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Fast long-term simulations of hot, reacting, moving particle beds with a melting zone

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ABSTRACT

Hot, reacting, moving particle beds as found in blast furnaces comprise a wide range of spatial and temporal scales. We present an approach that combines models for granular motion, interfacial mass, momentum and heat transfer as well as heterogeneous multi-step reactions efficiently. Although they correspond to vastly different scales, long-term investigations built upon data from discrete-element simulations become feasible. The strategy was first applied to 0.5 kg hematite reducing to iron under a CO atmosphere, where the most relevant reaction parameters were obtained from optimization towards experimental data. Then, we studied a 3D, full-scale blast furnace and its approach towards the thermo-chemical steady state over the course of 20 h.

The framework may be easily extended, which will allow for realistic simulations of moving particle beds over process-relevant durations. It has the potential to create digital twins so that different reactor types can be optimized and novel designs explored.

1. Introduction

Moving bed reactors can be found in various industrial applications such as chemical looping combustion (Kim et al., 2013; Shao et al., 2020; Wang et al., 2018), pyrolysis (Aylón et al., 2010; Liang et al., 2015), heat recovery (Feng et al., 2019; Qiu et al., 2020) and iron ore reduction in shaft (Shams and Moazeni, 2015; Hamadeh et al., 2018) and blast furnaces (BFs) (Kuang et al., 2018). They exhibit intense heat transfer and homogeneous as well as heterogeneous chemical reactions. In the present study, we focus on the BF process as a highly relevant but challenging example for this type of system. Despite its large CO₂ footprint and the need (Vogl et al., 2021) to replace it with alternatives with lower greenhouse emissions like hydrogen-based direct iron ore reduction (HYFOR) (Eisl et al., 2022) or H₂ plasma smelting reduction (HPSR) (Plaul et al., 2005; Sabat et al., 2014), the BF will continue to dominate global iron making for decades. Hence, it is essential to investigate possible optimizations, which can be explored most conveniently and risk-free with numerical simulations. However, developing a computational model that is both simple enough to allow for a feasible numerical treatment and extensive enough to contain the relevant physics and chemistry has proven extremely difficult because of its multiphysics and multi-scale nature. Fig. 1 illustrates the interplay between the mechanical (dense granular matter, particle-fluid interaction), thermal (intra and interphase heat transfer, phase change) and chemical (coke combustion, ore reduction etc.) problems. While most studies focused on one or a few selected phenomena like flow of liquid iron and slag in the hearth (Liu et al., 2020; Nijssen et al., 2022a,b; Vångö et al., 2018), raceway (RW) dynamics (Cui et al., 2020; Santana et al., 2019; Wei et al., 2019) or large-scale burden descent (Li et al., 2019; Wei et al., 2023; Yang et al., 2015; Yu and Saxén, 2012), a comprehensive model should incorporate all of these aspects. In this regard, the vastly different spatial and temporal scales present in the BF pose a massive obstacle. The huge number of small grains in a full-scale BF exceeds currently available computational resources by far, and grain collisions in the RWs last microseconds whereas large-scale burden motion and ore reduction take several hours. These issues led to the use of continuum models (Chu et al., 2006; Yagi, 1993) and massively coarse-grained parcel representations (Bierwisch et al., 2009; Sakai and Koshizuka, 2009).

But even then, the temporal scope was not sufficient for transient long-term studies. Hence, further model variants were devised to reach process-relevant durations, e.g. by artificially increasing the speed of the studied phenomena (Hou et al., 2020) or by exploiting system symmetries to reduce the computational effort (Yu and Shen, 2022). Some investigations assumed a steady state of selected, numerically expensive properties like the particle motion in the moving bed (Bluhm-Drenhaus et al., 2010; Krause et al., 2015, 2017) or in other applications involving

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Nomenclature

Greek lett	ters
α	volume fraction
δ_{n}	normal overlap between particles m
e	internal energy per mass $\dots m^2/s^2$
ε	emissivity
κ _n	normal tracer repulsion rate 1/s
λ_{cg}	coarse-graining factor
μ	viscosity kg/(m s)
ρ	mass density kg/m ³
ρ	mass density source term kg/(m ³ s)
σ	stress tensor kg/(m s ²)
Latin of s	ymbols
A_l	reaction resistance between layers l and $l + 1 \dots m/s$
B_l	diffusion resistance through layer <i>l</i> m/s
С	heat capacity $m^2/(s^2 K)$
d	diameter m
D	diffusivity m^2/s
е	particle energy per mass m^2/s^2
$E^{(a)}$	activation energy kg m^2/s^2
f	force density $kg/(m^2 s^2)$
\tilde{f}	effective volume factor
F	force kg m/s ²
F	diffusion resistance through stagnant fluid layer m/s
g	specific gravity m/s ²
$k^{(0)}$	reaction rate prefactor m/s
$k^{(\text{eff})}$	thermal conductivity kg m/(s ³ K)
Κ	kinetic energy per mass m^2/s^2
т	mass kg



Fig. 1. The multiphysics problem of BF modeling. Besides various other coupling mechanisms, mechanical, thermal and chemical aspects are interconnected via transport of heat and species, temperature-dependent fluid and particle properties, the interplay of CZ location and the granular velocity field, and the relationship between RW size and chemical efficiency.

dense granular matter (Bednarek et al., 2019; Haydar et al., 2023). In these studies, high-fidelity particle data from detailed simulations were employed to determine the steady state once and continue with faster computations.

In our previous publications (Lichtenegger, 2020; Lichtenegger and Pirker, 2020), we developed the foundations of an akin, deliberately simple approach which we refer to as *pseudo-steady CFD-DEM* that comes with two major advantages: (i) It is flexible with regard to the model that generates the underlying data for the fast simulations. As an alternative to expensive discrete-particle simulations, one could also carry out cheaper continuum calculations if one accepts the higher degree of phenomenology. (ii) It requires relatively little input data. Short, coarse-grained, full CFD-DEM simulations suffice to obtain the time-

mass change per time k	cg/s
pressure kg/(m	s ²)
energy density source term kg/(m	s ³)
heat flux density kg	g∕s ³
energy source term kg m ²	$^{2}/s^{3}$
position	m
velocity field	m/s
particle velocity	m/s
volume	m^3
temperature	. K
molar concentration mol	/m ³
molar change per time mo	ol/s
mass fraction	

Subscripts/Superscripts

m p q Q r u v V T x X Y

eq	equilibrium
ext	external
f	fluid
RCP	random close packing
р	particle
prod	product
react	reactant
List of abb	reviations
CFD	computational fluid dynamics
DEM	discrete element method
Nu	Nusselt number
Sc	Schmidt number
Sh	Sherwood number

averaged particle volume fraction and velocity fields. In a subsequent step, solid grains are replaced with non-interacting tracers which follow the previously computed field lines.

Without the enormous costs of continuing DEM calculations, it has become feasible to simulate the gas flow through the moving bed including interfacial heat transfer over the course of many hours (Lichtenegger, 2020). For other moving bed processes, this strategy might already allow to determine the thermal steady state. However, the BF is more complicated due to the presence of the cohesive zone (CZ) where solid iron (ore) melts and trickles down through the coke bed below. The location and shape of the CZ are a consequence of the particle temperature field, which develops over longer durations, but have a strong impact on the granular motion. Hence, one has to iteratively update the particle velocity field with the evolution of the CZ by interrupting the fast simulations with intermittent full CFD-DEM runs.

So far, we have only taken first steps with this approach and tested it with a hot BF slot model disregarding any chemistry (Lichtenegger and Pirker, 2020). The present work significantly extends our previous investigations in two regards: (i) We have included the most important chemical reactions into our framework. In particular, we employed an ore reduction model, for which we obtained the optimal chemical reaction parameters from a calibration experiment. Furthermore, we modeled coke combustion and the Boudouard reaction. (ii) It is well known that pseudo-2D simulations cannot completely replace 3D calculations (Bambauer et al., 2018). Therefore, we carried out three-dimensional, full-scale simulations of an industrial furnace. Taken together, these two points constitute a major step towards a realistic, multiphysics and multi-scale BF model that can be studied over process-relevant time scales and may be easily extended to account for additional phenomena. Ultimately, we envision digital twins which can massively improve the exploration and optimization of novel reactor designs. In the light of the approaching decarbonisation of various

branches of industry, such computational tools may provide invaluable help.

The paper is organized as follows: In Sec. 2, we recapitulate the governing equations for fluid flow and particle dynamics including thermal and chemical aspects. Furthermore, we explain the algorithmic strategy that allows us to decouple the contact-mechanical from the process time scale. Details on the simulated cases for chemical parameter calibration and for the full-scale BF study are provided in Sec. 3 with corresponding results shown in Sec. 4. Finally, we critically discuss our findings and point out directions for future work in Sec. 5.

2. Theoretical background

2.1. Fluid equations

A compressible, reacting fluid mixture in the presence of a secondary particle phase is described by the well-known filtered Navier-Stokes equations (Anderson and Jackson, 1967)

$$\frac{\partial}{\partial t}\alpha_{\rm f}\rho_{\rm f} + \nabla \cdot \alpha_{\rm f}\rho_{\rm f}\boldsymbol{u}_{\rm f} = \dot{\rho}_{\rm p-f} \tag{1}$$

$$\frac{\partial}{\partial t}\alpha_{\rm f}\rho_{\rm f}Y_i + \nabla \cdot \alpha_{\rm f}\rho_{\rm f}u_{\rm f}Y_i = \nabla \cdot \alpha_{\rm f}\rho_{\rm f}D_{\rm f}\nabla Y_i + \sum_{j\neq i}\dot{\rho}_{i,j} + \dot{\rho}_{{\rm p-f},i}$$
(2)

$$\frac{\partial}{\partial t} \alpha_{\rm f} \rho_{\rm f} \boldsymbol{u}_{\rm f} + \nabla \cdot \alpha_{\rm f} \rho_{\rm f} \boldsymbol{u}_{\rm f} \boldsymbol{u}_{\rm f} = \alpha_{\rm f} \nabla \cdot \boldsymbol{\sigma}_{\rm f} + \boldsymbol{f}_{\rm drag} + \boldsymbol{f}_{\rm ext}$$
(3)
$$\frac{\partial}{\partial t} \alpha_{\rm f} \rho_{\rm f} (\boldsymbol{\epsilon}_{\rm f} + \boldsymbol{K}_{\rm f}) + \nabla \cdot \alpha_{\rm f} \rho_{\rm f} \boldsymbol{u}_{\rm f} (\boldsymbol{\epsilon}_{\rm f} + \boldsymbol{K}_{\rm f}) =$$

$$- \nabla \cdot \alpha_{\rm f} p_{\rm f} \boldsymbol{u}_{\rm f} + \nabla \cdot \alpha_{\rm f} k_{\rm f}^{\rm (eff)} \nabla T_{\rm f} + \dot{q}_{\rm p-f} + \dot{q}_{\rm f}^{\rm (chem)} + \dot{q}_{\rm p-f}^{\rm (chem)}$$
(4)

accounting for transport of mixture and species-specific mass, momentum and energy.

Interfacial mass transfer per volume, $\dot{\rho}_{\text{p-f}i} = \sum_i \dot{\rho}_{\text{p-f}i}$, contains contributions from all heterogeneous reactions involving each species *i*. These are described with local mass fractions Y_i that are convected with the flow and diffuse with diffusivities D_{f} . The latter may be connected to the viscosity via the Schmidt number Sc $\equiv \mu_{\text{f}}/(\rho_{\text{f}}D_{\text{f}})$. Besides sources/sinks $\dot{\rho}_{\text{p-f},i}$ due to reactions with the granular phase, reactions within the fluid phase between species *i* and *j* affect the local species mass in terms of $\dot{\rho}_{i,j}$.

As a consequence of the filtering operation, source terms in the continuum equations are obtained from particle-based sources via a filter function g(r) which has a range comparable to the local mesh resolution. Hence, the mass of species *i* lost/gained per unit time at location r,

$$\dot{\rho}_{\text{p-f},i} = \sum_{k} g(\boldsymbol{r} - \boldsymbol{r}_{k}) \sum_{\boldsymbol{R}(i)} \dot{\boldsymbol{m}}_{k,\boldsymbol{R}(i)}$$
(5)

is obtained from contributions $\dot{m}_{k,R(i)}$ of all nearby grains *k* that participate in any reaction R(i) that involves *i* (cf. Eqs. (37) and (38)).

Similarly, momentum transfer between fluid and discrete phase, which in packed beds is mainly caused by the drag force density

$$\boldsymbol{f}_{\rm drag}(\boldsymbol{r}) \equiv -\sum_{k} g(\boldsymbol{r} - \boldsymbol{r}_{k}) \boldsymbol{F}_{k}^{\rm (drag)},\tag{6}$$

is obtained as a weighted sum of the drag forces $F_k^{(drag)}$ acting on all particles *k* located within the radius of the filter function g(r).

The interaction forces within the fluid phase are described by the stress tensor σ_f which often takes the Newtonian form

$$\boldsymbol{\sigma}_{\mathrm{f}} \equiv -p_{\mathrm{f}}\boldsymbol{I} + \mu_{\mathrm{f}} \Big(\nabla \boldsymbol{u}_{\mathrm{f}} + \big(\nabla \boldsymbol{u}_{\mathrm{f}} \big)^{\dagger} \Big) - \frac{2}{3} \mu_{\mathrm{f}} \boldsymbol{I} \nabla \cdot \boldsymbol{u}_{\mathrm{f}}.$$
(7)

In the present study, we accounted for a temperature-dependent viscosity $\mu_f(T_f)$ with the Sutherland equation (Sutherland, 1893). Further source terms in the momentum equation of external origin, such as weight, are contained in f_{ext} .

The fluid energy density consists of an internal contribution $\epsilon_{\rm f}$ and the kinetic energy density $K_{\rm f} \equiv u_{\rm f}^2/2$. Besides convective transport and pressure work, heat conduction with an effective thermal conductivity $k_{\rm f}^{\rm (eff)}$ (Syamlal and Gidaspow, 1985) plays an important role. The relationship between internal energy and temperature

$$\epsilon_{\rm f}(T) = \int_{T_0}^{T} dT' C_{\rm f}(T') + \epsilon_{\rm f}^{(0)},\tag{8}$$

is established by the specific heat capacity $C_{\rm f}(T_{\rm f})$, which can be conveniently approximated as a polynomial with coefficients from the JANAF tables of thermodynamics (Burcat and Ruscic, 2005).

Heat transfer

$$\dot{q}_{\rm p-f} \equiv -\sum_{k} g(\boldsymbol{r} - \boldsymbol{r}_{k}) \dot{\mathcal{Q}}_{k}^{\rm (p-f)} = -\sum_{k} g(\boldsymbol{r} - \boldsymbol{r}_{k}) \pi k_{\rm f} d_{k} \operatorname{Nu}(T_{\rm f} - T_{k})$$
(9)

between fluid and particle phase is driven by the temperature difference between each grain and the surrounding fluid. Although manyfold validated correlations for the Nusselt number Nu, such as that by Ranz and Marshall (Ranz and Marshall, 1952) exist, we remark that for large-scale flows in industrial plants, it seems necessary to significantly reduce the value of Nu to find agreement with measured data. Some authors have ascribed this discrepancy to the presence of dust in such systems (Slaby et al., 2006; Häring et al., 2010). Until further, more quantitative investigations shed some light on this phenomenon, the choice of Nu will remain a potential source of uncertainty for blast and shaft furnace simulations.

Furthermore, heat can be released or absorbed by chemical reactions within the fluid phase or between fluid and particles, leading to sources/sinks

$$\dot{q}_{\rm p-f}^{\rm (chem)} = \sum_{k} g(\mathbf{r} - \mathbf{r}_{k}) \sum_{R(i)} \dot{Q}_{k,R(i)},$$
 (10)

where $\dot{Q}_{k,R(i)}$ is the amount of heat released or absorbed at grain *k* due to its reaction R(i) with the fluid species *i*.

2.2. Particle equations

For dense, granular systems where long-lasting particle-particle contacts play a dominant role, DEM (Cundall and Strack, 1979) provides a convenient description. The properties of each grain, such as mass m_i , position r_i , velocity v_i , temperature T_i and composition are stored and subject to computations. In the following subsections, we summarize the various models for mechanical, thermal and chemical properties.

2.2.1. Mechanical model

The particle momentum and consequently trajectory are governed by Newton's second law

$$\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{r}_i = \boldsymbol{v}_i \tag{11}$$

$$\frac{d}{dt}m_i v_i = F_i^{(p-p)} + F_i^{(p-f)} + F_i^{(ext)},$$
(12)

where forces arise from particle-particle and particle-fluid interactions as well as from external sources like gravity $F_i^{(ext)} = m_i \mathbf{g}$. Forces between solid grains are often approximated with pairwise spring-dashpot models

$$F_i^{(p-p)} = \sum_{i \neq j} F_{i,j}$$
(13)

based on the deformation of elastic bodies (Hertz, 1882; Johnson, 1985). Both normal and tangential components of the force consist of elastic and of dissipative contributions. The tangential part is limited by Coulomb's law of friction. For more details, we refer the interested reader either to the original literature (Cundall and Strack, 1979) or to review articles (Guo and Curtis, 2015; Blais et al., 2019).

The main contributions to the particle-fluid interaction

$$F_i^{(p-f)} \approx -V_i \nabla p_f + F_i^{(drag)} \tag{14}$$

stem from pressure gradients and drag which we approximate with the Beetstra correlation (Beetstra et al., 2007) to account for polydispersity. Other forces such as lift were neglected in the present study because they play a minor role in packed beds but could be easily included if necessary.

2.2.2. Thermal model

The internal energy $E_i = m_i e_i$ of each particle changes because of (i) heat transfer with the surrounding fluid $\dot{Q}_i^{(p-f)} = \pi k_f d_i \text{Nu}(T_f - T_i)$ and with other particles $\dot{Q}_i^{(\text{p-p})} = \sum_{i \neq j} \dot{Q}_{i,j}$ and (ii) chemical sources $\dot{Q}_i^{(\text{chem})}$ (cf. Sec. 2.2.3) according to

$$\frac{d}{dt}m_{i}e_{i} = \dot{Q}_{i}^{(p-f)} + \dot{Q}_{i}^{(p-p)} + \dot{Q}_{i}^{(chem)},$$
(15)

which translates into temperature changes because of $\partial e_i/\partial T_i = C_i(T_i)$. Intergranular heat transfer $\dot{Q}_{i,j} = \dot{Q}_{i,j}^{(\text{cond})} + \dot{Q}_{i,j}^{(\text{rad})}$ takes place via conduction through solid contacts and via radiation. The former contribution depends on the material thermal conductivities k, the contact radius a and the temperature difference in terms of (Vargas and Mc-Carthy, 2001)

$$\dot{Q}_{i,j}^{(\text{cond})} = 4 \frac{k_i k_j}{k_i + k_j} a_{ij} (T_j - T_i).$$
(16)

Radiative heat transfer, which becomes increasingly dominant for high temperatures, is significantly more complicated. Wu et al. (2020) compared different levels of detailedness to model radiation in densely packed beds and suggested to approximate the grey-body radiation between two grains with emissivity ε as

$$\dot{Q}_{i,j}^{(\text{rad})} = \frac{1}{2/\epsilon - 1} \sigma A_i X_{ij} \left(T_j^4 - T_i^4 \right), \tag{17}$$

where σ is the Stefan-Boltzmann constant, A_i the surface area and X_{ii} the view factor between the two particles. Strategies to compute X_{ii} as discussed by Peng et al. (2020) have been recently extended by the usage of deep neural networks (Tausendschön and Radl, 2021; Wu et al., 2022).

If it is necessary to use a numerically cheaper alternative than the approaches above, one may choose to ignore the microstructure of the bed and employ empirical correlations to introduce a Eulerian effective thermal conductivity $k_{\text{cond}}^{\text{(eff)}} + k_{\text{rad}}^{\text{(eff)}}$, which gives rise to a local heat flux

$$\boldsymbol{q}_{\mathrm{p-p}} = -\alpha_{\mathrm{p}} \left(k_{\mathrm{cond}}^{\mathrm{(eff)}} + k_{\mathrm{rad}}^{\mathrm{(eff)}} \right) \nabla T_{\mathrm{p}}$$
(18)

in opposite direction of the gradient of the particle temperature field. One can then approximate the heat gain or loss of each grain by integrating the flux over an enclosing surface, which gives

$$\dot{Q}_{i}^{(\mathbf{p}-\mathbf{p})} \approx \frac{V_{i}}{\alpha_{\mathbf{p}}} \nabla \cdot \boldsymbol{q}_{\mathbf{p}-\mathbf{p}}.$$
(19)

Among the various empirical expressions for $k_{cond}^{(eff)}$ and $k_{rad}^{(eff)}$ (Carson et al., 2005; van Antwerpen et al., 2010; Qian et al., 2018), we employed

$$k_{\text{cond}}^{(\text{eff})} \rightarrow \frac{3\alpha_{\text{p}} - 1}{2\alpha_{\text{p}}} k_{\text{p}} \quad \text{for } k_{\text{p}} \gg k_{\text{f}}$$
 (20)

derived from the effective medium theory (Landauer, 1952; Kirkpatrick, 1973) and the Breitbach-Barthels correlation (Breitbach and Barthels, 1980)

$$k_{\rm rad}^{\rm (eff)} = 4F_{\rm E}\sigma d_{\rm p}T_{\rm p}^3 \tag{21}$$

in terms of the radiation exchange factor

$$F_{\rm E} = \left(1 - \sqrt{\alpha_{\rm p}}\right)\left(1 - \alpha_{\rm p}\right) + \frac{\sqrt{\alpha_{\rm p}}}{2/\epsilon - 1} \frac{B + 1}{B} \left(1 + \frac{1}{(2/\epsilon - 1)\Lambda}\right)^{-1} \tag{22}$$

$$B = 1.25 \left(\alpha_{\rm p} / (1 - \alpha_{\rm p}) \right)^{10/9} \tag{23}$$

$$\Lambda = \frac{k_{\rm p}}{4d_{\rm p}\sigma T_{\rm p}^3} \tag{24}$$

for radiative conductivity.

2.2.3. Chemical model

In the present work, we considered the following reactions between granular and fluid phase. Coke reacts with O₂ in the high-temperature RW regions to form CO and CO₂ and furthermore with CO₂ to regenerate CO (reverse Boudouard reaction),

$$(1+\gamma)C + \frac{2+\gamma}{2}O_2 \longrightarrow \gamma CO + CO_2$$
 (25)

$$C + CO_2 \longrightarrow 2CO,$$
 (26)

where γ is the CO-CO₂ ratio (Arthur, 1951). The CO₂ in Eq. (26) stems from the reactions with coke Eq. (25) and with iron ore, where hematite Fe₂O₃ is first reduced to magnetite Fe₃O₄, then wustite FeO and finally iron Fe,

$$3Fe_2O_3 + CO \longrightarrow 2Fe_3O_4 + CO_2$$
 (27)

$$Fe_3O_4 + CO \longrightarrow 3FeO + CO_2$$
 (28)

$$FeO + CO \longrightarrow Fe + CO_2.$$
 (29)

Instead of CO, H₂ or a mixture of both can act as reducing agent, but the main reduction work is still done by CO.

Reactions between a particle and reactants in the surrounding fluid phase will either consume the outer layers of the grain (shrinking particle) or transform them into a different material (shrinking core).

Multi-layer shrinking core model For successive reaction steps such as ore reduction Eqs. (27) - (29), multiple layers with an unreacted, shrinking core will develop (Valipour, 2009; Peters and Hoffmann, 2016; Kinaci et al., 2020; Schneiderbauer et al., 2020). Further examples comprise fluorination of uranium dioxide (Homma et al., 2005), calcination (Amiri et al., 2015), combustion of organic additives in clay bricks (Wesenauer et al., 2020) and hydrate-based CO₂ capture (Dashti et al., 2019).

Fig. 2 illustrates a simple description of such a multi-step reaction in analogy to an electrical resistor network. The differences between the concentration of gaseous reactant $x_{\text{react}}^{(f)}$ in the vicinity of the grain and the equilibrium concentrations x_{eq} of the various reactions at the layer interfaces act as the driving set of the various reactions at the layer interfaces act as the driving potentials to which diffusion through a stagnant fluid film and through the various material layers into the grain as well as finite reaction rates pose resistances. The hypothetical current through this network resembles the amount of reactant $\dot{X}_{react}^{(f)}$ consumed per unit time.

The reaction resistance between layers l and l+1 (where numbering starts from the outermost layer) with their interface located at r_1 is given by

$$A_{l} \equiv \left[k_{l} \left(\frac{r_{l}}{r_{p}}\right)^{2} \frac{1 + K_{l}^{(eq)}}{K_{l}^{(eq)}}\right]^{-1},$$
(30)

where

$$k_l = k_l^{(0)} e^{-E_l^{(a)}/(RT)}$$
(31)

is the reaction rate that depends on temperature via the activation energy $E_l^{(a)}$. The reaction is limited by the relative surface $(r_l/r_p)^2$ and the value of the equilibrium constant $K_i^{(eq)}$.

Diffusion through layer *l* poses another type of resistance

$$B_l \equiv \frac{r_l - r_{l+1}}{D_l} \frac{\tau}{\varepsilon},\tag{32}$$

which depends on the thickness $r_l - r_{l+1}$ of the layer, its tortuosity τ , porosity ε and the diffusion constant



Fig. 2. Multi-layer shrinking core model. A particle consisting of three concentric layers of different reaction states is surrounded by a thin film of stagnant fluid. For a reaction to take place on the interface between two layers, the gaseous reactant has to diffuse through the stagnant fluid and into the grain.

$$D_{l} \equiv \left[\frac{1}{D_{l}^{(m)}} + \frac{1}{D_{l}^{(K)}}\right]^{-1}.$$
(33)

The latter contains contributions from Knudsen diffusion of the gaseous reactant through material pores of width $d_{\text{pore},l}$ (Welty et al., 2020)

$$D_l^{(\mathrm{K})} = \frac{d_{\mathrm{pore},l}}{3} \sqrt{\frac{8RT}{\pi m_{\mathrm{react}}^{(\mathrm{mol})}}}$$
(34)

and of diffusing species within in a gas mixture, $D_l^{(m)}$. This molecular diffusion mechanism can be modeled with the Fuller-Schettler-Giddings correlation (Fuller et al., 1966).

Finally, diffusion through the stagnant fluid around each grain hinders the reaction in terms of

$$F \equiv \frac{a_{\rm p}}{{\rm Sh}D_l^{(\rm m)}},\tag{35}$$

where the Sherwood number Sh contrasts convective and diffusive mass transfer (Froessling, 1938).

Given the various resistances A_l , B_l and F, the difference of the reactant molar concentration $x_{\text{react}}^{(f)}$ from the chemical equilibrium value $x_{\text{eq},l}$ of each reaction leads to a molar consumption rate of the reactant $\dot{X}_{\text{react},l}^{(f)}$ on each interface. The corresponding equations are provided in Appendix A.

Once the molar amounts of consumed reducing agent are determined, it is straightforward to obtain the lost and the gained masses of the gaseous reactant and product, $\dot{m}_{\rm react,l}^{(f)}$ and $\dot{m}_{\rm prod,l}^{(f)}$, as well as those of the solid species $\dot{m}_{\rm react,l}^{(s)}$ and $\dot{m}_{\rm prod,l}^{(s)}$ of the corresponding layers. These mass changes reflect in the total grain mass

$$\frac{\mathrm{d}m_k}{\mathrm{d}t} = \sum_l \dot{m}_{\mathrm{prod},l}^{(\mathrm{s})} - \dot{m}_{\mathrm{react},l}^{(\mathrm{s})} \tag{36}$$

and in the source terms

$$\dot{\rho}_{\text{p-f},r} = -\sum_{k} g(\boldsymbol{r} - \boldsymbol{r}_{k}) \sum_{l} \dot{m}_{\text{react},l}^{(f)}$$
(37)

$$\dot{\rho}_{\text{p-f},p} = \sum_{k} g(\boldsymbol{r} - \boldsymbol{r}_{k}) \sum_{l} \dot{m}_{\text{prod},l}^{(f)}$$
(38)

of the fluid transport equations (2) for the reactant and product species r and p, where the inner sums run over all interfaces l of each particle k.

The reaction heat that is released or absorbed at each interface of each grain can be obtained from the difference of product and of reactant energies at the local temperature,

$$\dot{Q}_i = \dot{m}_{\text{prod},l}^{(\text{f})} e_{\text{prod},l}^{(\text{f})}(T) + \dot{m}_{\text{prod},l}^{(\text{s})} e_{\text{prod},l}^{(\text{s})}(T)$$

$$-\dot{m}_{\text{react},l}^{(f)}e_{\text{react},l}^{(f)}(T) - \dot{m}_{\text{react},l}^{(s)}e_{\text{react},l}^{(s)}(T).$$
(39)

The solid contributions stem from changes of the grain layer masses, which has further implications for the particle energy equation (15). Since the grain internal energy is given by the sum of the layer energies $m_i e_i(T) = \sum_l m_{i,l} e_{i,l}(T)$ and the heterogeneous reaction gives rise to a source term $\sum_l m_{i,l} e_{i,l}(T)$, the latter cancels the time dependence of the layer masses. Using $de_{i,l}/dt = C_{i,l} dT_i/dt$, one finds

$$m_i C_i(T_i) \frac{\mathrm{d}}{\mathrm{d}t} T_i = \dot{Q}_i^{(\mathrm{p}-\mathrm{f})} + \dot{Q}_i^{(\mathrm{p}-\mathrm{p})} \tag{40}$$

where the overall heat capacity is given as mass-weighted sum of the layer capacities.

Shrinking particle model Once a multi-layer shrinking core model has been implemented, deriving a shrinking particle model is straight forward and the expressions obtained in the previous section can be used with minor modifications. No internal interfaces between different material species exist, and the reaction takes place only on the grain surface. While diffusion into the particle can therefore not limit the reaction rate, diffusion through the surrounding gas according to Eq. (35) needs to be retained. In the present study, we applied such a simplified model for coke combustion and the reverse Boudouard reaction Eqs. (25) and (26) using similar reaction parameters as Shen et al. (2011). We remark that we did not attempt to resolve the complex RW chemistry. Instead, we assumed that all O_2 was finally converted into CO.

2.3. Parcel approach for polydisperse size distributions

In order to keep particle numbers in a manageable range even for large-scale problems, parcel approaches (Bierwisch et al., 2009; Sakai and Koshizuka, 2009) are commonly used. Several grains are grouped into a parcel with appropriately adapted material and interaction parameters (Brandt et al., 2023), which reduces the computational costs massively. Besides the contact mechanical properties, the particle-fluid mass/momentum/heat exchange terms need to be corrected, too. For example, if λ^3 grains of diameter *d* constitute a parcel with $D = \lambda d$, the coarse-grained drag force is given by $F_{cg}^{(drag)}(D) \equiv \lambda^3 F^{(drag)}(d)$. While a large amount of research has been conducted for monodis-

While a large amount of research has been conducted for monodisperse systems, much less has been done on polydisperse size distributions (Chu et al., 2016; Verma et al., 2017). Possible strategies comprise scaling all grains with a fixed λ so that the original size distribution is retained, or using a size-dependent λ such that all parcels have the same size (Lu et al., 2018). In a previous study (Lichtenegger and Pirker, 2018), we composed parcels of particles with different diameters, which corresponds to a more "macroscopic" perspective where a bulk material is decomposed into material lumps and contact parameters are obtained from calibration towards e.g. angle of repose or shear cell tests. More specifically, a broad size distribution of grains with Sauter mean diameter d_S is mapped onto monodisperse effective parcels with $\tilde{d} = d_S$, which carry a volume of

$$\tilde{V} = \tilde{f} \frac{\pi}{6} \tilde{d}^3 \tag{41}$$

each. The effective volume factor

$$\tilde{f} \equiv \frac{\alpha_{\rm RCP}^{(\rm poly)}}{\alpha_{\rm RCP}^{(\rm mono)}} \tag{42}$$

accounts for the fact that broad size distributions can occupy more space than monodisperse ones and can be obtained from correlations for the maximum packing fraction $\alpha_{\rm RCP}^{(\rm poly)}$ depending on the underlying size distribution. Any reference volume of densely packed particles should contain the same mass regardless if represented by *N* grains of the actual size distribution or by \tilde{N} monodisperse, effective parcels, i.e.

$$\frac{\pi}{6}\rho_{\rm p}N\frac{\sum_{i}d_{i}^{3}}{N} = \frac{\pi}{6}\rho_{\rm p}\tilde{f}\tilde{N}\tilde{d}^{3}.$$
(43)

Hence, each parcel represents

$$\tilde{n} \equiv \frac{N}{\tilde{N}} = \tilde{f} \frac{\tilde{d}^3}{\langle d^3 \rangle}$$
(44)

particles, where $\langle d^3 \rangle \equiv 1/N \sum_i d_i^3$ is the expectation value of the cube diameter computed over the underlying size distribution.

If the bulk density of a material is known, one may directly specify \tilde{f} from Eq. (42) without determining the underlying size distribution. Besides its effect on the parcel volume, \tilde{f} also enters the particle-fluid exchange rates, which are obtained from those for actual particles of diameter $d_{\rm S}$, as overall scaling factor (Lichtenegger and Pirker, 2018).

Finally, the monodisperse parcels may be coarse-grained with the rules of standard upscaling, which increases the number of particles in each parcel by λ^3 .

2.4. Simplifications for pseudo-steady conditions

If one is mainly interested in the global state of a moving bed reactor under regular operating conditions and less on the detailed particlelevel processes, it stands to reason to ignore granular stick-slip dynamics and approximate the particle motion with a velocity field v_p that does not change over long durations. Hence, one may replace the numerically expensive Eqs. (11) and (12) with

$$\frac{\mathrm{d}}{\mathrm{d}t}\boldsymbol{r}_i = \boldsymbol{\nu}_p(\boldsymbol{r}_i) + \delta\boldsymbol{\nu}_{\mathrm{relax}},\tag{45}$$

where $v_p(\mathbf{r})$ can be obtained as temporal average of the grain velocities in a CFD-DEM simulation, and δv_{relax} is a correction that prevents overpacking due to numerical inaccuracies. Possible approximations for the latter term may be constructed from the local volume fraction field or from a soft, repulsive force between particles with overlaps. Such simplifications allow to solve Eq. (45) with time steps Δt_{tracer} much larger than those usually encountered in DEM simulations. Ultimately, Δt_{tracer} has to be chosen sufficiently small to (i) follow the curvature of the particle velocity field and (ii) to picture any other particle equations such as those for temperature and chemical composition accurately.

For conditions not too far away from the thermal steady state of the reactor, one may further assume that the local fluid properties do not vary too quickly. Consequently, one may discard the transient terms of Eqs. (1) – (4) and solve their steady version for each large step. However, for reactors with a melting zone, the granular temperature distribution $T_p(r)$ is coupled to the particle velocity field. An initial guess for $T_p(r)$, with which $v_p(r)$ is obtained from a CFD-DEM calculation, will likely change over time and necessitate an update of $v_p(r)$. The same would be true for temperature-dependent particle properties, e.g. for polymers, which might be dealt with in an analogous fashion. Since the large-scale particle temperature field will generally change over durations far beyond DEM time scales, the combination of Eq. (45) with intermittent corrections from short-term CFD-DEM simulations is still much faster than actual (CFD-)DEM computations.

Taken together, the proposed approach is built upon the following simplifying hypotheses:

- The particle bed moves smoothly without any slip-stick irregularities. Its motion is determined by the shape and location of the CZ, which needs to be updated iteratively. The gas flow impacts the large-scale granular dynamics only weakly.
- The fluid momentum, thermal energy and chemical composition change only very slowly in time but may exhibit strong spatial gradients.
- Massive coarse-graining to reduce the particle number to a manageable amount is admissible. Consequently, the spatial resolution is limited, and the RWs can be pictured only insufficiently.
- Intergranular heat transfer may be approximated without detailed knowledge of the contact structure.

More details on the simplifications of the set of coupled particle-fluid equations for pseudo-steady conditions can be found in our previous publications (Lichtenegger, 2020; Lichtenegger and Pirker, 2020).

3. Simulation setup

We report on two case settings of vastly different size. First, we carried out small-scale calibration simulations to determine appropriate reaction parameters for ore. Then, we used their optimized values to study a full-scale BF over the course of many hours to obtain its thermal steady state. We considered two common charging patterns: (i) layers of ore and of coke distributed uniformly across the BF and (ii) a narrow, ore-free center surrounded by material layers.

The implementation of the pseudo-steady CFD-DEM approach including the various submodels in the framework of LIGGGHTS (Kloss et al., 2012) and CFDEMcoupling (Goniva et al., 2012) as well as the case setups can be found in our repository https://github.com/orgs/ ParticulateFlow upon request.

3.1. Reduction test

A cylinder with diameter $d_{cyl} = 83 \text{ mm}$ was filled with a fixed bed of approximately $m_{pellets} = 500 \text{ g}$ hematite pellets with diameters $10 \text{ mm} < d_{pellets} < 12 \text{ mm}$. After preheating the material sample to the target temperature of $T_{red} = 1223 \text{ K}$, a gas mixture of $60\% \text{ N}_2$ and 40% CO with the same temperature streamed with a mass flow rate of $\dot{m}_{gas} = 1.05 \text{ g/s}$ through the cylinder and reduced the hematite to magnetite, wustite and finally to iron. The reduction process was monitored via the mass loss of the pellets and compared to measurements on a corresponding system carried out at voestalpine AG (Feilmayr, 2022).

While we took literature values as specified in Sec. 2.2.3 for most chemical parameters for iron oxide reduction, the reaction rate prefactors $k_l^{(0)}$ and the pore diameters $d_{\text{pore},l}$ of the various layers were obtained from optimization (cf. Sec. 4.1).

Computations were carried out with different values of the CFD time step ranging from $\Delta t_{\rm CFD}^{\rm (trans)} = 1.25\cdot 10^{-3}$ s for fully transient calculations up to $100\Delta t_{\rm CFD}^{\rm (trans)}$ for the pseudo-steady variant. Coupling between the CFD and the DEM codes took place every CFD time step.

3.2. BF

A first impression of the case setup is provided by Fig. 3, which shows a snapshot of the simulation geometry and an illustration of the particle bed. The height of 27.2 m and maximum diameter of 14.8 m corresponded to the extensions of a real BF without the hearth region. Since we did not model the liquid phases and their drainage behavior within this study, we omitted the corresponding part of the reactor.

Oxygen-enriched, hot air with $T_{\rm in} = 1600$ K, entered through 32 inlets with a total mass flux of $\dot{m}_{\rm in} = 112.6$ kg/s and left through the furnace top at a prescribed pressure of $p_{\rm out} = 330$ kPa. Heat loss $\dot{Q}_{\rm wall}/A = h_{\rm wall}(T - T_{\rm wall})$ was accounted for in terms of a heat loss coefficient $h_{\rm wall} = 20$ W/(m²K). The same value had been used in the study of Zhang et al. (2016).

Relatively small amounts of fine coke and fine ore were inserted next to the walls and coarser coke and ore grains in the center. Using massive coarse graining, their total number was in the range of 800 000. The most important material properties are summarized in Table 1. The temperature and composition dependence of the heat capacities was obtained from seven-coefficient NASA polynomials (Burcat and Ruscic, 2005).

Coke was removed in front of the gas inlets and ore once it entered regions where the temperature exceeded $T_{\rm p}^{\rm (melt)} = 1675$ K. For the data-generating CFD-DEM simulations, coke removal took place with a predefined, artificially high rate of $m_{\rm RW} = 100$ kg/s per RW so that the time-averaged particle velocity and packing density fields converged faster. Afterwards, the particle velocity field was scaled down with a



Fig. 3. Sketch of the case geometry. A furnace with a height of 27.2 m and a maximum diameter of 14.8 m was filled with a packed particle bed consisting of coarse coke and ore in the center and fine coke and ore close to the walls). 32 gas inlets (red) were located around the circumference close to the bottom.

Table 1	
Particle material and contact properties.	

property	coke	fine coke	ore	fine ore
<i>d</i> _p [m]	0.044	0.02	0.015	0.006
$\rho_{\rm n}$ [kg/m ³]	950	950	3300	3300
λ	4	8	11	24
$f_{\rm n}$	0.81	1.2	1.03	1.2
$k_{\rm n}^{\rm r}$ [W/(mK)]	1.7	1.7	80	80
É [Pa]	$5 \cdot 10^{7}$	$5 \cdot 10^{7}$	$5 \cdot 10^{7}$	$5 \cdot 10^{7}$
ν	0.25	0.25	0.25	0.25
e	0.5	0.5	0.5	0.5
$\mu_{\rm f}$	0.9	0.9	0.8	0.8
$\mu_{ m rf}$	0.5	0.5	0.4	0.4

constant factor such that the descent velocity at the top of the furnace agreed with the value of $v_{\rm desc}\approx 0.002~\rm m/s$ observed in an actual plant.

The stiffness of the solid particles required small DEM time steps $\Delta t_{\text{DEM}} = 2.5 \cdot 10^{-5} \text{ s}$ in the CFD-DEM case, whereas we could use significantly larger steps $\Delta t_{\text{tracer}} = 5 \cdot 10^{-2} \text{ s}$ for tracers. While the requirement to follow the prescribed velocity field would have allowed even larger steps, we chose to equip the tracers with a linear, normal repulsion velocity $\delta v_{\text{relax}} = \kappa_n \delta_n n$ with $\kappa_n = \mathcal{O}(100)$ 1/s to prevent overpacking, which led to the above value of Δt_{tracer} . Since we postulated pseudosteady conditions, we could use a large CFD time step $\Delta t_{\text{CFD}} = 2 \text{ s}$. Within each of these steps, we solved the fluid equations (1) – (4) with $N_{\text{it}} = 10$ iterations.

Data-generating CFD-DEM simulations were carried out until they had equilibrated after the update of the CZ, which involved removal of all ore grains below the new CZ. Usually, it took about 24 s to 28 s of process time until the mean descent velocity of the burden had stabilized. The fast pseudo-steady CFD-DEM simulations covered episodes of 10 h, which was long enough so that the temperature field could evolve substantially, but not so long that too disruptive updates of the CZ would necessitate overly long equilibration times in the subsequent CFD-DEM calculations.



Fig. 4. Mass loss of an ore sample for different time steps. Transient calculations led to almost the same results as pseudo-steady computations with an up to 100 times larger step size.

Table 2

Ore reaction parameters. The pore diameters d_{pore} and the kinetic constants k_0 were obtained from optimization whereas the activation energies $E^{(a)}$ were chosen according to Valipour (2009).

reaction	d _{pore} [m]	k ⁽⁰⁾ [m/s]	$E^{(a)}$ [J/mol]
He to Ma	$0.56 \cdot 10^{-5}$	3700	113859
Ma to Wu	$0.88 \cdot 10^{-5}$	18	73674
Wu to Fe	$1.5 \cdot 10^{-7}$	10	69488

4. Results

4.1. Multi-layer shrinking core model

4.1.1. Calibration of chemical parameters

Since parameter optimization can be numerically very costly, we first investigated how large we could reasonably set the time step and carried out several rather short CFD-DEM simulations with literature values for the various chemical parameters. We ran a conventional, transient simulation with small time step (cf. Sec. 3) and several pseudo-steady ones to investigate how large an error this approximation induced. As expected pseudo-steady simulations with the same step size gave very similar results, and this behavior also held for up to 100 times the step size of the transient calculations. Fig. 4 shows that hardly any difference in the mass loss due to the reduction reaction could be found over a period of 100 s, which underlines the validity of the pseudo-steady approach and the accompanying opportunity to use large time steps.

This observation made it feasible to run a large number of simulations over longer durations such that we could attempt the optimization of several chemical parameters towards experimental findings (Feilmayr, 2022) for the mass loss over time. It can be seen in Fig. 5 that the optimized values provided by Table 2 reproduced the measured curve extremely well during the whole reduction process. Fig. 5b underlines over how different time scales the various steps took place. The initial reduction of hematite to rather small amounts of magnetite and further to wustite was significantly faster than the last reduction step from wustite to iron.

4.1.2. Parameter sensitivity

Notably, the sensitivity of the final results on the various parameters varied strongly. While changing the reaction rate prefactor $k_{WuFe}^{(0)}$ or the pore diameter of the iron layer by $\pm 50\%$ led to significant changes in the mass loss and changing the rate constant $k_{MuWu}^{(0)}$ by $\pm 50\%$ had an evident impact at least in the first phase of the process, similar variations in the pore size of magnetite or wustite or the other reaction constants did not change the final results visibly. We ascribe this behavior to the fact that the optimized iron pore diameter of $d_{Pore}^{Fe} = 1.5 \cdot 10^{-7}$ m was



Fig. 5. Ore reduction after parameter optimization. (a) The simulated mass loss agreed very well with the measured one over the whole observation period. An error of $\pm 50\%$ in $k_{\rm MaWu}$ would lead to a clearly visible deviation during the first part of the process, whereas an error of the same magnitude in $k_{\rm WuFe}$ or in $d_{\rm pore}^{Fe}$ would cause a larger discrepancy except at the very beginning. (b) Within the first 1000 s, hematite vanished rapidly and was mainly replaced by wustite (with an intermediate, slim magnetite phase). The reduction of wustite to iron took much longer and was still ongoing after 8000 s.

two orders of magnitude smaller than those of magnetite and wustite so that reactions involving iron took much longer and had therefore a larger impact. Hence, it is vital to obtain accurate estimates for the pore size of iron and the reaction rates from wustite to iron and from magnetite to wustite. The parameters for the other species and reactions seemed to be less important in our study.

4.2. Full-reactor thermo-chemical steady state

4.2.1. Flow patterns

Any long-term properties of interest such as the thermal or chemical steady state were results from the underlying flow patterns of the fluid and the granular phase. Fig. 6 shows that gas streamlines were visibly influenced by the CZ, which hindered its passage and forced it through the remaining coke windows. Upon closer inspection, the impact of the charging pattern can be identified. An ore-free center with higher porosity and grain size led to an increased gas flux in this region.

The particle trajectories on the other hand were less influenced by the material distribution. In both cases, grains moved along relatively straight paths towards/through the CZ and the remaining coke particles into the RWs, where they were consumed. However, those that were charged either at the center or very close to the walls had a longer residence time in the BF than those starting from intermediate locations. Figs. 6c and 6d show that there were zones with very low particle velocities originating from between the RWs, the so-called deadman, and next to the walls of the furnace belly.

The transport behavior of the gaseous species, in particular the increased flow through the ore-free center, reflects for example in the spatial distribution of the different iron oxides (cf. Sec. 4.2.3). The effects of the non-uniform particle velocity distribution can be seen most obviously in the temperature field and the resulting CZ (cf. Sec. 4.2.2).

4.2.2. Thermal properties

Starting from an initial guess for the temperature distribution in the BF, it took several hours until an approach towards the steady state was visible. It can be seen in Fig. 7 how the mean particle temperature increased rapidly (after a brief, weak drop which we ascribe to initial, numerical fluctuations) for several hours after which the increase slowed down. Even though the steady state had not been completely reached yet after 20 h, and the mean temperature still grew slowly, we believe that the system was reasonably close to its steady state at that time (cf. the discussion on the temperature field and the convergence of the CZ below).

Notably, the mean particle temperatures for the uniform-layer and the ore-free center case were almost identical until the first CZ update after 10 h and remained very similar afterwards despite their very different spatial distributions displayed in Fig. 8. Due to the higher gas flux through the center and the lower heat capacity of coke per volume than that of ore, a very strong temperature peak was present in the ore-free center ranging up almost to the top of the furnace. A charging pattern of uniform layers, on the other hand, led to a rather flat temperature distribution.

In both cases, temperatures below the CZ were very high due to the heat released by combustion of coke. Directly above the CZ, they dropped quickly for two reasons: (i) the latent heat absorbed during the melting process of iron (ore) and (ii) the endothermic, reverse Boudouard reaction regenerating part of the CO_2 from ore reduction into CO. However, since the reduction from wustite to iron and from hematite to magnetite are exothermic, a region of relatively constant temperature, the thermal reserve zone, established further up in the BF.

Fig. 8 demonstrates that the shapes of the CZs had almost converged because they hardly changed between 10 h and 20 h whereas a significant transformation had taken place within the first 10 h. Even though the CZ for the ore-free center case had a very pronounced peak while that of the uniform-layer configuration was flatter, both shared the presence of dips above the RWs. These structures may be explained with the granular velocity fields shown in Figs. 6c and 6d. Since particles above the RWs moved down faster than in the center or close to the walls, they had less time to heat up and reached their melting temperature further down.

4.2.3. Chemical properties

The chemical compositions of the ore grains that developed from the gas flow fields in Fig. 6 with accompanying species transport and the temperature distributions in Fig. 8 are displayed in Fig. 9. The dominant species was magnetite which had played a minor role in the reduction test described in Sec. 4.1. However, due to somewhat different conditions concerning temperature and CO concentration in the BF, it took much longer to transform magnetite into wustite. However, the last reduction step to iron took place quite quickly so that it became the prevalent species upon melting in the CZ as can be seen in Fig. 9. Since more gas moved through the center without ore than in the case of a uniform layer structure, adjacent ore could still reduce to a state of mostly iron before melting even though this happened much farther up in the BF.

The mass fractions of the iron oxides found at each height in the BF are provided in Fig. 10. The rapid decrease of hematite and a corresponding increase of magnetite can be clearly seen. While the observed behavior was qualitatively similar for both charging patterns, the uniform-layer configuration led to a slightly higher reduction degree at



Fig. 6. Gas and particle flow patterns. Gas streamlines (upper row) were slightly deflected at the CZ where they passed through horizontal coke windows. For (a) uniform material layers, the gas flow was distributed more evenly than for (b) the ore-free center with a central domain of higher velocity. In the particle velocity field (lower row), an almost stagnant region between the RWs was present. Hardly any differences regarding grain trajectories could be found between (c) the uniform-layer and (d) the ore-free center configuration.



Fig. 7. Mean particle temperature over time. After a rapid increase over the first few hours, the curves start to level off and approach their thermal steady state value.

the approximate height of the CZ. The fact that ore had not been completely reduced to iron was to be expected (Geerdes et al., 2004). To determine the final chemical state in the actual process, a description of the trickling bed below the CZ including a model for the direct reduction of liquid wustite in contact with coke would be necessary.

Fig. 11 provides details on how much of each gas species was consumed by which reaction. Most importantly, most CO took part in the ore reduction to become CO_2 which was partly regenerated into CO. While only a small fraction of CO left the BF, much more CO_2 did so. The amounts converted by ore reduction and the reverse Boudouard reaction were calculated from the mass influxes and outfluxes obtained from the simulation. We remark that the ratio of CO_2 to CO leaving the BF and the resulting gas utilization factor were too high for a real plant.

4.2.4. Numerical costs

The computational costs connected to full CFD-DEM simulations for flow field updates and to pseudo-steady CFD-DEM calculations of 10 h intervals on eight quad-core X5570 Xeon CPUs resulting in a total of 32 cores are provided in Fig. 12. The massive speed up of the latter in comparison to the former is demonstrated by the fact that it took only slightly longer to simulate 10 h of actual process time than a single flow field update of about 28 s. This acceleration of about three orders of magnitude was mainly caused by the much larger time steps



Fig. 8. Particle temperature fields. The same initial CZ (dotted line) developed into a much flatter structure for (a) the uniform-layer configuration than for (b) the ore-free center with large temperatures high up in the center of the BF. The dashed lines represent the CZs after 10 h, the straight ones after 20 h.



Fig. 9. Angle-averaged dominant iron oxide fractions. Iron became prevalent only briefly before reaching the CZ. In most of the furnace, magnetite was dominant. The black line represents the lower edge of the CZ.

that could be used in the pseudo-steady CFD-DEM simulations. The relative contributions to runtime, on the other hand, were not too different in both cases. While DEM calculations (particle motion, temperature and composition changes) caused significant costs, their CFD counterpart (solution of the Navier-Stokes equations) was relatively cheap. However, most time was spent with coupling of the particle and the fluid phase. This term comprised interprocessor communication, translation of Eulerian into Lagrangian information and vice versa as well as the computation of mass, momentum and heat transfer rates. For the pseudo-steady CFD-DEM simulations, the fraction of CPU time spent within the DEM code was comparatively smaller because the full force computation was replaced by the cheaper repulsion velocity $\delta v_{\rm relax}$ while more data connected to the various gas species had to be communicated and their transport equations solved.

Further, significant performance improvements for the pseudosteady CFD-DEM simulations can be most likely achieved simply by using larger time steps. In the present study, our choice was limited by the soft repulsion between grains to avoid overpacking, but a more elaborate treatment could allow larger steps closer to the limit dictated by temperature and composition changes. Concerning the data-generating



Fig. 10. Iron ore mass fractions over height. After hematite had largely vanished, magnetite made up most of the ore before being reduced to wustite which was transformed into iron just before melting. In the lower regions overlayed in grey, most iron (ore) was molten so that corresponding data are not available.

CFD-DEM simulations, a different strategy would have to be chosen because the step size cannot be increased easily. However, as already mentioned in the introduction, it would be equally admissible to carry out cheaper continuum simulations to generate the particle velocity and volume fraction fields.



Fig. 11. Gas mass fluxes. O_2 entering the furnace was combusted into CO which was mainly used to reduce the iron ore while a smaller amount left the furnace. Part of the resulting CO_2 was regenerated by the reverse Boudouard reaction whereas most of it left through the top. Values are given in kg/s. The left-hand value of each pair corresponds to the uniform-layer case, the right-hand one to an ore-free center configuration. The O_2 influx and the amount of CO from combustion was the same for both cases.



Fig. 12. CPU time of full CFD-DEM and pseudo-steady CFD-DEM simulations. One flow field update due to changes of the CZ took approximately as long as one pseudo-steady CFD-DEM episode covering 10 h.

5. Conclusion and outlook

In this study, we have a presented framework to deal with the multiphysics, multi-scale problem of hot, reacting moving bed reactors. To this end, we combined comparatively detailed, coarse-grained, full CFD-DEM simulations with extremely fast pseudo-steady CFD-DEM calculations. In a first step, the approach was used to carry out a large number of ore reduction simulations of a small hematite sample to calibrate the most important parameters of a multi-layer shrinking core model using optimization techniques. The achieved, very good agreement with experimental reference data demonstrated that this model is indeed flexible enough to describe the multi-step reduction process. Of course, a greater variety of measurements probing e.g. the sample mass or the grain size dependence would allow for an even more reliable parameter set.

In a second step, we applied the framework to a 3D, full-scale BF and studied the approach to the thermo-chemical steady state over the course of 20 h. We compared a charging pattern of uniform layers of coke and ore with one including an ore-free center. While the former led to a rather flat CZ, the latter was connected to high temperatures in the center of the furnace up to considerable heights with a strongly peaked CZ. However, the overall mean particle temperature and the reduction degree were surprisingly similar in both scenarios and were in qualitative agreement with expectations from a real BF.

In summary, our main finding with two derived insights are

- the capability of fast, pseudo-steady CFD-DEM to cover long process durations of hot, reacting particle beds using high-fidelity data from CFD-DEM,
- the sufficient flexibility of the multi-layer shrinking core model to describe the three-step reduction of hematite to iron *if* optimized parameters are used, and
- the strong influence of the BF charging pattern on the shape of the CZ with only a weak impact on furnace-wide averages of temperature and reduction degree.

Because we primarily wanted to demonstrate the feasibility of our approach to describe multiple physical processes acting on vastly different scales, we included representative models for mechanical, thermal and chemical properties without any claim of completeness. Clearly, they will need to be refined and their list extended to allow for simulations in quantitative agreement with a real plant. For example, we have neglected grain fragmentation and the presence of dust so far, which reflected in a too low pressure drop over the furnace height. Notably, excessive amounts of fines can cause irregularities such as temporary or permanent burden hanging. Processes of this type would be very difficult to describe with the proposed approach that assumes little temporal variation in particle and gas phase motion. However, if one could define sufficiently sensitive indicators for such events, one might switch from pseudo-steady simulations to fully transient CFD-DEM shortly before their occurrence. Of course, any such activities will require a sound description of dust transport and deposition in dense beds.

Besides model improvements within the framework, it is also possible to combine it with other, more detailed techniques for selected subregions of interest. In the present work, we prescribed RW shapes, but one could also decompose the computational domain and couple the full-scale calculations with small-scale simulations of much higher resolution accounting for the complex interplay of fluid mechanics and chemistry in the RWs. While online coupling (Queteschiner et al., 2018) would necessitate heavy algorithmic adaptions and might impair the overall performance, intermittent updates like we already performed them to determine the particle volume fraction and velocity fields would offer a compromise between increased accuracy and numerical costs. The RW subdomains would receive their boundary values like particle packing fraction and particle temperature from the large-scale simulations and in turn would provide gas mass flux, temperature and composition across their surface to the global calculations.

To conclude, we have shown that our approach for moving bed reactors like BFs is indeed capable to combine models bridging different scales and will lend itself to further improvements so that quantitatively accurate results can be obtained in future work. Extensive practical experience and numerous theoretical investigations of the BF have already led to a very good understanding of regular operating conditions. However, undesired, extreme events such as channel formation or burden hanging still pose serious problems. Improved insights into the underlying mechanisms, which may be gained safely in numerical simulations before proceeding to actual experiments and finally to the real plant, might allow to develop counter strategies.

Furthermore, adaptions of reactor design and novel process types will massively benefit from detailed digital shadows or even twins. While this holds for moving beds in general, it is particularly relevant for BFs. Despite the development of alternatives with a lower carbon footprint, their currently operated, huge number will remain substantial in the foreseeable future. Hence, any improvements of the BF process to reduce greenhouse emissions would have large, beneficial effects. Virtual experiments enable pre-studies on the feasibility of any such adaptions at little costs and safety risks.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Thomas Lichtenegger reports financial support was provided by voestalpine AG. Thomas Lichtenegger reports financial support was provided by Primetals Technologies Austria GmbH. Thomas Lichtenegger reports a relationship with K1-MET GmbH that includes: funding grants.

Data availability

Data will be made available on request.

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Appendix A. Solution of the multi-layer shrinking core network equations

For the specific case of a four-layer grain with an iron shell, a wustite and a magnetite layer as well as a hematite core, solution of the Kirchhoff law for the network depicted in Fig. 13 gives

$$\dot{X}_{\text{react,HM}}^{(f)} \equiv \frac{4\pi r_{p}^{2}}{W} \left[\left(A_{\text{WFe}} \left(A_{\text{MW}} + B_{\text{W}} + B_{\text{Fe}} + F \right) + \left(B_{\text{Fe}} + F \right) \left(A_{\text{MW}} + B_{\text{W}} \right) \right) \left(x_{\text{react}} - x_{\text{eq}}^{\text{HM}} \right) - \left(A_{\text{WFe}} \left(B_{\text{W}} + B_{\text{Fe}} + F \right) + B_{\text{W}} \left(B_{\text{Fe}} + F \right) \right) \left(x_{\text{react}} - x_{\text{eq}}^{\text{HM}} \right) - A_{\text{MW}} \left(B_{\text{Fe}} + F \right) \left(x_{\text{react}} - x_{\text{eq}}^{\text{WFe}} \right) \right]$$
(A.1)

$$\dot{X}_{\text{react,MW}}^{(f)} \equiv \frac{4\pi r_{\text{p}}^{2}}{W} \left[\left(\left(A_{\text{HM}} + B_{\text{M}} + B_{\text{W}} \right) \left(A_{\text{WFe}} + B_{\text{Fe}} + F \right) + A_{\text{WFe}} \left(B_{\text{Fe}} + F \right) \right) \left(x_{\text{react}} - x_{\text{eq}}^{\text{MW}} \right) - \left(B_{\text{W}} \left(A_{\text{WFe}} + B_{\text{Fe}} + F \right) + A_{\text{WFe}} \left(B_{\text{Fe}} + F \right) \right) \left(x_{\text{react}} - x_{\text{eq}}^{\text{HM}} \right) - \left(A_{\text{HM}} + B_{\text{M}} \right) \left(B_{\text{Fe}} + F \right) \left(x_{\text{react}} - x_{\text{eq}}^{\text{WFe}} \right) \right]$$
(A.2)

$$\dot{X}_{\text{react,WFe}}^{(f)} \equiv \frac{4\pi r_{\text{p}}^{2}}{W} \Biggl[\Bigl((A_{\text{HM}} + B_{\text{M}}) (A_{\text{MW}} + B_{\text{W}} + B_{\text{Fe}} + F) + A_{\text{MW}} (B_{\text{W}} + B_{\text{Fe}} + F) \Bigr) (x_{\text{react}} - x_{\text{eq}}^{\text{WFe}}) - A_{\text{MW}} (B_{\text{Fe}} + F) (x_{\text{react}} - x_{\text{eq}}^{\text{HM}}) - (A_{\text{HM}} + B_{\text{M}}) (B_{\text{Fe}} + F) (x_{\text{react}} - x_{\text{eq}}^{\text{MW}}) \Biggr]$$
(A.3)



Fig. 13. Mathematical representation of the four-layer shrinking core equations as resistor network. The differences between the local concentration of reactant x_{react} and the equilibrium values x_{eq} of the reactions taking place at the interfaces act as driving potentials.

$$W \equiv (A_{\rm HM} + B_{\rm M}) \Big(A_{\rm WFe} \big(A_{\rm MW} + B_{\rm W} + B_{\rm Fe} + F \big) + \big(A_{\rm MW} + B_{\rm W} \big) \big(B_{\rm Fe} + F \big) \Big) \\ + A_{\rm MW} \Big(A_{\rm WFe} \big(B_{\rm W} + B_{\rm Fe} + F \big) + B_{\rm W} \big(B_{\rm Fe} + F \big) \Big).$$
(A.4)

Expressions for the scenario where e.g. the hematite core has completely vanished are obtained by performing the corresponding limits $r_{\rm H} \rightarrow 0$, which simplifies the resistor network because of $A_{\rm HM} \rightarrow \infty$.

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