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Development of an atmosphere particle kinetic model for particle reactions in a combustion Flash-Reactor using CFD- methods.

Franz Edler* [a], Bernhard Geier [a], Wolfgang Reiter [a], Johannes Rieger [a], Christoph Spijker [b], Harald Raupenstrauch [b]

[a] *K1-MET GmbH, Linz, Austria*
[b] *Chair of Thermal Processing Technology, Montanuniversitaet Leoben, Leoben, Austria*

Abstract

The Reco-Dust-Process offers new possibilities for minimum waste strategies in the iron industry. The idea was realized in the Flash-Reactor pilot plant, which is meant to recycle steel mill dusts and residues. The input material is separated into two usable fractions, an iron ore substitute and a zinc oxide rich dust.
For a system scale-up the mathematic model was built to describe the process in CFD (Computational Fluid Dynamics) to support the research. The main process parameters are particle size and composition, gas composition, particle and gas temperature, burner and reactor geometry. With the "atmosphere particle kinetic model for particle reactions" it is possible to check the reactions in range of the calculation settings. First, a model for checking the user defined function of the gas and particle system was developed. Parcels with pre-calculated flue gas equilibrium compositions at certain temperatures are added in gas with fixed temperature and transported by the flue gas. With the rise of the temperature, heat and mass transfer of the particles are implied. In detail the behavior of the different parcel classes and the compositions over the residence time could be verified. Transition components are transferred to the gas phase and finally the evaluated "atmosphere particle kinetic model" is implemented in the main case.
The main case contains the real geometry of the Flash-Reactor or possible upscaling geometries. An Eddy Dissipation Concept (EDC) is chosen for the combustion model. The calculation results are compared with experimental data.

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Keywords: furnace modelling; Flash-Reactor; Eddy Dissipation Concept; steel mill dusts; Reco-Dust;

*Corresponding author. Tel.: +43 3842-402-5821
E-mail adress: franz.edler@k1-met.com

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Nomenclature

$n_{p,i}$	chemical reaction rate of the component i in the particle [mol/s]
$k_{r,i}$	reaction rate constant of the component i [m/s]
r_p	particle radius [m]
C_i	concentration of reactant of the component i [mol/m^3]
Ke_i	equilibrium constant of the component [-]
T	temperature [K]
$k_{f,r}$	forward reduction rate constant [m/s]
A_r	pre-exponential factor [consistent units]
β_r	temperature exponent [-]
E_a	activation energy for the reaction [J/kmol]
R	universal gas constant [J/kmol-K]

1. Introduction

The Reco-Dust Process is a new pyro metallurgical treatment for steel mill dusts and residues with low zinc contents. Due to their high iron contents, these dusts are supposed to be reused with in the steel production site, but with increasing zinc content a recycling is often limited or impossible without treatment. A treatment with the state of the art Waelz-Kiln is only economical with zinc contents around 20 %. Most dusts from the blast furnace route are between these boundaries, so they have to be recycled externally or landfilled.

Aim of the Reco-Dust process is the production of two fractions: A zinc rich oxide fraction and an iron rich fraction. The purity of the products has to be high enough to insure that both fractions can be used as secondary resources in the zinc or iron industry.

The process uses a reducing atmosphere provided by an oxygen/natural gas burner with temperatures around 1700 °C. Within these conditions, the fine dust melts and the zinc oxide is reduced and vaporized. The metallic zinc leaves the reaction chamber with the exhaust gas and is transferred into zinc oxide by means of a post combustion step. The exhaust gas is cooled and finally, a bag filter system can separate the zinc oxide from the flue gas.

The non-volatile components gather at the bottom of the reaction vessel being discontinually tapped as a slag.

The benefit of the RecoDust-Process is the absence of foregoing treatment steps for the dusts. However, a free floating and dry dust with a grain size less than 1mm represent limits to ensure a fast melting procedure. Unlike most other established processes, no additives are charged into the process, the reducing conditions are adjusted only by control of the oxygen flow in the mixing cyclone and the burner lance.

2. Model description

2.1. Atmosphere particle kinetic model

The atmosphere particle kinetic model is a useful tool to analyze the particle kinetic in dependence on the residence time. As a consequence it's possible to check the influence of different process parameters prior to the tests in the pilot plant. The most important parameters are gas temperature, gas composition, gas velocity, particle size, particle composition and the particle inlet temperature respectively. The flue gas equilibrium composition is pre-calculated by Cantera 2.2.1 [2], by fixed temperature and defined oxygen-fuel ratio.

The dimension of the cross section is based on the experimental reactor. So it gives an expression of the flow through the reactor. The fluid transport is solved in Eulerian framework and the dispersed phase is represented as Lagrangian particle. Turbulence was modeled by a realizable-k-ε model, which produces the most stable results. The flue gas composition is calculated with a reduced 17 species mechanism [3] designed to model natural gas combustion.

User defined function have been used for particle reactions and heat and mass transfer between fluid and disperse phases. The particle contemplation is based on a non-porous dust particle in different composition and size distribution. The atmosphere particle kinetic model investigates the composition change depending on the reaction rate in the particle for a certain size and atmosphere. The particle size is competed every time step.

$$n_{p,H_2} = 4\pi r_p^2 * k_{r,H_2} * \left(1 + \frac{1}{Ke_{H_2}}\right)\left(C_{H_2} - \frac{C_{H_2O}}{Ke_{H_2}}\right) \quad (1)$$

$$n_{p,CO} = 4\pi r_p^2 * k_{r,CO} * \left(1 + \frac{1}{Ke_{CO}}\right)\left(C_{H_2} - \frac{C_{CO_2}}{Ke_{CO}}\right) \quad (2)$$

$$k_{f,r} = A_r * T^{\beta_r} * e^{-\left(\frac{E_a}{RT}\right)} \quad (3)$$

The forward reaction rate constant (1) (2) is calculated by using the Arrhenius expression (3) in a form it is implemented in ANSYS Fluent v15. The reversible reaction rate constant is calculated by the equilibrium constant.

Table 1. Rate parameters for reaction parameters

	Particle-reaction	k_0 (m/s)	Ea (kJ/mol)	Ref.
R1	$3\ Fe_2O_3 + H_2 \Rightarrow 2\ Fe_3O_4 + H_2O$	160	92,1	[4]
R2	$Fe_3O_4 + H_2 \Rightarrow 3\ FeO + H_2O$	23	71,2	[4]
R3	$FeO + H_2 \Rightarrow Fe + H_2O$	2858,3	117,2	[4]
R4	$3\ Fe_2O_3 + CO \Rightarrow 2\ Fe_3O_4 + CO_2$	2700,0	113,9	[4]
R5	$Fe_3O_4 + CO \Rightarrow 3\ FeO + CO_2$	25	73,7	[4]
R6	$FeO + CO \Rightarrow Fe + CO_2$	17	69,5	[4]
R7	$ZnO_{(s)} + CO \Rightarrow Zn_{(g)} + CO_2$	9750	158,573	[5]
R8	$ZnO_{(s)} + H_2 \Rightarrow Zn_{(g)} + H_2O$	9270	183,677	[6]

The particle reactions from Table 1 are representing the key reactions for the first development step of the atmosphere particle kinetic model. The frequency factor (k_0) and activation energy (E_a) are taken from the reference papers recommended in the last column. For the user defined function, the pre-exponential factor is equal to the frequency factor and the temperature exponent is set to 0. So the Arrhenius expression is reduced to the same form as in [7]. The first model is based on a reaction limited to the particle reaction system.

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2.2. Flash-Reactor and Burner

The Flash-reactor pilot plant is built up in the technical center, chair of the Thermal Processing Technology at Montanuniversitaet Leoben. So the Simulation data can be compared with the experimental results. Especially new burner geometries, dust loading systems and the reactor design is part of further research. Therefore it is essential to develop a fast simulation model for a quick check and a verified detailed model for the heterogenic system. For the CFD study, a three dimensional unstructured grid of the reactor is meshed in the commercial software ANSYS ICEM v15.0 and imported in Ansys FLUENT v15.0 where it is converged into a polyhedron mesh.

The fluid transport and the turbulence model are the same as used by the atmosphere particle kinetic model. An Eddy Dissipation Concept Model EDC with a reduced 17 species gri mechanism [3] is used for the combustion process. The user defined function for the heat and mass transfer from the atmosphere particle kinetic model was used for the particle kinetics. The particle size distribution is implemented with a Rosin-Rammler distribution.

3. Results and Discussion

3.1. Results of the atmosphere particle kinetic model

Figure 1 shows the change of the particle mass, specific mass of the components and the particle temperature in dependence on the residence time, respectively. Fixed parameters of the system are the particle inlet temperature of 300 K, the inlet temperature of the flue gas 2100°C and the inlet velocities of particle and gas, respectively.

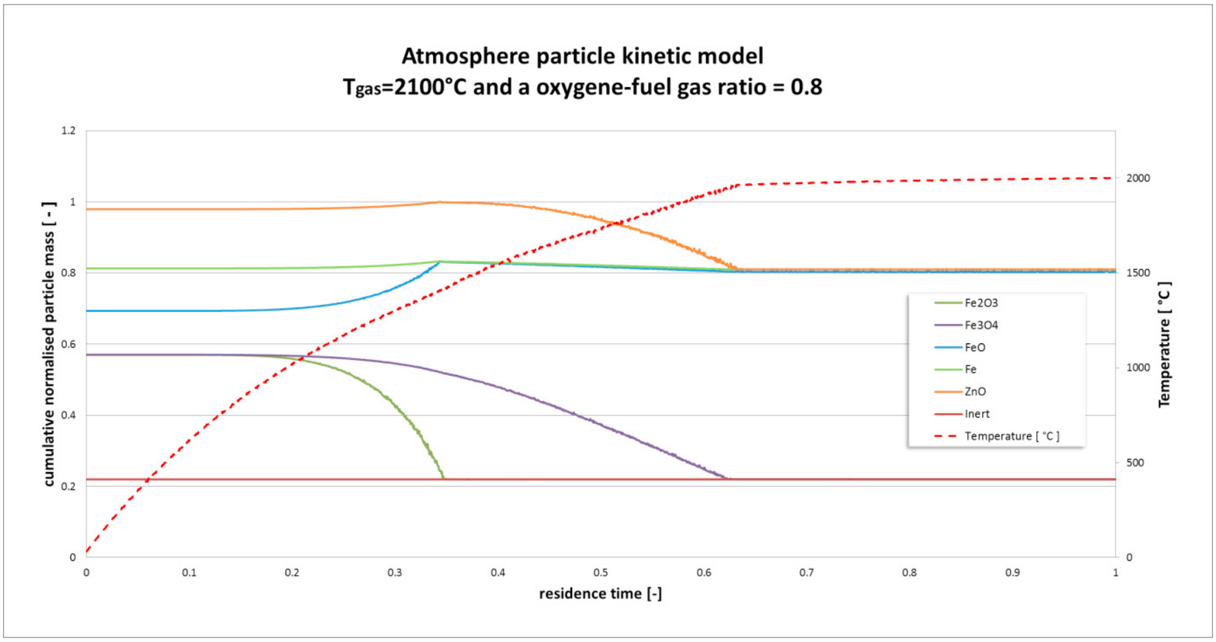

Fig. 1. Results of the atmosphere particle kinetic model. Cumulate mass and particle temperature of the composition over residence time of the particle.

Due to the fixed dimension of the grid, based on the geometry of the reactor, the residence time of the particle is influenced by the particle start velocity and the gas velocity of the system. Due to the increase of the temperature, the reaction changes the composition of the particle. The absolute mass of each component varies and is calculated in each time step. In top of Figure 1 the absolute mass of the particle by low residence time is constant. Up to higher residence times, the particle compositions will change at first. After the complete oxidation of metallic iron into Wüstite (FeO) and a particle temperature of a minimum at 1500°C, the kinetic of the zinc oxide (ZnO) starts and the total mass of zinc oxide in the particles gets reduced. Hematite (Fe_2O_3) is reduced to Magnetite (Fe_3O_4) and finally to Wüstite (FeO) at the given temperature and fuel gas composition. The inert mass in the particle is constant over the residence time. Studies of the particle kinetics are made for different ranges of flue gas temperature, particle temperature and oxygen fuel ratio, respectively.

3.2. Results simulation and experimental data

The particle inlet is located at the top of a cyclone charging hopper. Through a tangential secondary oxygen inlet the particles are mixed with pure oxygen and enter the reaction chamber. They are fed directly into the centrally mounted tube burner. The volatile components leave the reaction chamber with the flue gas at the side of the reaction vessel, while the oxide phase is collected at the bottom of the reactor. After post combustion, the exhaust gases are cooled and the re-oxidized zinc oxide (ZnO) is separated with the help of a baghouse filter.

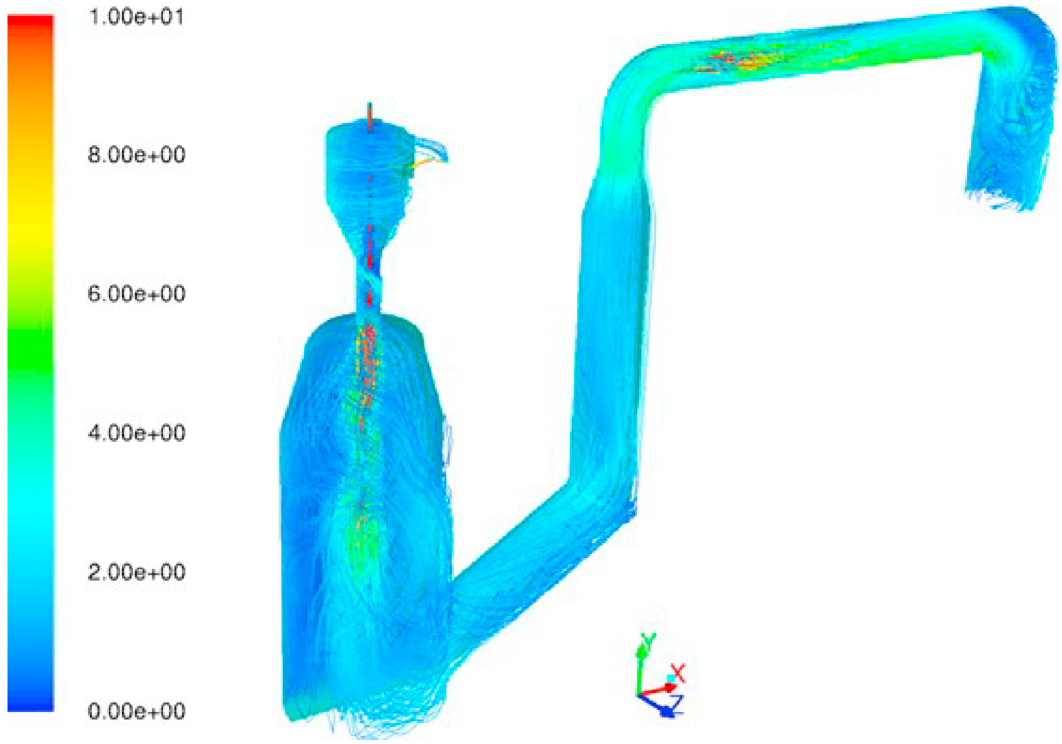

Fig. 2. Flash-Reactor simulation with a tube burner and a cyclone particle charging hopper. Path lines are colored by velocity magnitude [m/s].

The path lines of the velocity colored by the velocity magnitude are shown in the Figure 2. The color bar of the velocity does not represent the maximum of the velocity range. The velocity path lines only give an impression of the vortex stream in the reactor. So we can clearly see that in the reactor doesn't represent a plug flow. The residence time of the particle is influenced by the particle mass depending on the way through the reactor. In that case the particles are calculated by a Rosin-Rammler distribution and are based on the experimental data. The quick change of the flow direction over the slag reservoir causes a segregation of the particle distribution. Small particles will be taken by the gas flow up to the top of the reactor, where as other particles drop out on the reactor wall or on the slag reservoir surface. Small size particles which didn't drop out represent the carry-over of the system.

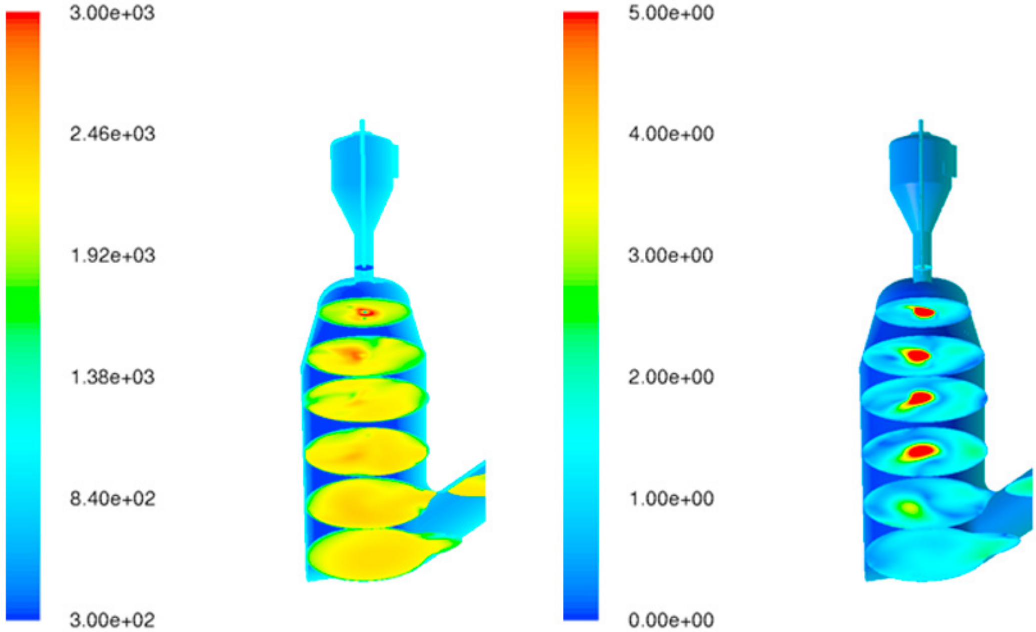

Fig. 3. Flash reactor simulation with a tube burner and a cyclone particle charging hopper. Left contours of Static Temperature [K], right Contours of Velocity Magnitude [m/s].

To minimize the particle carry-over effect on the reactor outlet is important for the process, caused by the particle carry-over the zinc oxide fraction gets an impact of iron oxide, which is not desired. Therefore the reactor geometry will have to be optimized in further research for the scale up process. Cross sections through the reactor show the inhomogeneous conditions in the different attitudes. In Figure 3 on the left hand side, the cross section showing the contours of static temperatures from 300 K to 3000 K and on the right hand side the contours of velocity magnitude from 0 [m/s] to 5 [m/s] give a good impression of the reactor conditions. The temperature and the velocity color bar don't represent the full range from minimum to maximum; it's a selected range for this view. The flame of this tube burner has an asymmetric shape, which is caused by the vortex and the back stream in the reactor.

3.3. Results of the Lagrangian phase

The Results of Lagrangian phase at every boundary were particles being trapped at the wall film or slag surface. The values are written in a file and post-processed by Matlab 2013. Furthermore it's possible to analyze the particle distribution, residence time, temperature and the composition due to their behavior to each other. The slag surface from the bottom of the reactor is discussed in detail.

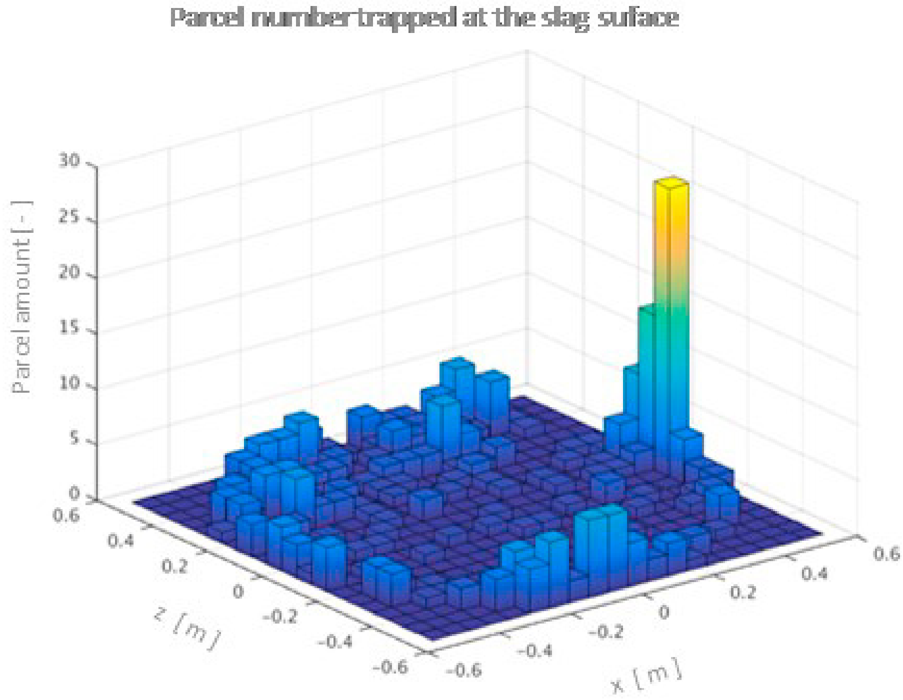

Fig. 4. Flash-Reactor simulation with a tube burner and a cyclone particle charging hopper. Parcel amount trapped at the slag surface countered at a raster of 50 mm.

Figure 4 presents the number of parcels, which are trapped at the slag surface in a raster-width of 50 mm. The calculated time of the simulation is only a few seconds so the parcel number is low. It is possible to analyze how many parcels are trapped on which location on the surface, as shown in Figure 4, the most particles are found on the right side near the reactor exit towards the exhaust pipe. In the center of the slag surface, the smallest amounts of parcels are collected.

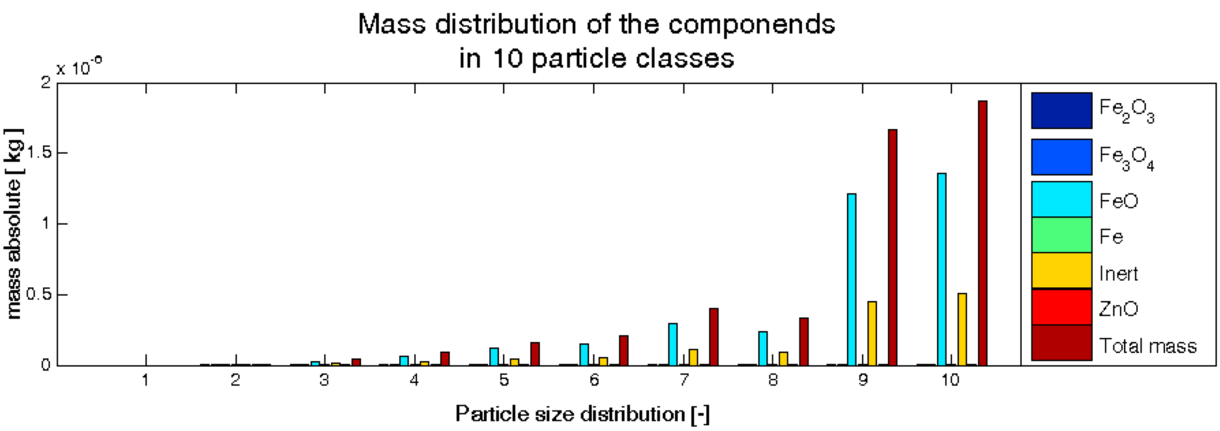

Fig. 5. Flash-Reactor simulation with a tube burner and a cyclone particle charging hopper. Sum of particle mass trapped at the slag surface; particle size distributions in 10 particle size classes.

As shown in Figure 5 the absolute mass of the particles with a diameter up to 150 µm has the most mass impact on the slag surface. These particle distributions have the lowest residence times and the most absolute mass impact of zinc oxide on the slag surface. The reactor wall and the carry-over are to be considered in the same way.

4. Conclusion

The results from the simulations are in good agreement with the experimental data. The atmosphere particle kinetic model is a suitable concept to improve the particle model within the calculation and to validate the system parameters. The implementation of the particle kinetic model into OpenFOAM (open source) is part of further research. To support the scale up development a fast model concept was proofed. A simple flamelet hybrid model with thermo parcels for geometry studies of the burner and the reactor design is still in validation.

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