

PARTICLE RESOLVED THERMO-CHEMICAL CONVERSION OF PULVERIZED COAL CLUSTERS

MATTHIAS KISS^{1,2,*}, MARKUS BÖSENHOFER^{1,2}, MAGDALENA
SCHATZL¹ AND MICHAEL HARASEK²

¹ K1-MET GmbH
Area 4: Simulation and Analyses
Stahlstraße 12, 4020, Linz, Austria
www.k1-met.com

² Technische Universität Wien
Institute of Chemical, Environmental and Bioscience Engineering
Getreidemarkt 9/166-2, 1060 Vienna, Austria
www.i5-0.com

*Corresponding: matthias.kiss@k1-met.com

Key words: CFD, Resolved Particle, Pulverized Coal Injection, Particle Cluster, Gas-Solid Chemistry

Abstract. Particle clusters form in the turbulent flow of boilers and furnaces when injecting pulverized coal. We study the difference in conversion behavior between a pulverized coal cluster and a single char particle. The simulations are done using a detailed particle model in OpenFOAM[®]. The detailed particle model is capable of spatially resolving char particles and solving transport equations for heat and mass inside the particles. Furthermore, a surface chemistry model is employed for modeling gas-solid reactions inside the pores of the particles. The char particles are coupled to the surrounding bulk fluid phase using explicit Robert-Neuman coupling. 2-D simulations are carried out for a cluster of 16 particles and a single particle. Both simulations use the same initial conditions, boundary conditions and particle properties. The results indicate that a lack of oxygen penetration toward the center of the particle cluster alters the thermo-chemical conversion behavior when compared to the single particle. On average this leads to a decrease of the thermo-chemical conversion rates for particles in the cluster, resulting in a reduction of solid carbon consumption compared to the same number of single particles.

1 INTRODUCTION

Pulverized Coal Injection (PCI) is commonly used to improve the efficiency of blast furnaces [1]. After injection the pulverized coal is subjected to turbulent flow conditions. Under these conditions a strong tendency for the particles to form clusters in turbulent flows exists [2]. The formation of such clusters might cause group combustion effects that alter the thermo-chemical conversion of the pulverized coal.

The efficiency of PCI is determined by the conversion ratio in the raceway zone. The residence time of injected coal particles is between 20 - 30 ms and ideally the particles are completely converted before leaving the raceway zone [1]. However, group combustion effects can lead to a slower thermo-chemical conversion of the particles inside a cluster [3]. Conversion of particles inside the cluster could be inhibited due to a lack of oxygen penetration towards the cluster center.

In the following work, we investigate the influence of clustering on the conversion of char particles. In section 2 we introduce the detailed particle model implemented in the open-source CFD toolbox OpenFOAM[®] [4]. In section 3, we will introduce the setup of the numerical simulations and the particle properties that were used to compare the thermo-chemical conversion of a single char particle and a cluster of 16 equidistant particles. In section 4, we discuss the differences in the thermo-chemical conversion behavior of the coal cluster and the single particle in detail. Finally, we will summarize our findings in section 5.

2 MODELING

All simulations are done using a resolved particle model implemented entirely in the open-source software package OpenFOAM[®] [4]. The porous particle is simulated as a Euler-Euler phase that combines both a solid and a gas phase. The particle porosity is the phase-fraction of the gas phase inside the particle. The particle pores themselves are not resolved. The gas phase inside the particle is assumed to be stagnant and to have constant pressure. Furthermore, the gas phase and the solid phase are assumed to be in thermal equilibrium and have the same temperature. The gas phase around the particle is treated as a Euler-phase, using a regular compressible solver available in OpenFOAM[®]. As a result, the simulations are split into two separate regions: the porous particle(s) and the surrounding gas-phase. These two regions interact via an iterative explicit-Robin-Neuman coupling [5]. The coupling is realized via a set of boundary conditions for temperature and species mass fractions. Furthermore, we introduce a Stefan flow boundary condition for the velocity at the interface. This velocity field results from the net production/consumption of gas-phase species at the particle surface due to the thermo-chemical conversion of solid carbon. The Stefan flow velocity \vec{U}_s is calculated using the following relation [6]:

$$\vec{n} \cdot \vec{U}_s = \frac{1}{\rho} \sum_{k=1}^{K_g} \dot{s}_k W_k \quad (1)$$

where \vec{n} is the particle's outer surface normal vector, ρ is the particle density, \dot{s}_k is the net molar production rate of each species k in the gas-phase, and W_k is the molecular weight of each species in the gas-phase.

The net production rate of each gas-phase species, \dot{s}_k is calculated via the surface chemistry model we implemented in OpenFOAM[®], using the SurfaceCHEMKIN [6] approach. Moreover, the tortuosity and specific inner surface are conversion dependent and are modeled according to the SKIPPY code [7].

The gas-phase chemistry is treated using the OpenFOAM[®] laminar combustion model. Mass diffusion is modeled using the Fuller correlation for binary diffusion [8]:

$$D_{ij} = \frac{1 \cdot 10^{-3} T^{1.75} \left(\frac{1}{W_i} + \frac{1}{W_j} \right)^{\frac{1}{2}}}{p \left(\left(\sum_i \nu_A \right)^{\frac{1}{3}} + \left(\sum_j \nu_B \right)^{\frac{1}{3}} \right)^2} \quad (2)$$

Here D_{ij} is the binary diffusion coefficient for two species i and j . T is the temperature, p is the pressure, W_i and W_j are the molecular weights of species i and species j , respectively. The sums over ν_A and ν_B are summing over the diffusion volumes of each atom/group/structure of each diffusing species i and j .

The effective diffusion coefficient D_{eff} of each species i in the mixture is calculated according to the Wilke mixture rule [9]:

$$D_{eff} = \frac{1 - X_i}{\sum_{j \neq i} X_j D_{ij}} \quad (3)$$

where X_i is the mole fraction of species i .

The current model supports stationary and constant sized particles. Therefore, the model is applicable for the part of the thermo-chemical conversion process before particle shrinking becomes relevant. Once Regime-III of the thermo-chemical conversion process is reached, all chemical reactions occur at the outer particle surface, leading to the shrinking of the external particle surface being an important effect to limit particle reactivity. Furthermore, particle movement or rearrangement that might further influence the conversion process, is disregarded in the simulation.

3 SIMULATION SETUP

We compare simulations of a static particle cluster of 16 2-D particles to simulations of a single particle with identical initial conditions and boundary conditions. The initial conditions and boundary conditions are listed in Table 1 and Table 2, respectively.

Table 1: Simulation initial conditions

temperature	pressure	velocity	N ₂	O ₂	CO ₂	H ₂ O
1500 K	1 · 10 ⁵ Pa	10 m/s	0.483	0.326	0.105	0.086

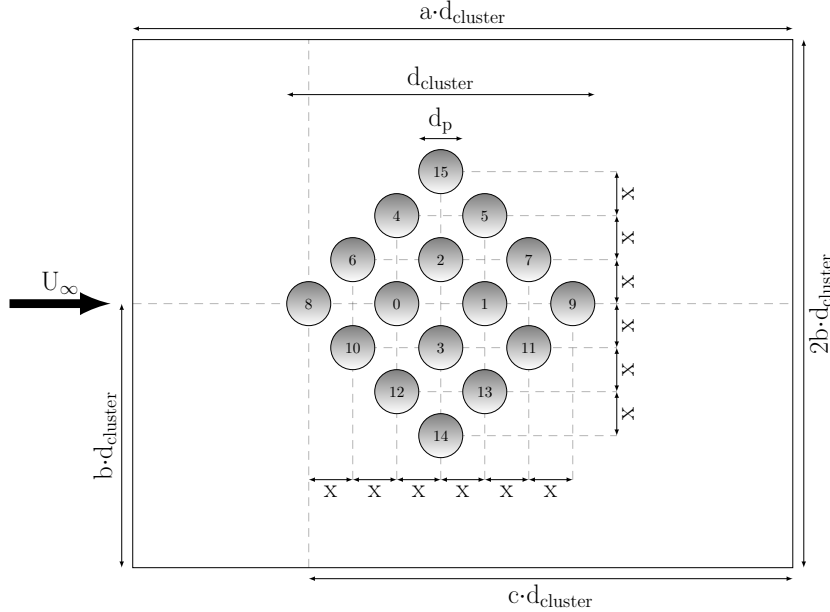


Figure 1: Schematic view of the particle cluster. The cluster as well as the whole simulation domain have the following dimensions: $d_p=100 \mu\text{m}$, $X=1.25 d_p$, $d_{\text{cluster}}=8.75 d_p$, $a=11.5$, $b=4$, $c=8$. The inlet is located at the left side, the outlet on the right. Initial conditions and boundary conditions are listed in Table 1 and Table 2, respectively.

The particle arrangement inside the cluster and all relevant dimensions of the simulation domain are shown in Figure 1.

Each particle has an identical circular shape of diameter $100 \mu\text{m}$ and uses the particle properties in Table 3, which were taken from [10, 11, 12]. The same particle properties have been used for simulations of pulverized char in literature before [13]. We employ a subset of the reaction mechanism proposed by Cai and Pitsch [14] which is originally based on GRI-Mech3.0 for the gas-phase chemistry. Inside the particles the surface reactions listed in Table 4 are used. These reactions were adopted from the mechanism proposed by Hecht et. al. [15].

Table 2: Simulation boundary conditions

boundary patch	temperature	pressure	velocity	mass fractions
inlet	constant (T_0)	zero gradient	constant (U_0)	constant (Y_{i0})
outlet	zeroGradient	constant (p_0)	flow stabilizing	zeroGradient
upper-wall	zeroGradient	zeroGradient	constant (U_0)	zeroGradient
lower-wall	zeroGradient	zeroGradient	constant (U_0)	zeroGradient
particle-interface	coupled	zeroGradient	Stefan-flow	coupled

Table 3: Particle properties [10, 11, 12]. Site density defines the number of surface sites available for surface reactions.

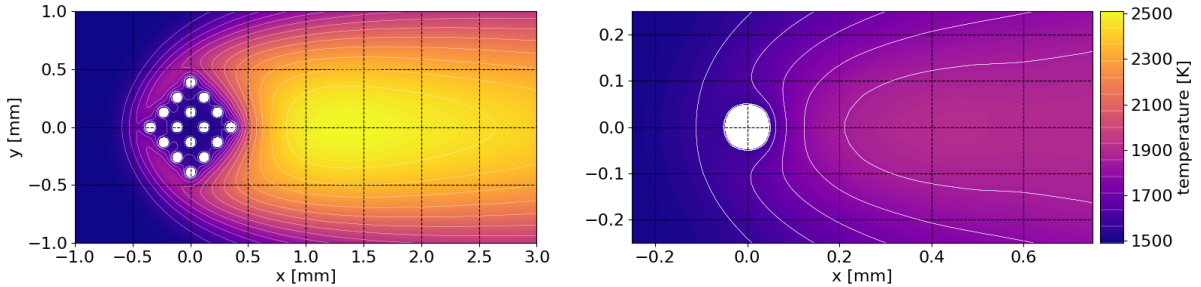
density	heat capacity	thermal conductivity	enthalpy of formation	site density	specific surface	porosity	tortuosity
560 Kg/m ³	2000 J/Kg/m ³	1.33 W/m/K	$2.1 \cdot 10^7$ J/Kg	$1.7 \cdot 10^{-8}$ kmol/m ²	577120 m ² /m ³	0.53	5

Table 4: Surface reactions, their pre-exponential factors (A), and activation temperatures (T_a) [15].

	surface reaction	A [kmol/m ² /s]	T _a [K]
I	$C_s + O_2 \rightarrow CO + O_s$	$3.3 \cdot 10^{14}$	20129
II	$O_s + 2C(b) \rightarrow CO + C_s$	$1 \cdot 10^8$	0
III	$C_s + O_2 \rightarrow O_{2s} + C(b)$	$9.5 \cdot 10^{12}$	17109
IV	$O_{2s} + 2C(b) \rightarrow C_s + CO_2$	$1 \cdot 10^8$	0
V	$C_s + CO_2 \rightarrow CO + O_s + C(b)$	$3.6 \cdot 10^{14}$	30193
VI	$C_s + H_2O \rightarrow H_2 + O_s + C(b)$	$4.35 \cdot 10^{13}$	26671

4 RESULTS AND DISCUSSION

In this section, we assess the influence of particle clustering on the thermo-chemical conversion of char particles. Therefore, we compare single particle and cluster simulations. The simulations were stopped after 5 ms, because after that time the gas-solid reactions shift towards the particle surface. This is equivalent to Regime-III gas-solid thermo-chemical conversion. Without a shrinking particle surface this leads to diverging temperatures at the particle surface. Currently, our model does not support any particle shrinking effects. However, 5 ms corresponds to about 1/4 to 1/6 of the particle residence time of 20 - 30 ms in the raceway zone [1]. Furthermore, it is questionable that a cluster


Figure 2: Temperature profiles around the cluster (left) and single particle (right). The single particle has the same diameter as the 16 individual particles in the cluster. The single particle is displayed larger for better comparison.

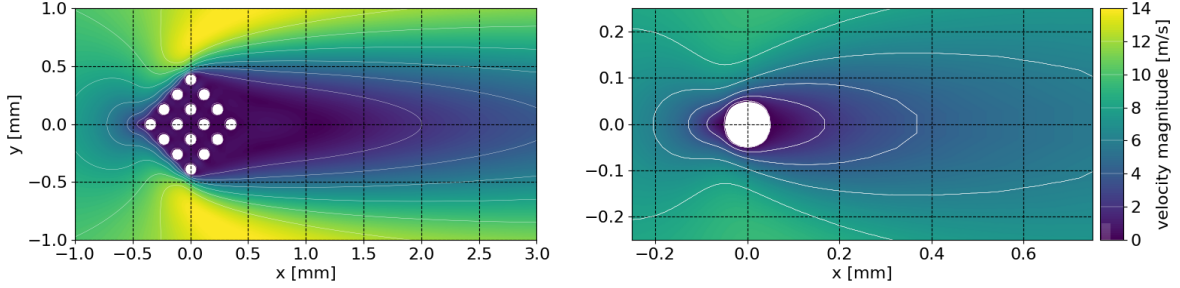


Figure 3: Velocity magnitude profiles around the cluster (left) and single particle (right). The single particle has the same diameter as the 16 individual particles in the cluster. The single particle is displayed larger for better comparison.

would stay in a static configuration throughout the whole raceway zone. It is more likely that the clusters break up and reform after some time [3].

Figure 2 shows that the gas-phase temperature around the cluster is higher than around the single particle. This is a consequence of the higher energy release rates caused by the thermo-chemical conversion of multiple particles. Figure 3 reveals that the flow around the particle cluster behaves similarly to the one around the single particle. It suggests that the particle cluster can be viewed as one macro-particle. It is also visible in Figure 3 that there is hardly any penetration of the bulk flow towards the cluster center. As shown in Figure 4 and Figure 6, this leads to a depletion of O_2 inside the cluster because of the oxidation of solid carbon to CO.

Therefore, diffusive mass transport is the driving force for moving O_2 into the cluster and transporting CO outside. The CO that diffuses outward towards the oxygen rich cluster boundary layer is then further oxidized to CO_2 , which produces most of the heat that is released in the thermo-chemical conversion of the simulated char particles. Figure 2 and Figure 5 show that the temperature around the cluster is indeed highest, where CO_2 has the highest mass fractions. This cluster conversion behavior as a whole is very similar to the conversion behavior of the single particle, where O_2 diffusion into the particle is slow compared to the O_2 consumption inside the particle by oxidizing solid carbon to

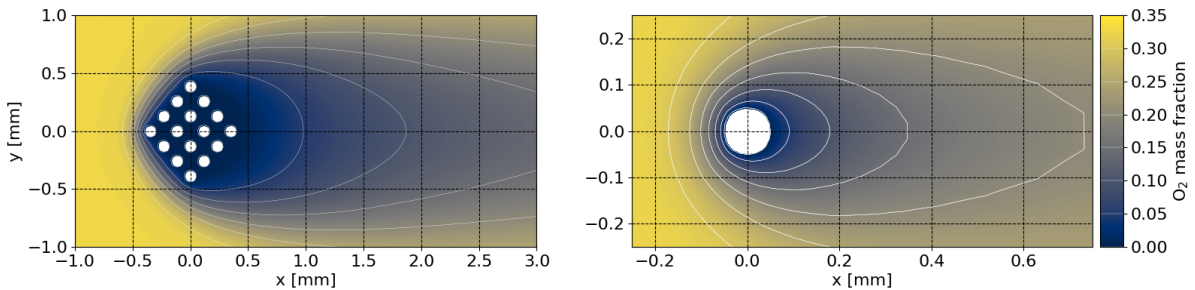


Figure 4: Oxygen profiles around the cluster (left) and single particle (right). The single particle has the same diameter as the 16 individual particles in the cluster. The single particle is displayed larger for better comparison.

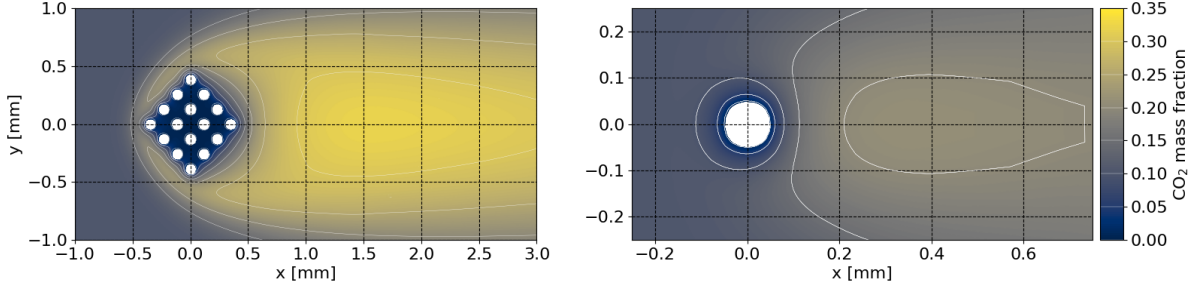


Figure 5: Carbon dioxide profiles around the cluster (left) and single particle (right). The single particle has the same diameter as the 16 individual particles in the cluster. The single particle is displayed larger for better comparison.

CO, which is then further oxidized to CO₂ in the boundary layer. This explains why the cluster as a whole behaves qualitatively similarly to the single particle.

Next we investigate the influence of clustering on the thermo-chemical conversion of each individual particle. Therefore, we calculated the total consumption of solid carbon at two times (0.1 ms and 5 ms) for each individual particle and then summed up the contributions of each particle in the cluster to receive the total solid carbon consumption of the cluster. Figure 7 shows the solid carbon consumption of each individual particle in the cluster, as a percentage of the total solid carbon consumption of the whole cluster. We further calculated the carbon consumption of the single particle simulation, and then divided it by the total solid carbon consumption of the cluster. This yields a percentage equivalent of the carbon consumption of the single particle simulation, that can be directly compared to the particles in the cluster. The particle numbering in Figure 7 corresponds to the one given in Figure 1. In the beginning of the simulation (0.1 ms), all particles of both simulations have almost the same carbon consumption. This is caused by the initialization of the simulations where the whole domain has the same oxygen concentration, which is consumed in the immediate surroundings of the particles. However, at the end of the simulation (5 ms) less solid carbon has been converted by the particles at the center of the cluster than the ones at the outside. The particle at the front of the cluster (particle 8

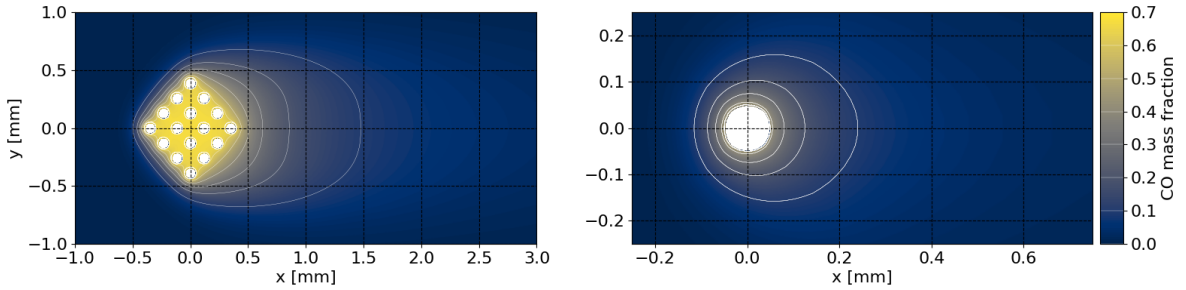


Figure 6: Carbon monoxide profiles around the cluster (left) and single particle (right). The single particle has the same diameter as the 16 individual particles in the cluster. The single particle is displayed larger for better comparison.

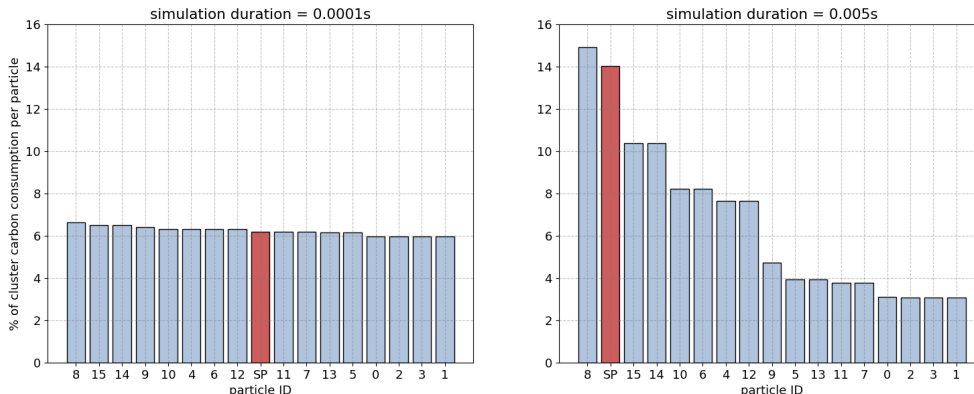


Figure 7: Comparison of the contribution of each individual particle (numbered 0-15) inside the cluster, as well as the single particle (sp) simulation to the total solid carbon consumption. All values are in percentages of the total amount of solid carbon that has been converted by the whole cluster. The single particle carbon consumption is also presented relative to the cluster carbon consumption.

in Figure 1) has the highest total carbon consumption of all particles in the cluster. This particle is at the stagnation point of the gas-phase flow field and O_2 is supplied through advection at higher rates than to other particles. Particle 8 of the cluster and the single particle have an almost identical total carbon consumption compared to the total carbon consumption of the cluster (15% and 14%, respectively). The four particles at the center of the cluster (particles 0, 1, 2, and 3) only contribute around 3% of the total carbon consumption each, which means that those four particles together converted less solid carbon than particle 8 alone.

Further comparison of the total cluster solid carbon consumption to the single particle simulation reveals that after 5 ms of simulation time the 16 particles in the cluster have thermo-chemically converted only 45% of the solid carbon, that 16 individual, un-clustered, particles would have converted.

5 CONCLUSION

In this work we were able to show that clustering has a significant effect on the conversion of char particles. Our detailed particle model coupled with a surface chemistry model akin to SurfaceCHEMKIN allowed us to show that most of the thermo-chemical conversion occurs at the outermost particles of the cluster, while the inner particles have very limited access to O_2 . Therefore, particles at the particle center are limited in their thermo-chemical conversion rates.

When comparing the total solid carbon that has been converted by each particle in the cluster, as well as an individual single particle, we can conclude, that after our 5 ms simulation the cluster has only converted 45% of the solid carbon, that would have been converted by the same amount of independent, un-clustered particles.

Our model is currently only capable of describing stationary, constant-size, particles.

Therefore, the simulations were stopped at 5 ms, before particle shrinking becomes relevant. Residence times of particles in the raceway zone are between 20 - 30 ms [1]. Therefore, further studies have to be made and it is planned to include particle shrinking in our model in the future.

It is essential for PCI, that the injected coal particles are thermo-chemically converted as completely as possible, before leaving the raceway zone. We have shown, that clustering can lead to an inhibition of the thermo-chemical conversion of char particles in the cluster. Therefore, the topic of particle-clustering during PCI warrants further research.

6 ACKNOWLEDGMENTS

The authors gratefully acknowledge the funding support of K1-MET GmbH, metallurgical competence center. The research program of the competence center K1-MET is supported by COMET (Competence Center for Excellent Technologies), the Austrian program for competence centers. COMET is funded by the Federal Ministry for Transport, Innovation and Technology, the Federal Ministry for Science, Research and Economy, the provinces of Upper Austria, Tyrol, and Styria, and the Styrian Business Promotion Agency.

Apart from funding, the project activities are financed by the industrial partners Primetals Technologies Austria, voestalpine Stahl and voestalpine Stahl Donawitz.

REFERENCES

- [1] Ahmed, H. New Trends in the Application of Carbon-Bearing Materials in Blast Furnace Iron-Making. *Minerals* (2018) **8**:561.
- [2] Haugen, N., Krüger, J., Mitra, D. and Løvås, T. The effect of turbulence on mass transfer rates of small inertial particles with surface reactions. *Journal of Fluid Mechanics* (2018) **836**:932–951.
- [3] Krüger, J., Haugen, N., Mitra, D. and Løvås, T. The effect of turbulent clustering on particle reactivity. *Proceedings of the Combustion Institute* (2017) **36**:2333–2340.
- [4] [Online]. Available: <https://openfoam.org>.
- [5] Kiss, M., Bösenhofer, M., Schatzl, M. and Harasek, M. Region Coupling Algorithms in OpenFoam. *16th Minisymposium Verfahrenstechnik and 7th Partikelforum, Vienna* (2020)
- [6] Coltrin, M. E., Kee, R. J, Rupley, F. M. and Meeks E. *Surface Chemkin-III: A Fortran Package for Analyzing Heterogeneous Chemical Kinetics at a Solid-surface Gas-phase Interface* Sandia National Laboratories (1996)
- [7] Muris, S. and Haynes, B. *SKIPPY: A Fortran Computer Code For The Modelling Of The Surface Kinetics In Porous Particles. Description And Application To The Combustion Of Coal Char* Departement of Chemical Enigneering, University of Sydney, Sydney

- [8] Fuller, E. N., Ensley, K. and Giddings J. C. Diffusion of halogenated hydrocarbons in helium. The effect of structure on collision cross sections. *The Journal of Physical Chemistry* (1968) **73**:3679–36851.
- [9] Fairbanks, D. and Wilke, C. Diffusion coefficients in multicomponent gas mixtures *Industrial & Engineering Chemistry* (1950) **42**:471–475.
- [10] Maloney, D. J., Monazam, E. R., Casleton K. H. and Shaddix, C. R. Evaluation of char combustion models: measurement and analysis of variability in char particle size and density. *Proc Combust Inst* (2005) **30**:2197–204.
- [11] Murphy, J. J. and Shaddix, C. R. Combustion kinetics of coal chars in oxygen-enriched environments. *Combust Flame* (2006) **144**:710–29
- [12] Zhang, X. and Bar-Ziv, E. A novel approach to determine thermal conductivity of micron-sized fuel particles. *Combust Sci Technol* (1997) **130**:79–95.
- [13] Farazi, S., Sadr, M., Kang, S., Schiemann, M., Vorobiev, N., Scherer V. and Pitsch, H. Resolved simulations of single char particle combustion in a laminar flow field *Fuel* (2017) **201**:15–28
- [14] Cai, L. and Pitsch, H. Optimized chemical mechanism for combustion of gasoline surrogate fuels. *Combustion and Flame* (2015) **162**:1623–1637.
- [15] Hecht, E., Shaddix, C. R. and Molina, A. Effect of CO₂ gasification reaction on oxy-combustion of pulverized coal char. *Proceedings of the Combustion Institute* (2011) **33**:1699–1706.