A COMBINED EXPERIMENTAL AND NUMERICAL APPROACH TO A DISCRETE DESCRIPTION OF INDIRECT REDUCTION OF IRON OXIDE

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ABSTRACT

Blast furnaces are complex counter-current reactors designed to reduce chemically iron oxides and melt them to liquid iron. The complex processes in blast furnace iron making involve various aspects of thermodynamics, fluid dynamics, chemistry and physics. Physical, thermal and chemical phenomena occurring within the process are highly coupled in time and space. In order to generate a more detailed understanding of the indirect reduction of iron ore, the innovative approach of the Extended Discrete Element Method (XDEM) is applied. It describes the ore particle as discrete entities for which the thermodynamic state e.g. temperature and reduction degree through a reaction mechanism is described individually for each particle. The flow within the void space between the particles is represented by classical computational fluid dynamics that solves for the flow and temperature distribution including the composition of the gas phase. Ore particles and gas phase are tightly coupled by heat and mass transfer, that allows particles to heat up and to be provided with the reducing agent i.e. carbon monoxide. Reduction of iron oxide is predicted by a set of equilibrium reactions that represent the phase diagram of iron oxides at different oxidation levels. The reaction mechanism was validated by experimental data for a single ore particle for different temperatures. A comparison between measurements and predictions yielded good agreement so that reduction of iron oxide to iron was represented by a single mechanism including all reduction steps.
The validated reaction mechanism was then applied to each particle of a packed bed that was exposed to defined gas flow with its temperature and composition. The predicted results were also compared to experimental data and very good agreement was achieved. Due to the resolution of iron reduction on a particle level, detailed results of the entire reactor were obtained unveiling the underlying physics of the entire process. Results showed the reduction state of each particle during the entire period and additionally revealed the inhibiting influence of a non-uniform flow distribution. It provided regions of the packed bed with

insufficient amounts of the reducing agent and thus, allowed identifying drawbacks for design and operation.

KEYWORDS

Iron reduction, packed bed, Extended Discrete Element Method (XDEM), experiments, modelling, Computational Fluid Dynamics (CFD)

INTRODUCTION

Further improvements of computational power and advances in numerical and mathematical modelling of multiphase systems allows to combine the advantages of two different numerical approaches to build hybrid or so-called coupled models: A discrete approach like DEM accounts for the solid phase whereas a continuous approach like Finite Element Analysis (FEA) or Finite Volume Method (FVM/CFD) accounts for the gas phase [1, 2, 3, 4, 5]. In this way each phase is inherently treated with the most appropriate approach. With these models it is possible to investigate furnace permeability and the influence of the gas flow on the solid descend in much greater detail. Some insights from such simulations are detailed local information about solid and gas flow patterns, local particle velocities, force and stress networks of the solid feedstock which can then be used to evaluate the shape of the cohesive zone or the residence time or temporal history of individual particles. Further it can be investigated how disturbances for example in the raceway influence the symmetry inside the furnace or the stability of the solid flow patterns.

Boom et al. [4, 5] investigate how different boundary conditions of a numerical model and the shape of particles influence the results from a DEM simulation. Comparing the results for a single particle geometry to a full diameter slot model with periodic boundary conditions and a full 3D simulation shows good agreement: Without gas flow deadman formation is almost non-existent. Regarding the shape of particles the size of a deadman increases when using non-spherical particles. This is explained by the resistance to rolling of non-spherical particles compared to spherical ones and thus a deadman formed is more stable. Finally, to study the influence of the gas flow Boom et al. [4, 5] use a slot type model of a lab scale blast furnace similar to the one by Zhou et al. [6]. The entire furnace diameter is considered and periodic boundary conditions are used. They compared isothermal, incompressible DEM-CFD simulation for two different gas flow rates. Results show that due to the lift created by the gas flow void space in the bed and thus permeability is increased when gas flow rate is increased. In accordance with findings by Nouchi et al. [7] the bed is supported by a force network resting below the belly of the reactor. An increase in gas velocity also reduces stresses on the particles located in the force network. Based on the observed loosening of the bed the layered structure is changed on the centre axis of the furnace in favour of a pellet free, coke only region. Lower compressive stresses allow the smaller and sphere shaped pellets to escape into available voids thus effectively shifting them towards the periphery. These pellet free regions due to the larger size and non-spherical shape of the coke particles cause a higher gas permeability just like the region close to the furnace wall. Thus in these regions higher volume flow rates are prevailing which cause higher lift on the burden. Accordingly the descend of the feedstock is decelerated in these regions. With this explanation the formation of a W shaped layer structure and a deadman can be explained. The authors remark that accounting for temperature gradients and compressibility in the gas phase might increase these effects due to expansion and compression phenomena.

Zhou et al. [3] extended their DEM model by an isothermal CFD approach for the gas phase. A blast furnace slot model of 4,5 m in height with coke particles of 40 mm and pellets of 30 mm in diameter. Pellets are allowed to shrink and disappear within the predefined cohesive zone. The shape of the latter can be chosen to be of V-, inverted V- or W shaped type. In contrast to the model by Adema et al. [5] pellet layers inside the cohesive zone are considered impenetrable. Solid flow pattern are in good agreement with findings of other researchers and from literature. For the case of a V shaped cohesive zone they observe considerable changes of solid flow and gas flow inside the cohesive zone in time.
Particles located inside the dead man are experiencing the largest compressive forces and stresses. This is caused by the burden column resting upon them. In contrast particles located on the inclined plane along the cohesive zone experience by far less compressive forces. This explains the loose packing and high solid descend velocities in these locations. In a previous paper [8] the authors present findings similar to the ones by Adema et al. Increasing the gas flow rate loosens the bed and reduces compressive stresses on particles and the number of contact partners. Further, vertical lift exerted by the gas phase is largest close to the raceway. Due to the impermeability of the pellet layers inside the softening zone the gas flow is redirected which causes high forces in horizontal direction exerted on the particles. 90 Kurosawa et al. [9] added shrinking and softening behaviour of particles due to load and material softening within the cohesive zone into their coupled model. In contrast to the model by Zhou et al. [3] ore layers in a specific region are not per se defined impenetrable but particles are allowed to overlap thus inherently reducing the void fraction of the bed and influencing the gas flow due to their softening. Softening is modelled by changing the Young's modulus of the ore particles. They calibrate their model by carrying out numerical compaction experiments for rectangular box of spherical particles but do not present any validation with experiments.

1. MEASUREMENTS AND EXPERIMENTS

Reduction experiments were carried out in a laboratory-scale experimental set-up as shown in fig. 1.

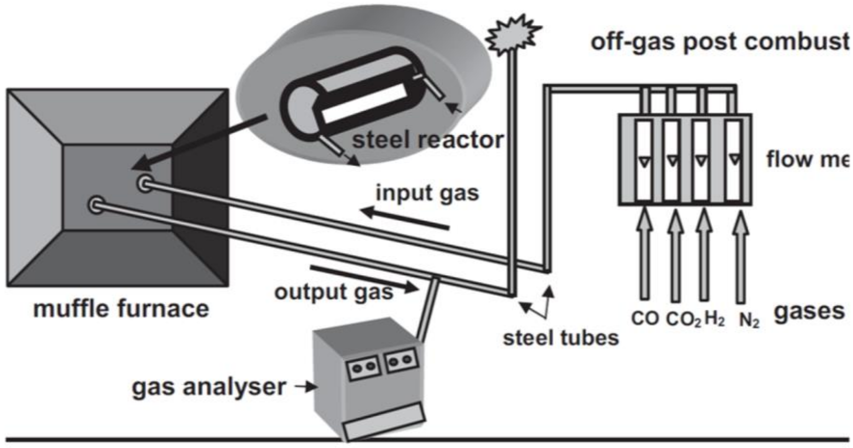


Figure 1: Experimental setup of the reduction system for iron bearing materials

The experimental set-up includes a muffle furnace, in which the reduction of iron bearing materials took place. A stainless reactor, of which the dimensions are depicted in fig. 5 for isothermal and non-isothermal reduction, respectively, was supplied with hot reducing gas (total gas flow rate of 8.0 NL/min). An off-gas analysis was connected to the furnace to determine the reduction progress. A boat, that was inserted into the reactor, contained 45 g and 15 g of iron ore pellets and nut coke, respectively (pellets/nut coke mass ratio 3:1). Pellets were sized 10 - 12 mm whereas nut coke was 12 - 14 mm. The reducing gas supplied was composed of 30 % CO and 70 % N2. Under non-isothermal reduction conditions, a slightly bigger reactor was used as shown in fig. 5b. The weight of pellets and coke amounted to 600 g and 200 g, respectively, whereby two differently sized types of nut coke were inserted: 12 - 14 mm and 35 - 40 mm. In order to represent reducing conditions in a blast furnace, the inflow temperature and composition was adjusted as listed in the following table 1.

Table 1: Temperatures and compositions for non-isothermal reduction conditions

Temperature [K]	Heating rate [K/min]	Gas composition [Vol. %]
298 - 673	8	100 % N2
673 - 1173	8	26 % CO, 14 % CO2, 60 % N2
1173 - 1273	2	30 % CO, 70 % N2
1273 - 1523	5	30 % CO, 70 % N2

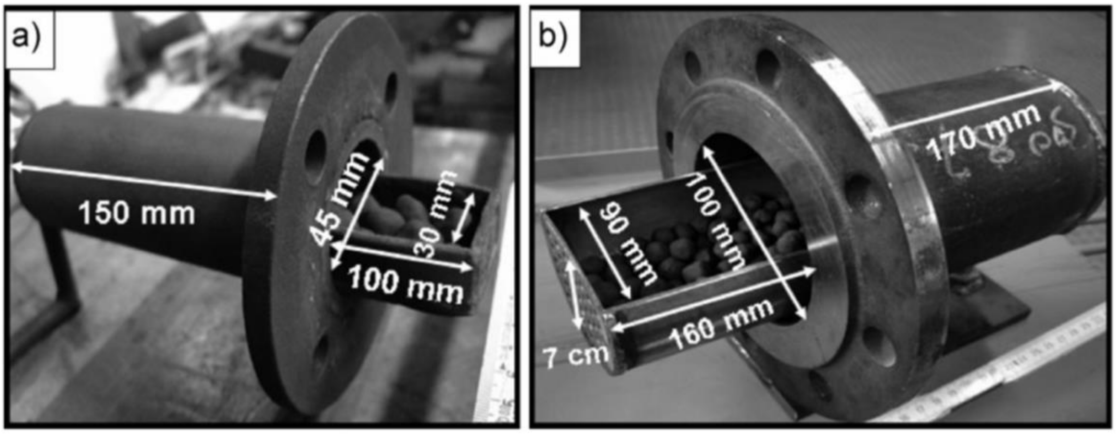


Figure 2: Reactor for isothermal (a) and non-isothermal (b) reduction of iron bearing materials

For more details of the experiments carried out the reader is referred to Moussa et al. [10]

2. NUMERICAL APPROACH

The numerical approach taken for the current investigations is a coupling between discrete and continuous simulation techniques. Thus, the fluid phase in the void space between the particles representing the reducing gas is treated by a Computational Fluid Dynamics (CFD) approach, while the solid phase as ore particles is resolved by a discrete approach similar to the Discrete Element Method (DEM). However, as an extension to the classical Discrete Element Method, that predicts motion of particles in space, the thermodynamic state of the each particle during ore reduction is determined.

Hence, DEM is extended by the thermodynamic state for each particle and its coupling via heat and mass transfer to the gas phase, and therefore, is referred to as the Extended Discrete Element Method (XDEM). Further details are found in the following references [11-16].

3. RESULTS

Mousa [17] conducted non-isothermal experiments with iron ore pellets under simulated blast furnace conditions. A small bed of pellets and mixtures of pellets and nut coke is placed within a closed steel tube inside a muffle furnace and exposed to a CO/N_2 reducing gas mixture. The chemical analysis of pellets is given in table 3 and the solid phase material properties are given in table 4.

Table 3: Chemical analysis of hematite pellet [17]

Element	Fe	FeO	SiO2	CaO	inerts
wt%	62	0.02	4.75	0.99	rest

Table 4: Properties of iron ore and nut coke for reduction experiments by Mousa [17]. Note: Densities are given as intrinsic densities of pure substances. Solid Phase Properties

Solid Phase Properties		
D_{pellet}	(m)	10-12 10^{-3}
ε_{pellet}	(-)	0.3
ρ_{pellet}	(kg/m^3)	3364
D_{coke}	(m)	12-14 10^{-3}
ε_{coke}	(-)	0.42
ρ_{coke}	(kg/m^3)	1243

The time varying inlet conditions for the non-isothermal scenario simulating conditions inside a blast furnace are given in table 5.

Table 5: Time varying parameters of reducing gas for non-isothermal iron ore reduction experiments by Mousa [17]

	Heating rate [K/min]	Temperature [K]	p_{CO} [bar]	p_{CO2} [bar]	p_{N2} [bar]	Volume flow [Nl/min]
phase 1	8	298 - 1173	0.26	0.14	0.6	8
phase 2	2	1173 - 1273	0.3	0.0	0.7	8
phase 3	5	1173 - 1273	0.3	0.0	0.7	8

For all experiments the integral reduction degree of the bed was evaluated through off-gas chemical analysis. For the gasification of coke by CO to form CO_2 kinetic parameters are taken from Babich et al. [18].

Fig. 3 compares the results of the model with the experimentally determined reduction curves for non-isothermal reduction of a bed of pellets (red) and a bed consisting of pellets and nut coke (green). The model correctly predicts the integral reduction behaviour of the bed in both cases very well. Hence, the current approach prove its excellent predictive capabilities by providing both very detailed results of the packed bed processes down to a particle level and also fulfilling integral balances for mass and energy.

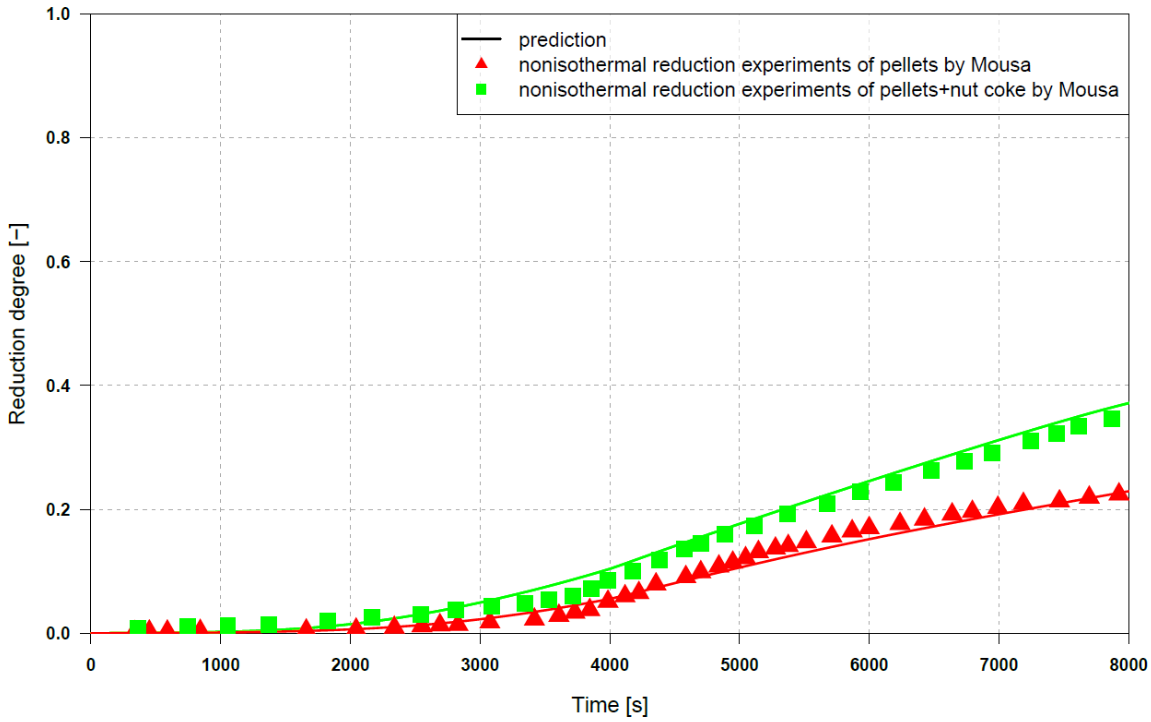


Figure 3: Comparison of model prediction and experiments for non-isothermal reduction of a bed of pellets (red triangles) and pellets mixed with nut coke (green squares).

Fig. 4 shows the particle bed and a longitudinal cut through the gas phase.

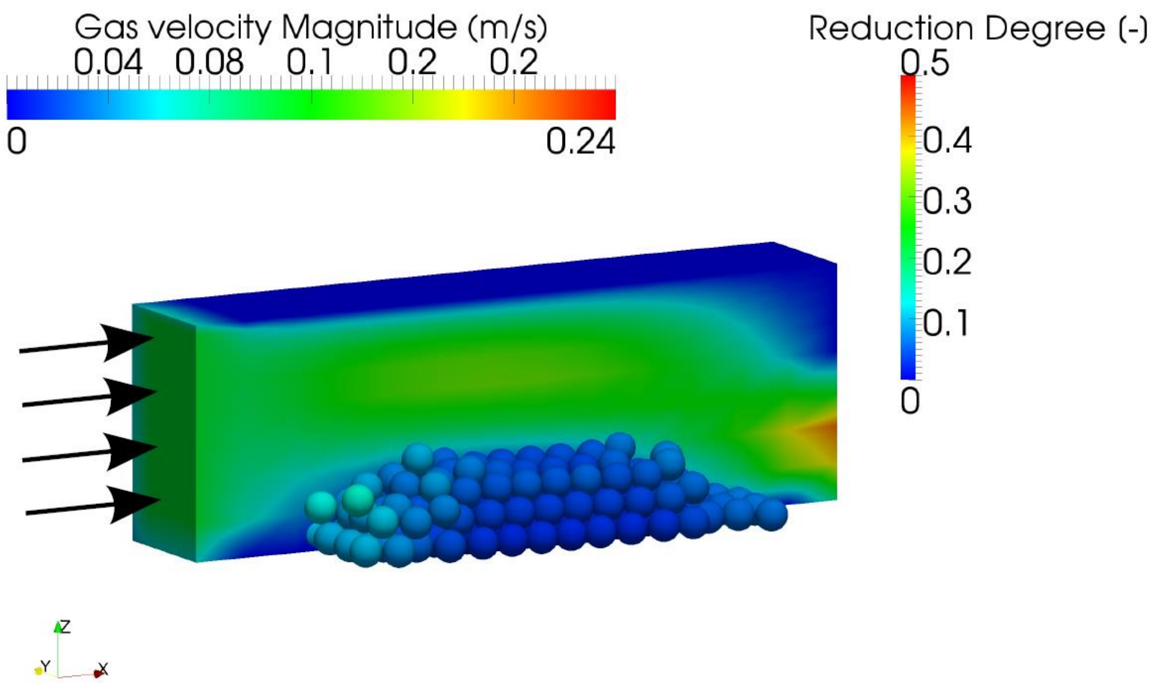
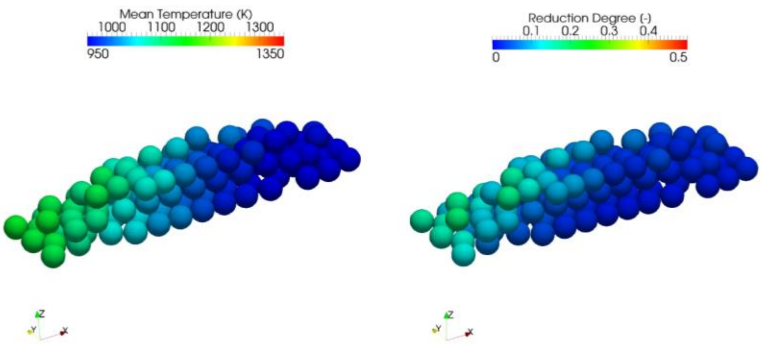


Figure 4: Gas phase velocity and particle reduction degree at t=4000 s

The reactor's gas inlet is located at the left of the bed and the gas streams in positive x-direction and the tubular reactor is still not completely filled with pellets due to the experimental setup. This again creates a free surface of particles and a free board above the

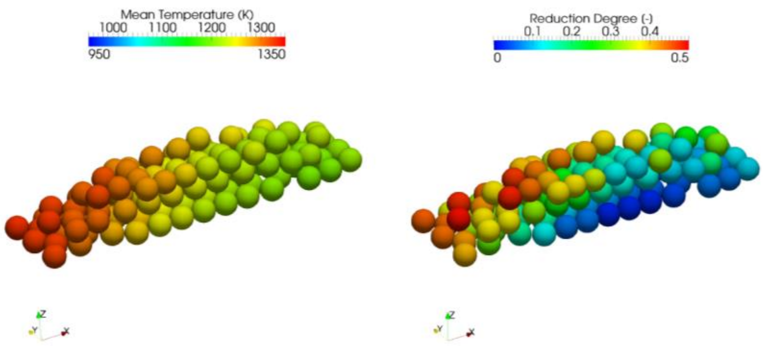
packed bed with significantly reduced flow resistance. Therefore, a considerable amount of the reducing gas is not forced to pass through the bed but instead streams over it. As a result particles close to the inlet are exposed to the streaming conditions specified at the inlet whereas particles located towards the outlet of the reactor are exposed to a gas stream of significantly lower velocity. Hence, the gas velocity influences the convective heat and mass transfer between particle surface and gas as the first reason for the heterogeneity inside the bed is limited mass transfer between particles and gas.
A second reason for the heterogeneity of the process is the spatial gradient in reducing gas potential: The product gas of the reduction which is emitted by particles close to the reactor inlet has a much lower reduction potential than the inlet gas. Thus downstream particles additionally suffer from the lower reduction potential of the gas since diffusion perpendicular to the gas stream is insufficient to compensate this effect.

Figs. 5 and 6 depict longitudinal cuts half way through the pellet bed in y-direction for two different points in time.



(a) Mean particle temperature (b) Reduction degree

Figure 5: Longitudinal cut through the bed of pellets at t=4000 s



(a) Mean particle temperature (b) Reduction degree

Figure 6: Longitudinal cut through the bed of pellets at t=8000 s

Particles are coloured according to their mean temperature and the reduction degree. A considerable gradient in reduction degree is observed. In addition to heterogeneity in mass transfer significant gradients in particle temperature further amplify the heterogeneity inside the bed. Firstly, a higher temperatures increases the reaction rate (Arrhenius equation). Secondly, as the reduction temperature is increased the equilibrium partial pressure ratio $p_{CO,eq}/(p_{CO,eq} + p_{CO2,eq})$ for the reduction of magnetite to wustite is decreased. The latter implies a higher reducing potential of the gas at higher temperatures and thus more favourable reducing conditions which in turn increases the reaction rate.

4. SUMMARY

The current studied presented a coupled DEM-CFD approach to predict reduction of iron ore in a packed bed. These efforts were enhanced and validated by experiments carried out in laboratory-scale reactor. Experiments yielded the integral reduction behaviour of the reactor derived by measuring the off-gas composition. Complementary predictions of the packed bed processes including the thermodynamic state of both iron ore particles and the surrounding flow agreed well with experimental data and allowed a detailed analysis of the underlying physics.

Results highlight that the gas and the solid phase are highly coupled in space and time during the process of indirect reduction. Axial gradients in temperature and composition form due to the heat and mass transfer between the packed bed and the streaming gas. Energy released or consumed by the indirect reduction provides an opposing trend to the gradients formed from the hot gas stream, thus reducing axial gradients within the bed and the gas phase. This observation is contrary to the results from the isothermal reduction experiments but shows the immense influence of time-varying boundary conditions on the process. Furthermore, the results indicate the mechanisms involved during the formation of the thermal reserve zones inside the blast furnace shaft due to the complex interaction of convective heat and mass transfer in conjunction with energy consumption and release by the reducing reactions.

REFERENCES

[1] S. Natsui, S. Ueda, F. Zhen-yun, J. Kano, H. Nogami, R. Inoue, T. Ariyama, Development of advanced blast furnace simulation model based on discrete element method, in: Proceedings of the 5th International Congress on the Science and Technology of Ironmaking (ICSTI), 2009.

[2] H. P. Zhu, Z. Y. Zhou, R. Y. Yanga, A. B. Yu, Discrete particle simulation of particulate systems: Theoretical developments, Chemical Engineering Science 62 (13) (2007) 3378-3396.

[3] Z. 540 Zhou, H. Zhu, A. Yu, P. Zulli, Numerical investigation of the transient multiphase flow in an ironmaking blast furnace, ISIJ International 50 (4) (2010) 515-523.

[4] A. Adema, Y. Yang, R. Boom, Coupled DEM-CFD modelling of the ironmaking blast furnace, in: Proceedings of the 5th International Congress on the Science and Technology of Ironmaking (ICSTI), 2009.

[5] A. Adema, Y. Yang, R. Boom, Discrete element method-computational fluid dynamics simulation of the materials flow in an iron-making blast furnace, ISIJ International 50 (2010) 954-961.

[6] Z. Zhou, H. Zhu, A. Yu, B. Wright, D. Pinson, P. Zulli, Discrete particle simulation of solid flow in a model blast furnace, ISIJ International 45 (12) (2005) 1828-1837.

[7] T. Nouchi, T. Sato, M. Sato, K. Takeda, T. Ariyama, Stress field and solid flow analysis of coke packed bed in blast furnace based on DEM, ISIJ International 45 (10) (2005) 1426-1431.

[8] Z. Y. Zhou, D. Pinson, H. P. Zhu, A. B. Zhu, B. Wright, P. Zulli, Discrete particle simulation of gas-solid flow in a blast furnace, Computers and Chemical Engineering 32 (2008) 1760-1772.
[9] H. Kurosawa, S. Matsuhashi, S. Natsui, T. Kon, S. Ueda, R. Inoue, T. Ariyama, Dem-Cfd model considering softening behaviour of ore particles in cohesive zone and gas flow analysis at low coke rate in blast furnace, ISIJ International 52 (2012) 1010-1017.
[10] E. Mousa, D. Senk, A. Babich, Reduction of pellets-nut coke mixture under simulating blast furnace conditions, Steel Research Int. 81 (9) (2010) 706-715.
[11] B Peters, The extended discrete element method (XDEM) for multi-physics applications Scholarly Journal of Engineering Research 2 (1), 1-20
[12] A. Mahmoudi, M. Markovic, B. Peters, G. Brem, An experimental and numerical study of wood combustion in a fixed bed using Euler–Lagrange approach (XDEM), Fuel 150, 573-582
[13] M. Michael, F. Vogel, B. Peters, DEM–FEM coupling simulations of the interactions between a tire tread and granular terrain, Computer Methods in Applied Mechanics and Engineering 289, 227-248
[14] E. Copertaro, P. Chiariotti, A. Donoso, N. Paone, B. Peters, G. Revel, A discrete-continuous approach to describe $CaCO_3$ decarbonation in non-steady thermal conditions Powder Technology 275, 131-138
[15] R. Navakas, A. Džiugys, B. Peters, A community-detection based approach to identification of inhomogeneities in granular matter, Physica A: Statistical Mechanics and its Applications 407, 312-331
[16] A. Mahmoudi, F. Hoffmann, B. Peters, Application of XDEM as a novel approach to predict drying of a packed bed, International Journal of Thermal Sciences 75, 65-75
[17] E. Mousa, Reduction of iron ore burden materials mixed with nut coke under simulated blast furnace conditions, Ph.D. thesis, RWTH Aachen (2011).
[18] U. Janhsen, A. Günbati, C. Sautner, E. Faraji, J. O. Wikstöm, T. Hilding, 635 J. Eriksson, O. Kerkkonen, P. Arendt, A. Babich, Ore agglomeration and ironmaking - changes in the microstructure of coke while passing the blast furnace with respect to the quality of the charged coke and the behaviour of nut coke in the blast furnace, Tech. Rep. EU 22439 EN, European Commission, Luxembourg (2007).