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MIXING AND REFINING DYNAMICS OF A GAS STIRRED THREE-PHASE REACTOR BY CFD ANALYSIS

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ABSTRACT

A multi-fluid model for gas stirred gas/liquid/liquid reactors with mass transfer between the two liquid phases has been developed within a commercial CFD code. Using the CFD model a scale up study of a gas/metal/slag reactor in which an unwanted impurity element is transferred from the metal to the slag phase has been carried out. Reactors of two different sizes and with four different gas-stirring rates have been simulated. The results show that scale up of the smaller reactor increases the metal productivity significantly, and that the refining rate increases significantly with increasing gas rate.

Keywords: CFD, refining, reactor

NOMENCLATURE

Nomenclature goes here, unless **all** symbols are defined in text.

INTRODUCTION

Gas-stirred reactors with two liquids are applied in refining processes where the purpose of the process is to mix the liquids well and facilitate fast transfer of impurities from one liquid to the other. Due to process conditions such as possible high temperatures and non-transparent liquids, it is often difficult to study the process in detail. Mathematical models describing the underlying physics may thus be helpful in understanding the local and global behavior of the process. In particular, they may serve as a tool for optimizing the reactor design and process conditions.

In order to quantitatively describe the refining process a mathematical model has been developed. Due to the complexity of the system, the model is not expected to predict results quantitatively accurate. Still the quantitative predictability is reasonable as shown below. The model will predict the qualitative behavior of the system, and the relative comparison between simulations with different parameter settings is expected to be very accurate. The model is thus applicable to analysis regarding process optimization and reactor design. In the following sections the model is presented and applied to a study on the scale up of a reactor with respect to gas rate optimization. Reactors may be designed in different shapes and with different types of gas inlets. We will focus on box shaped reactors with a bottom inlet for gas bubbles. The impurities are present in small concentrations, but the specification of the final product quality demands an even higher degree of purity.

MATHEMATICAL MODEL

To describe the mixing and refining dynamics of gas stirred reactors, a mathematical model has been developed within the framework of the CFD code Fluent (2005). The reactor is assumed to contain two liquids being mixed with the purpose of altering the composition of the liquids. Each liquid is a mixture of a solvent and different species present in small concentrations. Mixing of the liquids is driven by inert gas bubbles injected into the reactor. The gas injected into the reactor tends to accumulate at the top of the reactor where it acts as a cover gas. It is assumed that the reactor is well isolated such that near isothermal conditions prevail.

The mathematical model consist of an Eulerian multi-fluid model for the liquids and the cover gas. The gas bubbles are accounted for by a Lagrangian model interacting with the Eulerian phases through drag forces. The Eulerian phases also impose turbulent dispersion on the Lagrangian bubbles. Reasons for applying a mixed Eulerian/Lagrangian approach is given below. The dynamics of this mixing process is significantly affected by the size of the gas bubbles and the size of the liquid droplets. A model for bubble and droplet sizes is therefore necessary to describe the mixing dynamics properly. Droplet sizes are also important for the refining rate. Refining is accounted for by mass transfer of species between the liquid phases. The mathematical model presented below is later on applied to a three-phase reactor, but it will also be applicable to systems with more than three phases.

Eulerian Model

The fluid flow of the two liquids and the cover gas in the three-phase reactor is modeled by an Eulerian multi-fluid model. We solve for conservation of mass and momentum for each phase.

The continuity equation for phase q is

$$\frac{\partial}{\partial t}\left(\alpha_q \rho_q\right) + \nabla \cdot \left(\alpha_q \rho_q \vec{v}_q\right) = 0 \tag{1}$$

where α_q, ρ_q and $\vec{v}_q$ are the volume fraction, physical density and velocity of phase q respectively. In the phase mass conservation equations, mass transfer of species is neglected since the concentration of the impurity element is assumed never to exceed a few hundred ppmw.

The momentum equation for phase q is

$$\frac{\partial}{\partial t}\left(\alpha_q \rho_q \vec{v}_q\right) + \nabla \cdot \left(\alpha_q \rho_q \vec{v}_q \vec{v}_q\right) = \tag{2}$$

$$-\alpha_q \nabla p + \nabla \cdot \bar{\bar{\tau}}_q + \alpha_q \rho_q \vec{g} + \sum_{l=1}^{n} \vec{R}_{lq} + \vec{F}_p$$

where $\bar{\bar{\tau}}_q$ is the q^{th} phase stress tensor which for a Newtonian fluid is

$$\bar{\bar{\tau}}_q = \alpha_q \mu_q \left(\nabla \vec{v}_q + \nabla \vec{v}_q^T\right) - \frac{2}{3}\alpha_q \mu_q \nabla \cdot \vec{v}_q \bar{\bar{I}} \tag{3}$$

Here μ_q is the effective shear viscosity of phase q, p is the pressure shared by all phases, $\vec{R}_{lq}$ is an interaction force between phase q and l, and $\vec{F}_p$ is the interaction force from the gas bubbles accounted for by a discrete particle model. Turbulence is modeled by solving transport equations for turbulent kinetic energy and dissipation rate for each phase (i.e. *per-phase* k-ε model).

The interphase exchange is accounted for by the drag terms in the momentum equations. We have used the drag coefficient of Grace et al. (1976) which is based on a combination of the Morton Number

$$\mathrm{Mo} = \frac{g\mu_l^4(\rho_l - \rho_g)}{{\rho_l}^2 \sigma^3} \tag{4}$$

and the Eotvos Number:

$$\mathrm{Eo} = \frac{g(\rho_l - \rho_g)d_g^2}{\sigma} \tag{5}$$

The drag coefficient is calculated from a parameter J which is given by

$$J = 0.94H^{0.757} \qquad 2 < H \leq 59.3 \tag{6}$$

and

$$J = 3.42H^{0.447} \qquad H > 59.3 \tag{7}$$

where

$$H = \frac{4}{3}\mathrm{EoMo}^{-0.149}\left(\mu_l/0.0009\right)^{0.14} \tag{8}$$

The terminal velocity of the gas bubbles is

$$V_b = \frac{\mu_l}{\rho_l d_g}\mathrm{Mo}^{-0.149}\left(J - 0.857\right) \tag{9}$$

and the drag coefficent is

$$C_D = \frac{4}{3}\frac{g(\rho_l - \rho_g)d_g}{\rho_l V_b^2} \tag{10}$$

The drag coefficient is quite similar to the drag coeffiecent of Tomiyama (2004) for an air-water system, but is somewhat higher for a gas-metal system. For bubble swarms we modify the expression for the drag coefficent according to Wallis (1976) as follows

$$C_D = \frac{4}{3}\frac{g(\rho_l - \rho_g)d_g}{\rho_l V_b^2}\left(1 - \alpha_g\right)^{-1.7} \tag{11}$$

Since both the drag function and the particulate relaxation time depend significantly upon bubble and/or droplet size, the interphase interaction is strongly influenced by bubble and/or droplet size. Thus it is important to implement a good description of the average bubble and droplet size. A model for average bubble and droplet sizes is described below.

Lagrangian bubbles

Gas bubbles injected at the bottom inlet are accounted for by a Lagrangian discrete particle model. The model predicts translational motion of particles from Newton's second law. Bubbles are assumed to be spherical particles, although a shape factor may be implemented to describe motion of non-spherical particles. Newton's law gives the following equation for the velocity of the bubbles

$$m_p \frac{d\vec{u}_p}{dt} = \vec{F}_D + \frac{\vec{g}(\rho_p - \rho)}{\rho_p} \tag{12}$$

where m_p, $\vec{u}_p$ and ρ_p are the mass, velocity and density of the bubbles, ρ is the fluid density, $\vec{g}$ is the gravitational force and $\vec{F}_D$ is the drag force. Turbulent dispersion of bubbles is modeled by a stochastic discrete-particle approach known as *the discrete random walk model* (Fluent,2005). The applied drag force accounts for hindered settling, and is written in the following form

$$\vec{F}_D = \frac{\vec{F}_{SN}}{\left(1 - \alpha_p\right)^{5.1}} \tag{13}$$

Here $\vec{F}_{SN}$ is the drag force of Schiller and Naumann (1935) and α_p is the volume fraction of the gas bubbles.

The momentum transfer from the Lagrangian bubbles to the continuous phase is computed as

$$\vec{F}_p = \sum\left(\frac{18\mu C_D \mathrm{Re}}{24\rho_p d_p^2}\left(\vec{u}_p - \vec{u}\right)\right)\dot{m}_p \Delta t \tag{14}$$

where μ is the viscosity of the fluid, Re is particle Reynolds number, u is fluid velocity, C_D is drag coefficient, $\dot{m}_p$ is mass flow rate of bubbles, and Δt is the time step.

Bubble and Droplet Size

As mentioned above the size of bubbles and droplets significantly affect the dynamics of the reactor. The local average size $\tilde{d}$ of bubbles and droplets in the Eulerian phases is described by the following transport equation (Laux and Johansen, 1999):

$$\frac{\partial \tilde{d}}{\partial t} + \nabla \cdot \left(\alpha_d \rho_d \vec{v}\tilde{d} - D_{eff}\nabla \tilde{d}\right) = \alpha_d \rho_d \frac{d_{eq} - \tilde{d}}{\tau_{rel}} \tag{15}$$

Here D_{eff} is the effective dispersion coefficent, τ_{rel} is the relaxation time, and d_{eq} is the equilibrium diameter. The equilibrium diameter for bubbles and droplets in a turbulent flow is given by (Calderbank, 1958)

$$d_{eq} = C_1 \alpha_d^{0.5} \frac{(\sigma/\rho)^{0.6}}{\varepsilon^{0.4}} \left(\frac{\mu_d}{\mu}\right)^{0.25} + C_2 \qquad (16)$$

where α_d is the volume fraction of the dispersed phase being considered (i.e. either local bubble or droplet volume fraction), σ is the surface tension, ρ is the density of the continuous phase, μ_d and μ are the viscosities of the dispersed phase and the continuous phase, and ε is the turbulent dissipation rate of the dispersed phase. The coefficients C_1 and C_2 are given by empirical data. Note that Calderbank's relation was established for a system with only one dispersed phase. Still we have chosen to apply it to a system with multiple dispersed phases due to the lack of a more general relation.

Equation (15), which is based upon an Eulerian description, needs to be modified to be applicable to the gas bubbles, since the gas bubbles injected at the bottom are accounted for by a Lagrangian method. The Lagrangian version of Eq.(15) is given by

$$\frac{d\tilde{d}}{dt} = \alpha_d \rho_d \frac{d_{eq} - \tilde{d}}{\tau_{rel}} \qquad (17)$$

where diffusion has been neglected. The equilibrium size $d_{eq}$is given by Eq.(16) with ε being the turbulent dissipation rate of the continuous phase. Coefficients C_1 and C_2 will not be equal to the coefficients of the Eulerian bubbles and/or droplets.

Refining Model

The species conservation equation in an Eulerian frame of reference is given by the following equation:

$$\frac{\partial}{\partial t}(\rho_q \alpha_q C_{iq}) + \nabla \cdot (\rho_q \alpha_q \vec{v}_q C_{iq}) = -\nabla \cdot \alpha_q \vec{N}_{iq} + \dot{M}_{iq} \qquad (18)$$

where ρ_q, α_q and $\vec{v}_q$ are density, volume fraction and velocity of phase q, C_{iq} is mass fraction of species i in phase q, $\dot{M}_{iq}$ is mass transfer of species i to phase q, and $\vec{N}_{iq}$ is diffusion of species i due to concentration gradients in phase q

$$\vec{N}_{iq} = -\rho_q D_{iq} \nabla C_{iq} \qquad (19)$$

Here, D_{iq} is the diffusion coefficient for species i in the phase q.

The mass transfer rate for a species i in phase q is

$$\dot{M}_{iq} = \sum_p A_{pq} J_{iq\,jp} \qquad (20)$$

where A_{pq} is the interfacial area density between phase p and q and $J_{iq\,jp}$ is the mass flux of species i from phase q to species j in phase p. The exact description of the mass flux depends upon the nature of the refining physics. For typical absorption processes or processes involving infinite chemical reaction rates at the droplet interface we may derive the following equation for the mass flux (Deo and Boom, 1993):

$$J_{iq\,jp} = \frac{(L * C_{jp} - C_{iq})}{1/\rho_q k_{jp} + L/\rho_p k_{jp}} \qquad (21)$$

where L is the concentration ratio of the species in phase p and phase q at equilibrium, k_{iq} is the mass transfer coefficient of species i in phase q and k_{jp} is the mass transfer coefficient of species j in phase p. The mass transfer coefficient is given by the Sherwood number for spherical particles (Bird et al., 1960). The interfacial area density A_{pq} is a function of the droplet size d and the dispersed phase volume fraction α_d

$$A_{pq} = \frac{6\alpha_d}{d} \qquad (22)$$

Note that this only describes mass transfer at the interfaces between dispersed particulates and suspending fluid.

Boundary and Initial Conditions

Most boundaries in a geometry are treated as walls with *no-slip* condition for the fluid phases and reflective condition for the gas bubbles. Walls at the top of the geometry will allow gas bubbles to be ejected. Inlets are also treated as walls with the exception of a specified source of gas bubbles given as mass flow rate. If gas bubbles enter a region of pure gas, they are absorbed into the Eulerian gas phase (i.e. removed from the calculations).

Depending upon process conditions, different initial conditions may be studied. Here we assume that the fluids are at rest with a slag layer at the bottom, a metal layer above and gas at the top. Gas bubbles are introduced through a bottom inlet or a lance at the start of the process.

Implementation and Validation

The mathematical model described above is implemented in Fluent 6.2 with user defined functions and user defined scalars for bubble and droplet size, hindered settling, absorption of gas bubbles into the cover gas and mass transfer sources. Note that the turbulent dispersion model in Fluent 6.2's Eulerian multi-fluid model tends to generate stability problems in the presence of large scale interfaces, i.e. interfaces that separate continuous fluid layers. The gas bubbles, however, are significantly affected by turbulent dispersion and their distribution in turn strongly affects the flow pattern. This forces us to model the bubble phase in a Lagrangian manner and to leave out the effect of turbulent dispersion on the Eulerian droplets.

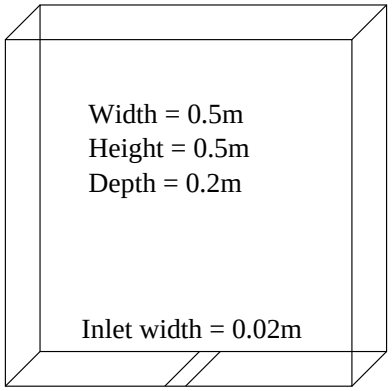

Figure 1: Box-shaped reactor with porous plug for gas injection.

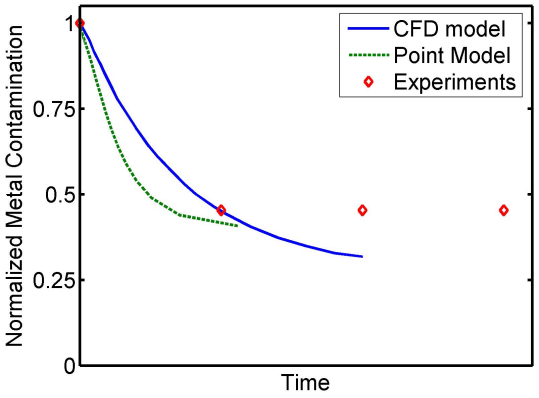

Figure 2: Normalized impurity concentration as function of time.

The model has been calibrated and validated against experiments in a transparent water-oil-air reactor. Water, silicone oil and air was used in a mixing experiment in a box-shaped glass tank illustrated in Fig.1. Gas bubbles were injected through a porous plug in the bottom of the reactor. The coefficients in the bubble and droplet size models were calibrated resulting in the values $C_1 = 0.37$ and $C_2 = 100\mu m$ for the Lagrangian bubbles and $C_1 = 2.0$ and $C_2 = 100\mu m$ for the Eulerian droplets (Olsen and Laux).

Refining experiments were carried out in a commercial gas stirred slag-metal reactor with gas injected through a lance. In Fig.2 we see the refining progress of an impurity in the metal phase. Results from the CFD model presented above are compared to experimental results and results from a point model. The point model assumes perfect mixing above the lance inlet and no mixing below. We see from Fig.2 that the CFD model is capable of describing realistic refining processes with reasonable accuracy for engineering purposes. Due to confidentiality further details of the experiment can not be released.

ANALYSIS OF REACTOR SCALE UP

Numerical calculations have been carried out to study the effect of gas rate and scale up upon the mixing and refining dynamics of a gas-metal-slag reactor with material properties at the system temperature as given in Table 1. The reactor has a volume of 50 l and is similar to the box shaped reactor described above with the exception of its content. Initially the reactor is filled with 40% slag at the bottom, 40% metal in the middle and 20% gas at the top. The metal contains 100 ppmw of a given impurity. Based on empirical observations it is assumed that slag is the continuous phase. The numerical calcultions have been carried out on a two-dimensional mesh of 10000 cells (i.e. 5mm resolution) with a timestep of 0.0002 sec. The Fluent solver uses a SIMPLE-based time-stepping algorithm (Flu, 2005) and allows the user to chose between different spatial (convective terms) and temporal discretization schemes. We have used first order accurate schemes for turbulence and species transport equations and higher order schemes for velocity and mass conservation equations. For discretization in time a second order implicit scheme was applied. The calculations were carried out until the contamination level declinded to 27 ppmw (25 ppmw being the equilibrium concentration). Typical CPU times were about 3 weeks on a single CPU node.

REST OF THE PAPER IS NOT SHOWN

Table 1 Normalized material properties at process conditions.

ρ_{slag}	1.07
ρ_{metal}	1
ρ_{gas}	$7.72 \cdot 10^{-5}$
μ_{slag}	259.01
μ_{metal}	1
μ_{gas}	0.02
$\sigma_{gas/slag}$	0.54
$\sigma_{gas/metal}$	1
$\sigma_{metal/slag}$	0.86
L	3
D_{slag}	0.15
D_{metal}	1

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