Supporting Information

CFD-DEM study of mass transfer mechanisms in riser flow

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Model Verification

Transient diffusion case

In this test case, the implementation of mass diffusion in the CFD-DEM code is verified. Single phase simulations have been performed in a rectangular duct of 0.2 × 0.02 × 0.02 meters, where the grid length in all dimensions are h = 0.005 m. At the bottom X-Y plane, the gas mass fraction of A is equal to 1, while 'zero flux boundary conditions' are applied at the side walls whereas the gas mass fraction of A in the remaining part of the simulation domain is zero. The inflow velocity is zero, so there is a diffusive flux of A throughout the simulation domain. The analytical solution of the axial gas mass fraction profile is determined by the error function (derived from 2$^{\text{nd}}$ Fick's law):

$$\frac{w_z - w_0}{w_s - w_0} = 1 - \text{erf}\left(\frac{z}{2\sqrt{D_{AB}t}}\right) \tag{1}$$

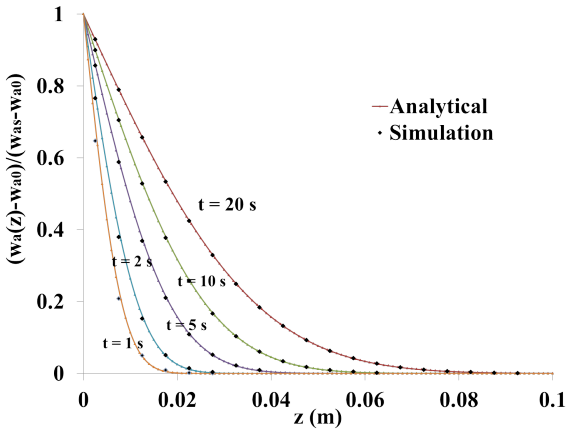

Figure S1: 1D profile of instantaneous gas mass fraction of A. h = 0.005 meters.

It can observed that the numerical solution is agreeing very well with the analytical results (see Figure S1).

Graetz-Nusselt test case

The Graetz-Nusselt case is performed in order to verify the correct implementation of mass convective terms of the convection-diffusion equation. In this case, there is an inflow velocity at the bottom plane of the domain and free-slip boundary conditions are applied for the gas phase at the side walls. Concerning the species field, the mass fraction at the bottom plane is equal to 1 and at the side walls equal to zero. So the side walls behave, in this case, as reactive surfaces where component A reacts at infinitely high rate (mass diffusion controls). The verification consists of the comparison of cross-sectional profiles of the gas mass fraction

Table S1: Simulation data.

Simulation Data			
D_{AB}	$2.0{\cdot}10^{-5}$ (m/s^2)	ρ_g	1 (kg/m^3)
μ	$2.0{\cdot}10^{-5}$ (kg/m·s)	P	101325 Pa
H × W × D	0.2 × 0.02 × 0.02	U	0.03 m/s

of A under fully developed flow conditions. To attain fully developed concentration profiles, the inverse of the Graetz number should higher than 0.1:

$$Gz = \frac{D_{AB} \cdot z}{U \cdot D^2} \geq 0.1 \tag{2}$$

where U is the gas velocity and D is the distance between the side walls. According to this criterion, fully developed flow is found at an axial distance z = 0.015 m. The analyti-

Table S2: Boundary conditions.

Bottom plane	$w_{A,w}$=0.0
Side walls	$w_{A,c}$=1.0
Top wall	$\frac{dw_A}{dz}$=0.0

cal solution of the mass fraction profile in a squared duct under fully developed conditions is:

$$\frac{w_{wall} - w(y,z)}{w_{wall} - w_{center}} = \left(1 - \frac{6}{5}\left(\frac{y}{Y}\right)^2 + \frac{1}{5}\left(\frac{y}{Y}\right)^4\right) \tag{3}$$

where Y is half distance between the side walls, y is the distance to Y.

Under these conditions the analytical solution for the Sherwood number is 2.977. This value was compared to Sh number computed from simulation data:

$$Sh_w = \frac{M_A D}{w_{A,w} - w_{A,m}} \tag{4}$$

where M_A is the mass flux towards the side walls, $w_{A,w}$ the mass fraction of A at the wall, $w_{A,m}$ the cup-averaged mass fraction of A and D the distance between the side walls. From Table S3, it can be seen that the error between the simulation result and the analytical solution is very small. Actually this can be significantly reduced when h = 0.0012 m. In Figure S2, the mass fraction profile of component A is plotted and compared to the analytical solution at an axial coordinate where the flow is fully developed. This test case demonstrates

that the implementation of the convective mass transfer terms is correct.

Table S3: Sherwood number of test case.

NX × NY × NZ	M_A (kg/m^{2}s)	w_m	Sh	Sh(an)	Error %
9 × 9 × 20	$1.89 \cdot 10^{-4}$	0.052	3.038	2.977	2.023
17 × 17 × 20	$1.97 \cdot 10^{-4}$	0.066	2.997	2.977	0.671

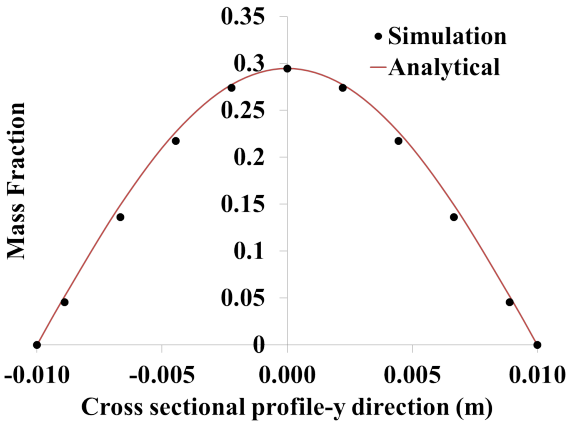

Figure S2: Cross-sectional profile mass fraction of A.

Chemical reaction in packed bed system

A packed bed reactor is simulated in order to validate the implementation of the chemical conversion due to a catalytic reaction. A mass transfer limited case is simulated, where the particles act as sink points with a mass fraction of component A equal to zero. Free-slip boundary conditions at the side walls are applied. The dimensions of the packed bed system are 0.2 × 0.02 × 0.02 meter. The cell numbers are 50 × 5 × 5. The gas density and viscosity are 1.2 kg/m^3 and $2.0 \cdot 10^{-5}$ kg/(m·s) respectively. The bed height is equal to 0.1 meter, and the bed porosity is equal to 0.9345. In this simulation pure A gas is injected through the bottom plane of the simulation domain at a gas superficial velocity of 4 m/s. Component A converts due to a fast reaction at the particle surface. Thus, the behavior of this system

can be described by a 1D plug flow model where the reaction rate is dictated by the mass transfer coefficient (Gunn correlation) which is approximately 0.238 m/s.

In this case, the analytical solution is :

$$\frac{w_A}{w_{A,0}} = \exp\left(-\frac{(1-\epsilon)a_v k_{mt} z}{U}\right) \tag{5}$$

The 1D profile of the gas mass fraction of component A at steady state is compared to the analytical solution in Figure S3 We can see that results almost overlap and the difference is negligible.

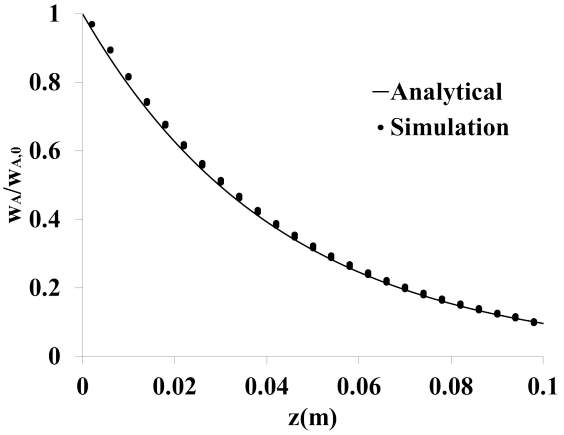

Figure S3: Validation of source term implementation.

Mass transfer contributions under riser flow conditions.

In this table, an overview of relevant research related to mass transfer on ri ser flows is presented. In the remarks column, a comment has been added to categorize each one of these contributions by the main topic related to mass transfer that is covered in these works or specifying a different fluidization regime than riser flow.

Table S4: Mass transfer contributions under riser flow conditions.

Authors	Modelling/Experimental	Physical/chemical system	Remarks
Resnick and White, 1949[1]	Experimental	Naphthalene Sublimation	Bubbling
Fryer and Potter, 1976[2]	Experimental	Ozone Decomposition	Bubbling
Dry et al., 1987[3]	Experimental	Heat pulse tracer	G-S
Watanabe et al., 1991[4]	Experimental	Heat transfer	G-S
Kagawa et al., 1991[5]	Experimental	Ozone Decomposition	G-S
Jiang et al., 1991[6]	Experimental	Ozone Decomposition	G-S
Pagliolico et al.,1992[7]	C/A Exp.	Ozone Decomposition	Validation
Sun and Grace, 1992[8]	Experimental	Ozone Decomposition	Effect of PSD
Kumar et al., 1992[9]	Experimental	Naphthalene Sublimation	Mass transfer
Ouyang et al., 1993[10]	Experimental	Ozone Decomposition	G-S
Vollert and Werther, 1994[11]	C/A - Exp.	NO Oxidation	Validation
Fligner, 1994[12]	C/A	Hydrodynamics	Validation
Van der Ham et al. 1994[13]	Experimental	Naphthalene Adsorption	Mass transfer
Li and Kwauk, 1994[14]	EMMS	Hydrodynamics	Clusters
Koenigsdorff and Werther, 1995[15]	C/A	Hydrodynamics	Validation
Ouyang et al., 1995[16]	Experimental	Ozone Decomposition	G-S
Pugsley,1996[17]	C/A	Hydrodynamics	Validation
Schöenfelder, 1996[18]	C/A - Exp.	Ozone Decomposition	Validation
Zethraeus, 1996[19]	C/A	Hydrodynamics	Validation
Kunii and Levenspiel, 1997[20]	C/A	Hydrodynamics	G-S
Venderbosch,1999[21]	Experimental	CO Oxidation	Sh overall
Zhu et al., 1999[22]	Experimental	Heat transfer	G-S-downer
Contractor et al., 2000[23]	Experimental	Argon gas tracer	G-S
Bolland et al., 2001[24]	Experimental	Ozone Decomposition	Sh overall
Wang and Li, 2002[25]	TFM/EMMS- Exp.	Naphthalene Sublimation	Validation
Subbarao and Gambhir, 2002[26]	Experimental	Naphthalene Adsorption	Sh overall
Scala,2007[27]	Experimental	CO Oxidation	Bubbling
Subbarao, 2008[28]	Cluster model	Mass Transfer	Cluster model
Dong et al., 2008a[29]	TFM/EMMS	Naphthalene Sublimation	Validation
Dong et al., 2008b[30]	TFM/EMMS	Ozone Decomposition	Validation
Chalermsinsuwan et al., 2009[31]	TFM/EMMS	Ozone Decomposition	Sh overall
Prajongkan et al.,2009[32]	TFM/EMMS	Ozone Decomposition	Sh overall
Breault et al., 2009[33]	Experimental	Naphthalene Sublimation	Sh cluster-bulk
Shuyan et al., 2009[34]	CFD-DEM	Naphthalene Sublimation	Particle-Sh
Hou et al., 2010[35]	TFM/EMMS	Ozone Decomposition	Validation
Wang et al., 2010[36]	TFM/EMMS	Hydrodynamics-Mass Transfer	Review
Ge et al., 2011[37]	TFM/EMMS	Hydrodynamics-Mass Transfer	Review
Kashyap et al., 2012[38]	TFM/EMMS-Exp.	Ozone Decomposition	Sh overall
Chen et al.,2012[39]	TFM/EMMS	Gas desulfurization	Validation
Hou et al.,2013[40]	TFM/EMMS- Exp.	CO oxidation	Validation
Li et al., 2013[41]	Experimental	Ozone Decomposition	G-S
Wang et al., 2015[42]	Experimental	Ozone Decomposition	G-S

* G-S: Gas-solid contact efficiency. PSD: Particle size distribution.

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