number provides a tool for predicting scale-up effects, and even though the model presented here is oversimplified due to the assumption of an infinitely thin active layer, it nevertheless provides an insight to the use of the Peclet number for the scale-up of rotary kilns.

Diffusion in the concentric flow region was found not to add much to the mass transfer at realistic conditions where the Peclet number is much higher than 1. At high $k/\omega$ values, however, concentration gradients become fully concentrated near the surface of the bed, and here the assumption that the active layer is infinitely thin becomes a poor assumption as all mass transfer is occurring within this layer. This model shows that the entrapment of gas perpendicular to the bed is relatively unimportant compared to diffusion effects at these higher $k/\omega$ values.

Further work should be concentrated at developing a suitable model for the active layer, where a 1-dimensional dispersion model would be a fair assumption, based on the results of this work. It will also be important to determine the depth of the active layer, and how the movement of particles over each other affects the dispersion of the gas in the direction perpendicular to the bed surface. This information will give the boundary
conditions for the concentric flow region, with the aim of generating a comprehensive model including all mechanisms for mass transfer.

SYMBOLS

A  Cross sectional area of the bed of the rotary kiln [m$^2$]
$C_A$  Concentration of reactant A in the bed of the rotary kiln [kmol/m$^3$]
$C_{A,s}$  Concentration of reactant A at the surface of the bed [kmol/m$^3$]
$D_e$  Effective diffusivity of reactant A [m$^3$/s]
$Pe$  Peclet number modified to suit a rotary kiln
$R$  Radius of kiln [m]
r  Reaction rate of a reactant A [kmol/m$^3$/s]
$V$  Velocity of gas within the bed [m/s]
x  Co-ordinate in x direction
y  Co-ordinate in y direction

Greek Letters

$\varepsilon$  Voidage of the bed of the rotary kiln
$\phi$  Thiele modulus
$\eta$  Effectiveness factor
$\lambda$  Dimensionless distance
$\tau$  Tortuosity
$\upsilon$  Dimensionless velocity
$\omega$  Angular velocity of kiln [rad/s]
$\psi$  Dimensionless concentration
REFERENCES


APPENDIX

Proof that $u_x = \lambda_y$ and $u_y = -\lambda_x$

Consider a point on the radius, which is at $\lambda = 1$ in dimensionless coordinates. By definition,

$$v_x = \frac{V_x}{\omega R}$$

Substituting for $V_x$,

$$v_x = \frac{\omega R \sin \theta}{\omega R} = \sin \theta$$

From the diagram,

$$\lambda_y = \sin \theta = v_x$$

At points away from the wall, $u_x$ and $\lambda_y$ decrease proportionately. It can similarly be shown that $v_y = -\lambda_x$. 
Chapter 4 - Hydrodynamics and dispersion in the active layer of rolling rotary kilns

ABSTRACT

In previous works (Heydenrych et. al. 2001a,b), it was shown that the top layer (or active layer) of a rolling bed is the only active place in the case of a fast reaction. The bottom part of the bed (also referred to as the concentric flow region) is almost depleted and therefore hardly reacting. Therefore in this work we focus on that active layer. A hydrodynamic model is prepared to predict its shape. A mass balance on particles moving within a rotary kiln gives the shape of the active layer, defined as that part of the bed where particles move past each other. The derivation is based on the assumption that there is a linear velocity gradient of particles in the direction perpendicular to the surface of the bed that is constant at any place on the bed surface. Published data was used to establish a relationship between this velocity gradient on the one hand, and bed fill and rotation speed on the other. A dispersion model was derived to predict the mass transfer of gaseous reactant within the active layer. As void spaces are created and destroyed due to the action of particles moving over each other, mixing of gas occurs. A dispersion coefficient was derived, based on this gas mixing, and was found to be proportional to the velocity gradient and to the square of particle diameter. Applying these relationships to a 1-dimensional concentration gradient model gave a correlation for the mass transfer coefficient at the bed surface. This model does not correctly predict the proportionality of mass transfer with bed speed reported in the literature. We conclude that another effect, probably the granular temperature of the particles, is important for modeling of dispersion and mass transfer in the active layer of rolling rotary kilns.
1. INTRODUCTION

The movement of particles in a rotary kiln is shown in Figure 1. Particles move concentrically with the wall of the kiln (passive layer) until they reach a point near the surface, at which point the particles slide down the top surface of the bed. This differentiation of the two zones within a rotary kiln has been recognized from the earliest published data on rotary kilns (Sullivan et al. 1927, Saeman, 1951).

![Diagram of rotary kiln](image)

Fig. 1. Particles slide over each other to the lower half of the bed in the active layer.

This motion of particles relative to each other leads to much improved heat and mass transfer rates compared to what one would expect from a stationary bed (Heinen et al. 1983).

Traditionally, heat and mass transfer rates have been modeled using a coefficient based on the surface area of the bed (Tscheng & Watkinson, 1979). Recently, Heydenrych et al. (2001) have shown that in certain
cases, particularly for slow processes, the active layer, and the volume of the bed can also become important. Those models were developed for mass transfer, and in this paper we will continue to model mass transfer as an example of transfer processes that occur in rotary kilns.

In this paper, we explore the physical processes that occur within the active layer in order to model the depth and shape of the active layer, and to predict the dispersion that can be expected. With these models, we estimate concentration profiles and finally calculate the effective mass transfer coefficient (based on bed surface area) for the active layer for a given rotation speed, particle size and bed depth.

2. HYDRODYNAMICS OF THE ACTIVE LAYER

2.1 The shape of the active layer

The movement of particles sliding over each other in the active layer causes dispersion of the gas in the voids. This dispersion effect is typically much greater than the effect of gaseous diffusion in the active layer, and concentration gradients in this layer are much less than one would predict by the diffusion model presented by Heydenrych et. al. (2001).

The most important parameter for predicting the degree of dispersion will be the gradient of the velocity profile near the surface of the bed.

If we take any line from a point on the bed surface to a point on the wall within the bed, the mass on either side of that line must remain constant in
time. We will use a line extending from 1 to 2 to 3 on Fig. 1; we expect the mass within the volume 1, 2, 3, 4 to remain constant.

\[ \frac{\partial}{\partial x} \left( \frac{d^2}{2} \right) = \text{const.} \]

Figure 1. Mass balance over envelope described by 1-2-3-4

The volume flow rate out of the control volume (using solids in this case) has earlier been shown (Heydenrych et al., 2001) to be:

\[ Q = \frac{\alpha L (1 - \varepsilon)}{2} \left( R^2 - x^2 - (h + d)^2 \right) \]  \hspace{1cm} (1)

Boateng & Barr (1997) have measured the velocity profiles in the active layers of rotary kilns, and found that for certain materials, the gradient of the velocity profile is substantially linear in the direction perpendicular to the bed surface (1-2) on Fig. (1). They also found that this gradient does not change along the x-direction in Fig. 1. We will make this assumption to calculate the volume flow rate into the control volume through the envelope extending from 1 to 2 in Fig. 1. It would be useful to understand through granular dynamics simulations (Hoomans, 2000) how the choice of materials affects these assumptions.
Firstly, let us define the gradient of the velocity profile (called shear rate by Boateng and Barr (1997)) as the variable \( m \). Also, defining the velocity of particles at the surface and at depth \( d \) as \( v_s \) and \( v_d \) respectively, it follows that

\[
v_s - v_d = dm
\]  

(2)

and assuming that \( v_d \) is determined by the x-component velocity of the passive layer,

\[
v_d = -r \omega \sin \theta = -(h + d) \omega
\]  

(3)

Substituting for \( v_d \) into Eq. (2),

\[
v_s = dm - (h + d) \omega
\]  

(4)

The volume flow rate of solids between points 1 and 2 can now be calculated, using the average velocity between these two points:

\[
Q = \frac{(1 - \varepsilon)Ld(v_s + v_d)}{2}
\]  

(5)

Eliminating \( Q \) using Eqns. (1) and (5), we get:

\[
v_s + v_d = \frac{\omega}{d} \left( R^2 - x^2 - (h + d)^2 \right)
\]  

(6)
Chapter 4 – Active layer models

Substituting Eq. (3) for $u_d$ and Eq. (4) for $u_S$ and simplifying,

$$d = \sqrt{\frac{R^2 - x^2 - h^2}{m/\omega - 1}}$$  \hspace{1cm} (7)

Note that with $d$ and $m/\omega$ known, $u_S$ can be determined as a function of $x$ from Eq. (4).

The $m/\omega$ term is interesting. It means that $m$ has to be greater than $\omega$ to have any physical significance. If we choose a linear relationship for $m = b\omega$, ($b$ is a constant), it means that $b$ must be greater than 1. Literature data suggests $b$ is about 15. Also, if (as the data suggests), $m = b\omega + c$, then the only requirement is that $c > \omega$, or $b > 1$. In reality, this constraint is easily met.

A constraint that is not as easily met is the constraint that the active layer depth $d$ must be less than the bed depth $H$, where $H = R - h$. This constraint can be represented by

$$d(x=0) < R - h$$  \hspace{1cm} (8)

Substituting Eqn. (6) for maximum active layer thickness at $x = 0$, we can show that this constraint simplifies to

$$\frac{m}{\omega} > \frac{2R}{H}$$  \hspace{1cm} (9)

The relationship between $m$ and $\omega$ has been measured by Boateng & Barr (1997). They measured particle velocities at various depths using an optical probe. Their measurements indicate that for a given material and
bed depth, m increases linearly with $\omega$. Their data for beds consisting of polyethylene beads and limestone are shown in Fig. 2. The data shows some scatter, and simulation using granular dynamics (Hoomans, 2000) might shed some light on where the true relationships lie. We will look for a relationship describing m not just as a function of rotation speed $\omega$, but also as a function of bed fill, based on the experimental data.

![Image of graph showing linear relationship](image)

**Figure 2.** The linear relationship $m = 19\omega + 7.5$ is shown by the solid line. Data points marked PE represent polyethylene beads, and LS represents limestone.

Noting that the constraint for the value of m imposed by Eqn. (9) also follows a linear relationship with $\omega$, it would be interesting to see if m also follows a linear relationship against R/H. We have plotted the data of Boateng & Barr (1997) against R/H in Fig. 3.
Figure 3. Shear stress $m$ shown as a function of $R/H$ for polyethylene particles. 

The data appears to correlate reasonably well with $R/H$, indicating that the shear stress is indeed dependent on $R/H$. In fact for shallower beds (high $R/H$), there appears to be a linear relationship, and the slope of the lines correlates well with the slope of $2\omega$ suggested by the criterion of Eqn. (9). In fact, a suggested relationship for small bed depths

$$m = 2\omega \frac{R}{H} + c$$

is shown on Fig. 3, with $c = 9.5 \text{ s}^{-1}$. This model fits the data for $R/H > 3$ very well. The intercept is apparently independent of rotation speed, and represents the shear stress that will result from particles sliding down the surface of an inclined chute with an infinitely deep bed, and will depend on particle characteristics only. In this case, one can expect a layer of finite thickness, and constant $m$ as flow increases – i.e. bed depth proportional to flow rate of particles.
Noting from the data of Boateng & Barr (1997) that a typical value for shear stress $\mu/W$ is 10, we conclude that the constraint (Eqn. 9) will become important when $R/H < 5$. In terms of bed fill, it means that for bed fill less than 5.2%, $m$ must be larger than the narrow range expected from the data shown in Fig. 2.

A further implication of the bed height expression, Eqn. (7), is that $d \propto \sqrt{\omega / (m - \omega)}$. Substituting for $m$ using Eqn. (10),

$$d \propto \sqrt{\omega / \left(\sqrt{2R/H - 1}\omega + 9.5\right)}.$$  This means that at low rotation speeds, $d \propto \omega^{0.5}$. As rotation speed increases, $d$ becomes less dependent on rotation speed.

Our model does not predict the shear stress well at lower $R/H$ values (fuller beds). In this case, geometric constraints and perhaps centrifugal forces may affect the shear stress profile.

Substituting Eqn. (10) into Eqn. (7), we can determine the shape of the active layer at any point in the active layer, at any given kiln rotation speed. The active layer profile predicted by this model for various bed depths and rotation speeds is shown in Fig. 4.
Heinen et al. (1983) have published a good deal of data showing observed active layer thickness as a function of bed depth and rotation speed. Fig. 5 shows how the measured data at the deepest part of the active layer compares with our model for predicting bed depth (at x = 0).
Figure 5. Our model predicts a deeper active layer than visually measured data at $x = 0$, probably due to wall effects that were confirmed by Boateng & Barr (1997).

Some of the scatter in the data can be attributed to the fact that Heinen et al. (1983) reported rotation speeds within limits, rather than the exact values. We used the average values within those limits.

2.2 The volume of the active layer

In some cases, it is useful to know the volume of the active layer, and Jauhari et al. (1998) presented a model for predicting mass transfer rates based on the volume of the active layer. To calculate the volume of the active layer, it is useful to show that, using our model, the shape of the active layer is described by an ellipse with the central axis located on the bed surface.
An ellipse (Fig. 6) is described by

\[
\frac{(y-c)^2}{b^2} + \frac{x^2}{a^2} = 1
\]  

\(\text{(11)}\)

We would like to rewrite the expression for active layer depth in the form shown in Eqn. (11).

\[
\left(\frac{m}{\omega} - 1\right)d^2 = R^2 - x^2 - h^2
\]  

\(\text{(12)}\)

Substitute for \(d^2\) using the relationship \(y = -d - h\)

\[
\frac{(y + h)^2}{m/\omega - 1} + x^2 = R^2 - h^2
\]  

\(\text{(13)}\)

\[
\frac{(y + h)^2}{(m/\omega - 1)(R^2 - h^2)} + \frac{x^2}{R^2 - h^2} = 1
\]  

\(\text{(14)}\)
Writing in terms of the dimensions of Fig (6),

\[ a = \sqrt{R^2 - h^2} \]

\[ b = \frac{\sqrt{R^2 - h^2}}{\sqrt{m/\omega - 1}} \]

\[ c = -h \]

Because the axis of the ellipse lies along the surface of the bed, the area of the active layer is half of the area of the ellipse. Therefore the volume of the active layer is

\[ V_{\text{act}} = \frac{\pi}{2} ab \cdot L = \frac{\pi L (R^2 - h^2)}{2 \sqrt{m/\omega - 1}} \] (15)

Consider the points on the ellipse where \( y = h \). The slope of the tangent = \( \infty \) for all \( h \). Because the slope of the wall is finite for \( h < 0 \), it implies that there is always a point where the active layer theoretically extends beyond the bed (for \( h < 0 \), all \( m/\omega \)). For practical purposes, this is usually insignificant, and should not detract from the usefulness of the model.

### 3. DISPERSSION WITHIN THE ACTIVE LAYER

As particles slide over each other, the size of inter-particle voids changes. This gas moves to adjoining voids, where it mixes with gas at another concentration, assuming that a concentration gradient exists. Consider Figure 7, where successive layers of particles are separated by a distance
approximately equal to the particle diameter $d_p$. In a full cycle, the top layer of particles moves horizontally by a distance $2d_p$ relative to the bottom layer. Noting that the top and bottom layers are separated by a vertical distance of $2d_p$, the time it takes for this cycle will be $1/m$ seconds (where $m$ is the velocity gradient, in m/s per m active layer depth).

Figure 7. Description of voids in one cycle of particles sliding over each other.

Assume also that a steady-state concentration gradient exists with molar flux $W_A$, where we need to determine dispersion coefficient $D$,

$$W_A = -D \frac{dC}{dy} \quad (16)$$

Noting that at steady state the concentrations remain constant at a given $x$, a nett mole balance from position 1 to position 2 over a full cycle gives:

$$N = C_1 f(V_1 - V_2) - C_2 f(V_1 - V_2) \quad (17)$$

The parameter $f$ is used to take into account that the gas can move in any of 6 orthogonal directions, and therefore will have a minimum value of $1/6$. If the particles slide over each other as rigid layers, then the movement of
the gas will primarily be in the y-direction (perpendicular to the bed surface), and \( f \) could have a value as large as \( \frac{1}{2} \). We will use \( f = \frac{1}{6} \) as our default value.

Substitute \( \alpha = \frac{V_2}{V_1} \) – this will later be estimated from the change of voidage from a loosely packed solid to a tightly packed solid.

\[
N = V_1 f (1 - \alpha)(C_1 - C_2) \tag{18}
\]

The time required for one cycle is \( \frac{1}{m} \). It follows then that the molar flow rate \( F_A = Nm \), and from the definition of molar flux \( W_A \), \( W_A = \frac{F_A}{A} \).

\[
W_A = mV_1 f (1 - \alpha)(C_1 - C_2) \tag{19}
\]

\[
\frac{V_1}{A} = \frac{\varepsilon_1 d_p A}{A} = \varepsilon_1 d_p \quad (\varepsilon_1 \text{ is voidage for loose-packed solid.}) \tag{20}
\]

\[
W_A = d_p m \varepsilon_1 f (1 - \alpha)(C_1 - C_2) \tag{21}
\]

\[
\frac{dC}{dy} = \frac{C_1 - C_2}{d_p} \tag{22}
\]

\( D \) is determined by dividing Eqn. (21) by Eqn. (22):

\[
D = d_p^2 m \varepsilon_1 (1 - \alpha) \tag{23}
\]

\( \alpha \) can be determined from the ratio of the voidage of loosely packed spheres vs tightly packed spheres.
\[ \varepsilon_1 = \frac{V_1}{V_v + V_1} \quad \Rightarrow \quad \frac{V_v}{V_1} = \frac{1}{\varepsilon_1} - 1 \]  

(24)

\[ \frac{V_2}{V_1} = \alpha = \frac{1}{\varepsilon_1} - 1 = \left( \frac{1 - \varepsilon_1}{1 - \varepsilon_2} \right) \frac{\varepsilon_2}{\varepsilon_1} \]  

(25)

For loose-packed spheres, \( 1 - \varepsilon_1 = \pi/6 \)

\[ \varepsilon_1 = 0.4764 \]

For tight-packed spheres,

\[ \varepsilon_2 = 0.32 \]

\[ D = 0.246 \text{ fm} d_p^2 \]  

(26)

The effect of binary diffusion must be adjusted to take account of the reduced area for diffusion due to the solid particles, and the tortuosity of the path. This diffusion contribution \( D_c \) is commonly estimated by \( D_c = \varepsilon^2 D_a \). The overall dispersion coefficient for the active layer \( D_{al} \) will be the sum of \( D \) and \( D_c \):

\[ D_{al} = 0.246 \text{ fm} d_p^2 + \varepsilon^2 D_a \]  

(27)

4. CONCENTRATION PROFILES AND MASS TRANSFER

Having established the dispersion coefficient applicable to the active layer, we now need to establish the concentration profile that exists within the active layer in order to predict mass transfer. For the purposes of this paper, we will assume that the gas is fully depleted at the boundary of the
active and passive layers. If the thickness of the layer containing undepleted gas is much smaller than the width of the bed surface, then wall effects can be neglected, and we have the situation of gas dispersion and reaction in a flat plate. This analysis has already been done by Thiele (1937) for diffusion in a catalyst.

Thiele (1937) showed that the concentration profile in a flat plate can be determined (using our notation) as

$$\Psi = \frac{e^{z\phi} + e^{-z\phi}}{e^\phi + e^{-\phi}}, \text{ where } \phi = d \frac{e^{-k}}{D_{ad}}$$

and the effectiveness factor is then

$$\eta = \frac{\tanh(\phi)}{\phi}$$

The above equations (28) and (29) are valid for $d$ being measured from the catalyst surface to the center of the plate, where there is no flux at the center due to symmetry. In our case, we have assumed more stringent conditions, needing to assume that there is no flux at the bottom of the active layer due to depletion of the gas (we do not have symmetry). In this case, Eq. (29) reduces to

$$\eta = 1/\phi$$

For this model, with reaction only occurring at the surface of the bed, we will find it convenient to express the reaction rate for the system in terms of a mass transfer coefficient at the bed surface $k_s$:
\[
\int P_A dV = k_s AC_{A,s} = \eta \varepsilon k C_{A,s} \cdot V_{al} \tag{31}
\]

Neglecting end effects, \( d = V_{al}/A \). Then we can rewrite Eqn. (31) in terms of \( k_s \), and substitute Eqn. (29) to get

\[
k_s = \eta \varepsilon k d = \sqrt{D_{al} \varepsilon k} \tag{32}
\]

The rate constant \( \varepsilon k \) can be calculated (Heydenrych et al. 2001) using

\[
\varepsilon k = \frac{6(1-\varepsilon)Sh \cdot D_a}{d_p^2}, \quad Sh = 3.8 \tag{33}
\]

5. MODEL VALIDATION

Jauhari et al. (1998) reported their mass transfer coefficients as \( k_s A \), enabling us to make a direct comparison of our model with their results, by multiplying Eqn. (33) by bed surface area \( A \). We note that the results of Jauhari et al. (1998), measured at a bed fill of 4.3\%, are substantially proportional to rotation speed (Fig. 8), whereas our model predicts a much smaller dependency on rotation speed, although the predicted values are in the correct range.
Chapter 4 – Active layer models

Figure 8. Our model does not correctly predict the linearity of $k_s A$ with rotation speed, but predicts values in the correct order of magnitude. $f = 1/6$, $d_p = 3$ mm.

The very small dependency of $m$ on rotation speed (using Eqn. (10) to estimate $m$, with $c = 9.5$ s$^{-1}$), results in a similarly low dependency of $D_{al}$ on rotation speed. Using Eqn. (33) we calculated $\xi k = 8.36$ s$^{-1}$.

Clearly, this model does not accurately predict mass transfer rates for the conditions used by Jauhari et al. (1998). There may be two reasons for this. The first is that mass transfer at the top surface of the bed, where the freeboard gas slip velocity becomes important, has not been taken into account in this model. In their measurements, Jauhari et al. (1998) operated under the constraint that the superficial axial gas velocity was less than the drum wall velocity. This could lead to particularly low slip velocities on the outer surface of the bed at the slower rotation speeds. This could lead to the lower than expected mass transfer rates that were measured at rotation speeds lower than 1 rpm.

The second possible reason for deviation from our model could be due to the nature of the movement of particles in the active layer. Our model
assumes that the particles slide over each other, creating and destroying voids in an orderly fashion. Boateng and Barr (1997) have measured the granular temperature of particles in the active layer: a term that describes the random movement of particles due to collisions within the active layer. The frequency of these collisions may be much higher than the frequency predicted by our model. Moreover, the granular temperature measured by Boateng and Barr (1997) is dependent on depth \((d)\) within the active layer, which means that \(D_{al}\) changes with bed depth, giving a different dependency of \(k_sA\) on rotation speed. It may also affect the mass transfer resistance at the surface of the bed.

6. DISCUSSION AND CONCLUSIONS

A relatively simple model has been derived to predict bed depth and the surface velocity of particles, based on the assumption that the shear stress in the active layer is constant for a given bed depth and rotation speed.

A correlation was presented to predict the shear stress within the active layer as a function of bed depth and rotation speed, \(m = 2(R/H)\omega + c\). The constant \(c\) is expected to depend on the properties of the bed material; data for polyethylene beads gives \(c = 9.5 \text{ s}^{-1}\).

By examining the effect of particles sliding over each other within the active layer, where void spaces are filled and formed again, a correlation was established for prediction of the effective dispersion coefficient \(D_{al}\). Our model predicts that as long as the reactant does not occur deeper than the active layer in significant concentrations, the mass transfer coefficient for the bed surface can be calculated as \(k_s = \sqrt{D_{al}ek}\).
These attempts to fundamentally predict the mass transfer coefficient were not successful, based on the data measured by Jauhari et al. (1998). The most probable reason for the poor correlation of our model with experiment was the fact that neither bed surface effects nor the granular temperature of the bed material were considered in the determination of the effective dispersion coefficient $D_e$. Future work should be directed at a means of predicting $k_sA$ based on granular temperatures within the active layer.
SYMBOLS

A  bed surface area, m²
a, b, c  symbols used to describe an ellipse (Fig. 6)
$C_A$  gas-phase concentration, mol m⁻³
$C_{A0}$  gas-phase concentration at $t = 0$, mol m⁻³
$C_{A,s}$  gas-phase concentration at the bed surface, mol m⁻³
d  active layer depth, m
D  binary diffusion coefficient, m² s⁻¹
$D_{al}$  total dispersion coefficient for the active layer, m² s⁻¹
h  perpendicular distance from radial center of the kiln to the bed surface, m
H  bed depth at the deepest point (H = R−h), m
k  gas-phase reaction constant, s⁻¹
$k'$  gas-phase reaction constant for use in active layer, s⁻¹
$k_s$  mass transfer coefficient, m s⁻¹
L  axial distance, m
m  shear stress in the active layer (velocity gradient of particles) s⁻¹
$P_A$  reaction rate, mol s⁻¹ m⁻³
Q  volumetric flow rate, m³ s⁻¹
R  kiln radius, m
V  volume, m³
f  factor for the directionality of dispersion, $\frac{1}{6} < f < \frac{1}{2}$
W  molar flux, mol s⁻¹
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Greek letters

\( \eta \)  effectiveness factor, dimensionless
\( \varepsilon \)  bed voidage, dimensionless
\( \lambda \)  dimensionless distance; depth below bed surface/active layer depth
\( \phi \)  thiele modulus, dimensionless
\( \psi \)  concentration, dimensionless

REFERENCES


Chapter 5 - A combined mass transfer model for both active and passive layers in rolling rotary kilns

ABSTRACT

A mass transfer model for rotary kilns that incorporates both the active and passive layer concentrations, is applicable over a wide range of rates. Dispersion effects are included in the active layer model, and convection (gas entrainment) effects are included in the passive layer model. At high $k/\omega$, it is not sufficient to use the contribution of the active layer alone. Gas entrained into the passive layer at the wall of the kiln makes an important contribution to mass transfer. At low $k/\omega$, the passive layer model alone can be used only for deep beds and low Thiele modulus $\phi$, otherwise the combined model is recommended.

By making two assumptions: 1) that molecular diffusion is unimportant compared to dispersion, and 2) that the particle velocity profile in the active layer ($m$) is proportional to rotation speed, then the effectiveness factor for the bed is dependent on $k/\omega$, $h/R$ and $R/d_p$. Of these, only $R/d_p$ is dependent on the scale of the kiln.
1. INTRODUCTION

In rolling rotary kilns, there are two distinct regions within the bed: the passive layer, where particles are transported concentrically with the kiln until they reach a point near the surface of the bed, and the active layer, where these particles cascade down the surface of the bed. It has previously been assumed that the active layer is the most important area for mass or heat transfer. Heydenrych et. al. (2001a) have shown that for slower reactions (or drying), mass transfer in the passive layer can also play an important role. In this paper, we explore the transition from mass transfer domination in the active layer, to kinetic domination in the passive layer.

The work of Heydenrych et. al. (2001a) explores how gas entrapped in the voids between particles in the active layer, gets depleted as it moves concentrically in plug flow through the passive layer with the particles.
They used an effectiveness factor to characterise the radial concentration profile, and a gas-phase volumetric rate constant to include mass transfer limitation, for example, for 1st order processes:

\[-P_A = \eta \varepsilon k C_A \quad (1)\]

By rewriting Eqn. (1) in terms of dimensionless units, they showed that the effectiveness factor depends only on \( k/\omega \) and the bed fill. In later work (Heydenrych et al. 2001b), the effect of diffusion within the passive layer was investigated. They showed that diffusion effects in the passive layer are unimportant for slower reactions (\( k/\omega < 1 \)), but importantly, they also showed that an additional dimensionless number must be used to describe mass transfer in faster reactions when diffusion effects are to be taken into account. This can either be a Thiele modulus, or (as they chose), a Peclet number, defined as

\[ Pe = \frac{\varepsilon \omega R^2}{D_e} \quad (2)\]

Jauhari et al. (1998) have measured mass transfer rates for the drying of relatively small silica-alumina particles soaked with decane. These results gave far greater mass transfer rates than the passive layer model (Heydenrych et al. 2001a) predicts. Jauhari et al. (1998) used an empirical model based on the volume of the active layer to predict mass transfer as a function of rotation speed.

Heydenrych et al. (2001c) developed a more fundamental model to predict the shape and volume of the active layer. This model was largely based on the assumption that the velocity gradient \( (m) \) of particles within the active layer is constant at any point within the active layer, given a set of
conditions such as bed depth and rotation speed. They predicted that bed depth could be described by

$$d = \frac{R^2 - x^2 - h^2}{m/\omega - 1}$$  \hspace{1cm} (3)$$

This model was further extended to predict the dispersion coefficient in the active layer caused by the change of void volumes that mix the gas as particles slide over each other. If successive layers of particles in the active layer move as rigid layers, then the dispersion of gas will occur predominantly in the direction perpendicular to the surface of the bed, and the volume of gas dispersed in this way can be calculated from the difference in voidage of a loose-packed structure and a tight-packed structure. Using these assumptions, they derived the following expression to calculate dispersion in the active layer:

$$D_{al} = 0.123 \, m \, d_p^2 + e^2 D_a$$  \hspace{1cm} (4)$$

They further showed that a relatively simple 1-dimensional model could be derived to predict the concentration gradients in the active layer in the case of fast reactions \((k/\omega > 10)\), giving rise to a correlation for a mass transfer coefficient for the surface of the bed:

$$k_s = \sqrt{D_\alpha e k}$$  \hspace{1cm} (5)$$

Using published data to predict the change of the velocity gradient \(m\) with rotation speed, they showed that the model represented by equations (3), (4) and (5) is inadequate for predicting the mass transfer rates measured by Jauhari \textit{et. al.} (1998). In this paper, we would like to adapt the fundamental approach adopted by Heydenrych \textit{et. al.} (2001c) to one that better predicts
the measured data of Jauhari et. al. (1998), for use as a design tool for rotary kilns and applicable to reaction rates that cover a large range. As a design tool, we need a model that is relatively easy to use, and some simplifying assumptions will be made so that the model can easily be solved on a spreadsheet.

2. COMBINED MODEL

We continue with the approach of Heydenrych et. al. (2001a), but we now explore whether the rate constant $k$ from Eqn. (5) is affected by the continuous mixing of gas as the layers of particles slide over each other.

On the basis of Sørensen & Stewart (1974) the “rate constant” describing the rate of evaporation of gas from the surface of wet particles in a stagnant packed bed can be formulated:

$$k = \frac{6ShD_a}{d_p^2} \frac{1 - \varepsilon}{\varepsilon}, \text{Sh} = 3.8$$

Heydenrych et. al. (2001a) have shown that estimation of $k$ using Eqn. (6) works well to predict the slow rate of evaporation from relatively large particles in the passive layer of rotary kilns, where there is a batch-wise saturation of gas in the voids as the gas rotates with the kiln. The gas concentration in the voids changes according to

$$k(C_{sat} - C_A) = \frac{dC_A}{dt}$$

(7)
Integrating Eqn. (7),

$$\frac{C_{sat} - C_4}{C_{sat} - C_{A0}} = e^{-kt} \tag{8}$$

In the active layer, the same process can be expected to occur, except that at regular intervals, the gas volume is flushed out with fresh gas. The frequency of this cycle is given by the shear rate $m$, and therefore, time ($t$) in Eqn. (8) is the inverse of $m$.

We wish to determine the rate of generation within a control volume $V$ in the active layer:

$$P_A dV = (C_4(t) - C_{A0}) m \bar{e} dV = (C_{sat} - C_{A0})(1 - e^{-k/m}) m \bar{e} dV \tag{9}$$

Replacing the generation term $P_A$ with a more conventional rate expression, as we have used in previous work,

$$\varepsilon k'(C_{sat} - C_{A0}) dV = (C_{sat} - C_{A0})(1 - e^{-k/m}) \varepsilon m dV \tag{10}$$

$$k' = m(1 - e^{-k/m}) \tag{11}$$

Lastly, we assume an alternative correlation to describe the relationship of $m$ with rotation speed. This is essentially an empirical correlation, chosen to reflect the mass transfer rates measured by Jauhari et. al. (1998):

$$m = 70 \omega \tag{12}$$

It is interesting to note that by substituting $m$ in Eqn. (12) into Eqn. (3), we find that the shape and volume of the active layer is not dependent on
rotation speed. Fig. 2 shows how this model predicts the maximum active layer depth \((x = 0)\) compared to the maximum active layer depths that were measured for various materials and drum sizes, and reported by Heinen et. al. (1983). For each material type, the active layer depth was measured at a range of rotation speeds.

![Graph showing the comparison between calculated and measured active layer thicknesses for different materials](image)

**Figure 2.** Surprisingly, relatively the steep particle velocity gradient of Eqn. (12) predicts deeper active layer depths than those measured visually.

Using this correlation for \(m\) and replacing \(k\) with \(k'\) of Eqn. (11), we predict the mass transfer rates measured by Jauhari et. al. (1998) as follows:
Figure 3: By using $m = 70\omega$ and $k' = m\left(1 - e^{-k/m}\right)$ we can reasonably predict measured mass transfer rates from $k_s = \sqrt{D_{al}\varepsilon k'}$. The data of Jauhari et al. (1998) is represented by ◆, and the line represents model predictions.

Within the active layer, we predict concentration profiles at the deepest part ($x = 0$), and assume that at any other part of the bed ($x \neq 0$) the same concentrations will be found at a given depth $y$ from the surface. The dimensionless concentration is estimated as:

$$\Psi = \frac{C_A(\lambda)}{C_{A0}} = \frac{e^{\phi} + e^{-\phi}}{e^\phi + e^{-\phi}}, \text{ where } \phi = d_{max} \sqrt{\frac{\varepsilon k'}{D_{al}}} \quad (13)$$

Heydenrych et al. (2001b) have shown that $k/\omega$ and either $Pe$ or $\phi$ can be used to describe concentration profiles in a rotary kiln. In this case, we prefer to use $\phi$ because we have assumed that all diffusion effects take place in the active layer. When binary diffusion is negligible, we can have

$$D_{al} = 0.123 \, m \, d_p^2 \quad (14)$$
We can now determine $\phi$ from Eqn. (13) by substituting for $d_{max}$ using Eqn. (3), $D_{al}$ using Eqn. (14), $k'$ using Eqn. (11) and finally $m$ using Eqn. (11):

$$
\phi = \frac{R}{d_p} \left[ 9.72 \varepsilon \left[ 1 - e^{-\left( \frac{1}{\lambda} \right) \frac{1}{70}} \right] \right] \left[ 1 - \left( \frac{h}{R} \right)^2 \right]
$$

(15)

This reduces the model to the same dimensionless numbers that were introduced by Heydenrych et al. (2001a), but with the addition of the third term $R/d_p$. This is the only term that takes dispersion (and therefore scale-up effects) into account, bearing in mind that Eqn. (15) is only valid when dispersion effects are much greater than molecular diffusion.

We will solve the model using the Thiele modulus $\phi$ as the third independent variable (rather than $R/d_p$), because it increases the validity of the simulation results: it is possible to include binary diffusion effects, or to use any other model to determine the Thiele modulus $\phi$.

The active layer model described above predicts the concentrations that occur at the interface between the active and passive layers of a rotary kiln. We use these concentrations as the starting concentrations for the gas as it moves concentrically around the kiln in the passive layer, getting depleted as it moves as it would in a plug flow reactor. This is similar to the model described by Heydenrych et al. (2001a), except that the surface boundary is not flat, but in the shape of the active layer interface, and the starting concentrations vary along the boundary. This is not easily solved analytically, but it can be quite easily solved numerically.

Both the active layer model and the concentric flow model described above give a concentration profile over the radial cross-section of the bed. The
effectiveness factor for a bed area (active or passive layer) is found by integrating the dimensionless concentrations over that area as described by Heydenrych et. al. (2001b):

\[
\eta_{al} = \frac{\int \psi dA}{A_{al}} \quad \text{and} \quad \eta_{pl} = \frac{\int \psi dA}{A_{pl}} \quad \text{with} \quad \psi = \frac{C_A}{C_{A0}}
\] (16)

We use the same \(C_{A0}\) for both the active and passive layers; \(C_{A0}\) is the freeboard gas concentration. We need to calculate the effectiveness of the active layer \(\eta_{al}\) separately from the effectiveness of the passive layer \(\eta_{pl}\) in order to get an overall effectiveness factor \(\eta\), defined in terms of \(k\).

\[
\eta_{ekA_{bed}} = \eta_{al} k' A_{al} + \eta_{pl} k A_{pl}
\] (17)

\[
\eta = \eta_{al} \frac{k'}{k} \frac{A_{al}}{A_{bed}} + \eta_{pl} \frac{A_{pl}}{A_{bed}}
\] (18)

The term \(k'/k\) is determined by substituting \(m\) (Eqn. 12) in Eqn. (11):

\[
\frac{k'}{k} = \frac{70\omega}{k} \left(1 - e^{-\frac{k}{70\omega}}\right)
\] (19)

Note that \(k'/k\) is only a function of \(k/\omega\). Therefore the model for predicting the dependent variable \(\eta\) is a function of three independent variables (we consider \(\varepsilon\) as a constant): \(k/\omega\), \(h/R\) and \(\phi\). We could replace \(\phi\) with \(R/d_p\) using Eqn. (15), but then the results we show would be limited to the model represented by Eqn. (15), so we have chosen \(\phi\).
3. MODEL PREDICTIONS

A contour plot of the concentrations found using the model developed here is shown in Fig. 4, where \( h/R = 0.5 \) and \( k/\omega = 1 \) for a clockwise rotating kiln. This shows the continuity of concentrations on the right hand side active/passive layer interface, where the gas moves into the bed. On the left hand side of Fig. 4, there is a very steep concentration gradient, which is unlikely to occur in practice due to diffusion effects there, and the transport of depleted gas from the active/passive layer interface into the active layer. We expect, from the findings of Heydenrych *et. al.* (2001b,c), that the diffusion effects in the active layer will predominate over the transport effects in the passive layer. If this is the case, then the model should be a reasonable indication of what happens in practice.

![Contour plot of dimensionless concentrations](image)

*Figure 4. A contour plot of dimensionless concentrations using the combined active/passive layer model with \( h/R = 0.5 \) and \( k/\omega = 1 \).*

Note how we find a high concentration at the wall of the kiln, because at the wall, the active layer depth is very small, and gas enters the concentric region at almost the maximum concentration.

Effectiveness factors at various \( \phi \) are shown as a function of \( k/\omega \) in Fig. 4.
Chapter 5 – Combined model

Figure 5. Effectiveness factors at various $\phi$ for a bed fill of 37% ($h = 0.2$). The thick line represents the passive layer model (Heydenrych et. al. 2001a).

Not surprisingly, for larger kilns (larger $\phi$) we always have a lower effectiveness factor. It is interesting to note that the active layer does not simply add to the mass transfer predicted by the passive layer model (Heydenrych et. al. 2001a), but predicts lower values than that model. This is due to the depletion of gas within the active layer, effectively blocking reactive gas from entering the passive layer.

Fig. 6 shows lines of constant $R/d_p$. These are useful because typically we find that $k$ varies along the length of a kiln as the solid dries (intraparticle diffusion limiting) or as the solid material becomes converted, in the case of reaction. This means that $\phi$ is not constant along the length of a kiln, and as reaction proceeds, we move from right to left on Figure 3 along a line of constant $R/d_p$. 

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Figure 6. Lines of constant $R/d_p$ are useful because typically $R/d_p$ remains constant over the length of a rotary kiln.

Fig. 7 shows a similar plot to Fig. 5, but with a shallow bed, with bed fill of 5.2% ($h = 0.8$). The trends are similar, but the effectiveness factor here is less dependent on $k/\omega$ because of the increased ratio of bed surface area (active layer) to bed volume. Here, the passive layer model of Heydenrych et. al. (2001a) is poor at predicting the shape of the curves, and cannot be used even for slow reactions. As bed fill decreases, the active layer volume increases relative to the passive layer volume. Therefore, even with slow reactions (low $k/\omega$), the active layer predominates and we do not benefit fully from the entrainment effect of the passive layer as it draws gases into the bed.
Figure 7. Effectiveness factors at various $\phi$ for a bed fill of 5.2% ($h = 0.8$). The thick line represents the passive layer model (Heydenrych et al. 2001a).

In both Fig. 5 and Fig. 7, we find low effectiveness factors even at low $k/\omega$. This is unlikely in practice, because when the reaction rate is slow, usually the Thiele modulus is also low. The lines of constant $R/d_p$ can be used as a guide. If we consider a rotary kiln using pelleted particles of 10 mm diameter, then a 4 m diameter kiln would give $R/d_p = 200$. It would be quite unusual to find a rotary kiln of a scale larger than $R/d_p = 1000$, because then molecular diffusion would predominate, and we can approximate this using the dispersion model with an appropriate value of $d_p$. We find that $d_p = 2$ mm gives $D_{ul}$ values that are consistent with molecular diffusion, and this gives $R/d_p = 1000$ for the 4m diameter kiln.

To examine the contribution of active layer at various bed heights, we determine what fraction the active layer contributes towards the total effectiveness of the bed. This is calculated using the 1st term of Eqn. (18) divided by the total effectiveness factor:
Fig. 8 shows for $\phi = 3$ how the active layer is always more important for shallower beds. It is also interesting to note that for deep beds, the passive layer continues to play a significant part in mass transfer even at high reaction rates.

The fraction contributable to the active layer does not approach unity for a combination of two reasons. The first is that even for fast reactions that occur very close to the surface of the bed, there is always an appreciable amount of gas that moves into the passive layer at the wall of the kiln, where the depth of the active layer tends to zero. The second effect is that
the weighting due to the term $k'/k$ comes into play, lessening the active layer contribution. For $k/\omega = 100$, for instance, $k'/k = 0.23$.

Noting that we typically get higher effectiveness factors using shallower beds, and that commercially operated kilns typically use shallow beds, it would be interesting to see if there is a maximum mass transfer that can be achieved for a given kiln radius. Fig. 9 shows an effectiveness factor based on total volume, rather than the bed volume that the previous effectiveness factor was based on. We see that there may be a maximum at the highest reaction rates ($k/\omega = 100$), but in general, a deeper kiln gives better mass transfer.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure9.png}
\caption{For fast reactions, it is possible to get an optimum bed fill, but in most cases better mass transfer will be obtained with a deeper bed.}
\end{figure}
Figure 10. Active layer contribution for a deep bed (fill 37%) does not change much as dispersion increases. \( \phi \) (top to bottom) = 0.1, 0.3, 1, 3 and 10.

Figure 11. Active layer contribution for shallow beds (fill = 5.2%) is highest for the larger kilns.
Chapter 5 – Combined model

For a shallow bed, we see a reversal in the order (Fig. 11), and we find that the active layer contribution becomes increasingly important as $\phi$ increases.

4. COMPARISON WITH MEASURED DATA

Applying the combined model to Jauhari’s data (Fig. 12) gives small but significant changes to the active layer model that was fitted using Eqn. (12).

![Figure 12](image)

Figure 12. The combined model predicts improved mass transfer rates at the higher rotation speeds compared to the active layer model. Jauhari’s data is represented by •.

The increase in the slope of the line predicted by the combined model is due to two effects: Firstly, at lower rotation speeds, dispersion is low, and the omission of binary diffusion from the model results in the prediction of lower mass transfer rates. The second effect is the additional effect of the passive layer. Figure 11 shows that for shallow beds, even at high $k/\omega$, there is still a significant contribution by the passive layer that the active layer model does not account for.
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The data of Heydenrych et. al. (2001a) is shown in Fig. 13 against their passive layer model (thick line) and the combined model (thin line), using $R/d_p = 4.7$. Although the scatter of the data does not allow definite conclusions to be made, it appears that the combined model fits the data better than the passive model alone.

![Figure 13](image)

*Figure 13. The combined model (thin line) fits the data of Heydenrych et. al. (2001a) somewhat better than the passive layer model (thick line).*

5. CONCLUSIONS

In attempting to simplify the relationship for the velocity gradient in the active layer to $m = 70\omega$, we arrive at some useful relationships that provide insight into the scale-up of rotary kilns. In particular, we showed that the dimensionless number $R/d_p$ describes the dispersion effect within a rotary kiln, and therefore it is the parameter that is important for scaling of rotary kilns.
Practically, we find a large range of $R/d_p$ in rotary kilns. An active layer model that calculates the mass transfer coefficient $k_s$ at the surface of the kiln can be used when the fraction contribution of the active layer is high. In many cases, this is not true, and the combined model presented here should be used. Also, the passive layer model used alone will in most cases incorrectly predict the mass transfer, having a totally different form (as a function of $k/\omega$) to that predicted by the combined model.

It would also be incorrect to assume that the mass transfer predicted by an active layer model can be added to the mass transfer from a passive layer model: we have shown that in many cases the depletion of gas in the active layer prevents activity in the passive layer.
## SYMBOLS

- **A**  cross-sectional area \([\text{m}^2]\)
- **CA**  gas-phase concentration, \([\text{mol m}^{-3}]\)
- **CA0**  gas-phase conc. at \(t = 0\) or freeboard gas conc., \([\text{mol m}^{-3}]\)
- **CA,s**  gas-phase concentration at the bed surface, \([\text{mol m}^{-3}]\)
- **d**  active layer depth, \([\text{m}]\)
- **d_{\text{max}}**  active layer depth at \(x = 0\), \([\text{m}]\)
- **D_a**  binary diffusion coefficient, \([\text{m}^2 \text{s}^{-1}]\)
- **D_{al}**  total dispersion coefficient for the active layer, \([\text{m}^2 \text{s}^{-1}]\)
- **D_e**  effective dispersion coefficient for a packed bed \([\text{m}^2 \text{s}^{-1}]\)
- **d_p**  particle diameter \([\text{m}]\)
- **f_{al}**  fraction contribution of active layer towards total effectiveness factor \([\%]\)
- **h**  perpendicular distance from radial center of the kiln to the bed surface \([\text{m}]\)
- **k**  gas-phase reaction rate constant \([\text{s}^{-1}]\)
- **k’**  gas-phase reaction constant for use in active layer, \([\text{s}^{-1}]\)
- **k_{gs}**  mass transfer coefficient on the surface of particles \([\text{m s}^{-1}]\)
- **k_s**  mass transfer coefficient of active layer \([\text{m s}^{-1}]\)
- **L**  length of the rotary kiln in axial direction \([\text{m}]\)
- **m**  shear stress in the active layer (velocity gradient of particles) \([\text{s}^{-1}]\)
- **P_A**  reaction rate of A \([\text{moles m}^3(\text{bed}) \text{s}^{-1}]\)
- **Q**  volumetric gas flow rate \([\text{m}^3 \text{s}^{-1}]\)
- **R**  internal radius of the rotary kiln \([\text{m}]\)
- **V**  volume \([\text{m}^3]\)
- **V_{\text{bed}}**  volume of bed \([\text{m}^3]\)
- **V_{al}**  volume of the active layer \([\text{m}^3]\)
- **x**  direction parallel to the surface of the bed in the radial cross-section; \(x = 0\) is at the centre \([\text{m}]\)
- **y**  direction perpendicular to the surface of the bed \([\text{m}]\)
Greek symbols

\( \varepsilon \) bed voidage \([m^3(gas) \cdot m^{-3}(bed)]\)

\( \lambda \) dimensionless distance; depth below bed surface/active layer depth

\( \eta \) effectiveness factor of reaction

\( \phi \) Thiele modulus

\( \psi \) concentration, dimensionless

\( \omega \) angular velocity \([s^{-1}]\)

REFERENCES


Publications


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Acknowledgements

At first glance, a doctoral thesis would seem to be an individual task, but it ultimately involves many people. I would like to acknowledge the support and contributions of the following people in particular:

My wife René, who supported my move to the academic world, and indeed supported me in this doctorate all the way. My whole family needs to be thanked, because an endeavour such as this inevitably encroaches on the time spent with them. And when I am with them physically, they will attest that I often seem far away . . .

To the University of Pretoria, and in particular Prof. Uys Grimsehl, who did many things to provide the environment for completing this thesis. He ensured that, apart from the core educational tasks that I was responsible for, my time was free to concentrate on my research. He supported my application to complete my studies at the University of Twente in every way. In that respect, I also owe a debt of gratitude to the other members of the Department of Chemical Engineering at the University of Pretoria for taking up some of my duties when I left the Department on study leave to the University of Twente.

Finally, to the University of Twente and my promotors Prof. Geert Versteeg and Prof. Hans Kuipers for initiating and funding my stay at the University of Twente. Special thanks go to my assistant promotor Dr. Bert Heesink who taught me how to add value to papers in review – a skill that I hope to be able apply much more effectively in my career in future. He spent a lot of his time not only on the thesis, but both he and Prof. Geert Versteeg spent a lot of time hosting me and my family in our stay in Enschede – something I value greatly.
My hope is that in future we will be able to keep regular contact, and strengthen the friendships we have established here, by actively promoting the co-operation agreement between our respective Universities.
Curriculum vitae

Mike Heydenrych spent his primary and secondary school years in Rustenburg, a town about 100 km west of Pretoria. He obtained his bachelors degree at the University of the Witwatersrand in 1982 and then completed two years of compulsory National Service. From 1984 he worked at CSIR (a contract research organisation) as a team member, determining the hydrodynamics of Fischer-Tropsch slurry reactors. The information was used by SASOL to scale up their slurry reactors, and these are now used industrially.

Still working for CSIR, he completed an MSc (at the University of the Witwatersrand) on the determination of the intrinsic kinetics of the oligomerisation of ethene on a supported nickel catalyst. He then spent two years as a consulting engineer for Megkon, a company specialising in systems engineering. In 1993 he returned to CSIR to work on fluidised bed combustors: full-scale design, supplying units to industry. After that, he headed a large minerals processing development project that that sparked his interest in mass transfer in rotary kilns.

In 1997, he joined the University of Pretoria where he is responsible for the undergraduate and postgraduate courses in Reactor design and for the Final year plant design project.

He is married to René with 2 children, Nicole and Graeme.