PARSCALE - AN OPEN-SOURCE LIBRARY FOR THE SIMULATION OF INTRA-PARTICLE HEAT AND MASS TRANSPORT PROCESSES IN COUPLED SIMULATIONS

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Abstract. *We introduce the open-source library `ParScale` for the modeling of intra-particle transport processes in non-isothermal reactive fluid-particle flows. The underlying equations, the code architecture, as well as the coupling strategy to the widely-used DEM solver `LIGGGHTS®` is presented. A set of verification cases, embedded into an automated test harness, is presented that proofs the functionality of `ParScale`. To demonstrate the capabilities of `ParScale`, we perform simulations of a non-isothermal granular shear flow including heat transfer to the surrounding fluid. We present results for the conductive heat flux through the particle bed for a wide range of dimensionless cooling rates and particle volume fractions. Our data suggests that intra-particle temperature gradients need to be considered for an accurate prediction of the conductive flux in case of (i) a dense particle bed and (ii) for large cooling rates characterized by a critical Biot number of ca $Bi_{crit} \approx 0.1$.*

1 Introduction

Over the past ten years the coupling of the Discrete Element Method (DEM) and Computational Fluid Dynamics (CFD) has been widely adopted by academia and industry to predict fluid-particle flows [1]. Recently, the trend towards CFD-DEM has been fueled by the introduction of open-source toolboxes [2]. These tools are able to model momentum and thermal energy transport of the particles and the fluid with high computational efficiency, hence provide a detailed understanding of granular flow behavior. However, these tools typically do not take intra-particle transport phenomena into account, and hence are unable to model intra-particle processes, e.g., heterogeneous reactions and diffusion inside a porous particle. Unfortunately, in these reactive particulate systems intra-particle processes often play a central role, and hence may dictate overall reactor performance. Thus, spatially-resolved particle properties (e.g., the local gas concentration in the pores of the particle) need to be resolved to account for all relevant phenomena in the system [3].

`ParScale`, a newly developed open-source library implemented in a C/C++ environment and publicly available through *www.github.com* [4], closes this gap. At the current development state, `ParScale` contains a plurality of models that aim on predicting heat and mass transfer, as well as homogeneous and heterogeneous reactions inside flowing porous particles. Also, it is possible to account for a single or multiple-reactions, i.e., a whole reaction network. Due to a modular class-based structure, and the integration into an automated test harness, easy extendibility and a high software quality is ensured. Our contribution outlines the governing equations for modeling various intra-particle phenomena in Section 2. In section 3 a number of verification cases is presented in order to demonstrate and verify the functionality of `ParScale`. Section 4 demonstrates the coupling to `LIGGGHTS` $^{\circledR}$ and evaluates the need to account for intra-particle temperature gradients in non-isothermal granular shear flows cooled by a surrounding fluid.

2 Simulation method and parallel coupling strategy

The key purpose of `ParScale` is to predict intra-particle target properties (e.g., the temperature) as a function of time and space in a spherical particle and a fixed grid consisting of equidistant grid points. The governing equations for the relevant transport phenomena within each particle are outlined in the next section.

2.1 Transport within a particle

To illustrate a typical transport equation to be solved by `ParScale` within a single particle, the Fourier equation in spherical coordinates with $\lambda_{eff} = const.$ is considered:

$$\rho \, c_p \, \frac{\partial T}{\partial t} = div(\lambda_{eff} \, grad \, T) + s_T \tag{1}$$

where ρ is the density, c_p is the heat capacity, T represents the target property profile, t is the time, λ_{eff} is an effective conductivity (e.g., for heat), and s_T is a volume-specific

source term (e.g., for thermal energy). By introducing the thermal diffusivity

$$a = \frac{\lambda_{eff}}{\rho\, c_p} \tag{2}$$

and for constant thermal conductivity λ_{eff}, Eqn. (1) can be re-written as

$$\frac{\partial T}{\partial t} = a \cdot \left(\frac{\partial^2 T}{\partial r^2} + \frac{2}{r}\, \frac{\partial T}{\partial r} \right) + \frac{s_T}{\rho\, c_p} \tag{3}$$

where r is the radial coordinate.

This partial differential equation (PDE) can be discretized in space, e.g., using a central-differencing scheme, in the spirit of the so-called method of lines. The resulting system of ordinary differential equations (ODEs) needs to be solved using a robust integration approach, since the source s_T might lead to a stiff behavior of the PDE. Specifically, we have chosen the flexible multi-step, variable-order solver CVODE, which uses a (modified) Newton-iteration approach to robustly integrate stiff systems of ODEs. CVODE contains a plurality of direct and iterative linear solvers for solving the resulting matrix-algebra problems, and is actively maintained by the Lawrence Livermore National Laboratory (LLNL, U.S.A) as part of the SUNDIALS package [5]. ParScale inherits the flexibility of CVODE, and hence can handle quickly changing environmental conditions, or fast, strongly exothermal reactions in porous particles.

For the second verification example (see Section 3.2), we will consider a single heterogeneous reaction in a porous particle. The corresponding transport equations are presented in the next section.

2.2 Chemistry model

The following equations model the mole-based reactive species balance equations in a particle with constant porosity ϵ. We have adopted the notation of Noorman et al. [6], and from which we have extracted typical system parameters for a relevant application. Here we focus on a single chemical reaction, which is considered to be irreversible and involves a solid species A, as well as a gas species B educt that forms a solid product C and a gaseous product D:

$$aA(s) + bB(g) \rightarrow cC(s) + sD(g) \tag{4}$$
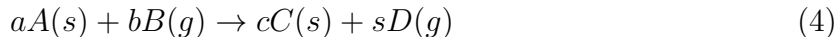
Relevant real-world examples following this scheme are (i) the reduction of iron oxides by H_2, (ii) the oxidation of ZnS and FeS with O_2 to metal oxides (i.e., ZnO, Fe_2O_3), or (iii) the combustion of coal with a high ash content. The mole balance equation for each species in the porous particle is written as:

$$\frac{\partial \varepsilon c_i}{\partial t} = -\frac{1}{r^2} \frac{\partial}{\partial r}(r^2 N_i) + s_i \text{ with } i = B, D \tag{5}$$

Here ε is the phase fraction, c_i is the gas concentration (in $kMol$ per m^3 gas volume), N_i the (convective and diffusive) flux, and s_i is the chemical source term. A typical model for s_i is

$$s_i = \sum_{j=1}^{NR} \nu_{ij} \cdot \prod_{k}^{NG+NS} c_k^{n_{k,j}} \tag{6}$$

where NR is the number of reactions, NG is the number of gas components, ν_{ij} is the stoichiometric coefficient of species i in reaction j, NS is the number of solid species, and $n_{k,j}$ is the reaction exponent. The mole balance equations for the solid phase are similar, however, exclude the flux and are based on molar concentrations in $kMol$ per m^3 total volume:

$$\frac{\partial c_i}{\partial t} = s_i \text{ with } i = A, C \tag{7}$$

For spatial discretizing a second order central differencing scheme is used. For further information about the underlying equations, available models and discretizion schemes the interested reader may refer to the online documentation in the public repository of `ParScale` [4]. The next section briefly outlines the run modes of `ParScale` including the coupling to the open-source DEM-based solver `LIGGGHTS`®.

2.3   Parallel coupling strategy

Besides a stand-alone mode the current development state of `ParScale` provides coupling capabilities to `LIGGGHTS`® and selected solvers of `CFDEMcoupling` in parallel. The key idea is that `ParScale` acts as a slave to the master (i.e., `LIGGGHTS`®), and can exchange its data containers between individual processes as requested by the master. In this paper we will only focus on the coupling to `LIGGGHTS`®. The coupling to `CFDEMcoupling` is handled via `LIGGGHTS`® data structures, and hence is in fact unproblematic. Figure 1 illustrates the underlying coupling algorithm.

At every timestep t_n `LIGGGHTS`® advances the particle position and velocity (and other integral particle quantities if desired) in the simulation domain. After this computation is finished, the coupling is realized by updating the particle surface temperature ($T_{surface}$) directly, or (alternatively) a heat transfer coefficient α together with the fluid temperature in the vicinity of the particle. In addition, a conductive heat flux due to particle-particle collisions ($\dot{q}_{cond}$) can be imposed as well. `ParScale` initializes from the last time step t_{n-1}, and calculates all internal property fields according to the imposed boundary conditions. Additional coupling options are available, e.g., `LIGGGHTS`® is able to push a particle-unique environment temperature (i.e., fluid temperature) to `ParScale`. This enables `ParScale` to react due to changes in the environment temperature, e.g., if the particle enters a region if a fixed temperature. Furthermore, the coupling provides the option to reset the value of the target property to a certain value. Due to the automatic sub-time
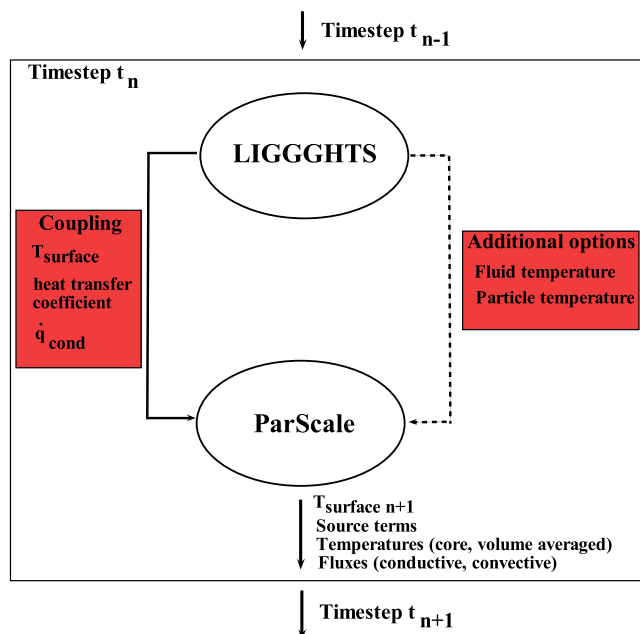
Figure 1: Coupling between `LIGGGHTS`® and `ParScale` for one timestep including required coupling parameters and additional coupling options.

stepping of `CVODE`, the internal `ParScale` timestep does not necessarily has to correspond to the DEM timestep from `LIGGGHTS`®. After `ParScale` completed its calculation of the surface temperatures of all particles, these temperatures are updated and taken into account by `LIGGGHTS`® at timestep t_{n+1} when computing conductive fluxes. Furthermore, source terms due to reactions, the core and the volume-averaged temperature and surface fluxes handed over to `LIGGGHTS`®.

Next, two verification cases, i.e., transient cooling of a sphere and a heterogeneous reaction, is presented.
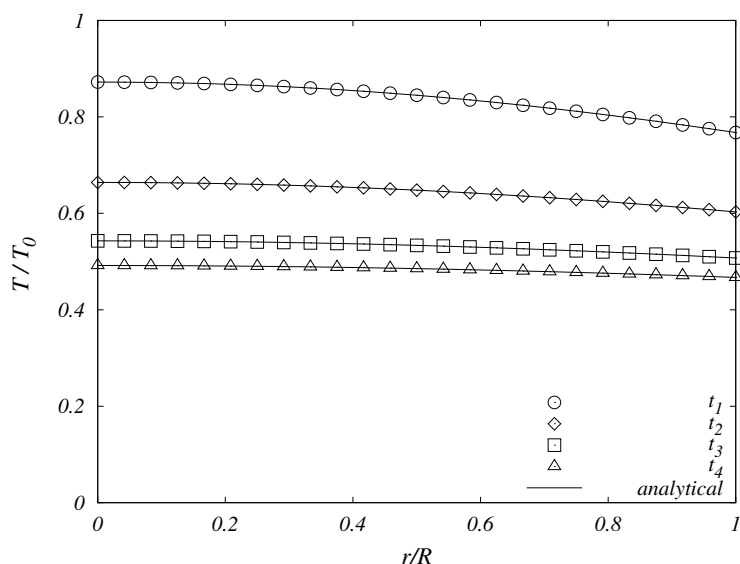
3 Verification cases

3.1 Cooled sphere

The first verification case considers a classical situation in which a spherical particle (initially having the uniform temperature T_0) is convectively cooled by an ambient fluid with temperature T_{enviro}. Table 1 summarizes the parameters of this case. Figure 2 illustrates the comparison between the numerical solution by `ParScale` and the analytical solution provided by [7] for a number of time coordinates.

As expected, excellent agreement (i.e., an average error of 10^{-6}, and a maximum error of 10^{-5}) between analytical and numerical solution can be found.

Table 1: Parameters for the verification case 'cooling of a sphere'.

c_p	300	$[\mathrm{J\,m^{-3}\,K}]$
ρ	1000	$[\mathrm{kg\,m^{-3}}]$
λ_s	1	$[\mathrm{W\,m^{-1}\,K^{-1}}]$
α_p	100	$[\mathrm{W\,m^{-2}\,K^{-1}}]$
r_p	$5 \cdot 10^{-3}$	$[\mathrm{m}]$
T_0	800	$[\mathrm{K}]$
T_{enviro}	300	$[\mathrm{K}]$
t_1, t_2, t_3, t_4	2,5,8,10	$[\mathrm{sec}]$

Figure 2: Numerical (symbols) and analytical results (lines) for the temperature distribution in a convectively cooled sphere at $t_1 = 2$ s, $t_2 = 5$ s, $t_3 = 8$ s, and $t_4 = 10$ s.

3.2 Heterogeneuous reation

This verification case considers a single reaction, and follows the analytical solution provided by Wen [8] for a relative reaction speed (characterized by the Thiele Modulus) of ≈ 3.16. All parameters are chosen following the copper oxidation case considered in Noorman et al. [6]. The three basic assumpions are (i) an isothermal particle, (ii) a reaction rate that is first-order with respect to the gas-phase species, (iii) and a reaction of: $2\,CO + O_2 \rightarrow 2\,CuO$. Figure 3 shows a comparison of the solid and fluid concentration inside the particle for two characteristic times.

Figure 3 (a) shows an excellent agreement for the early stage of the reaction. The small, but noticable, differences in the later stage (see Figure 3 (b)) are due to the pseudo-steady state assumption that needs to be adopted when deriving the analytical solution provided
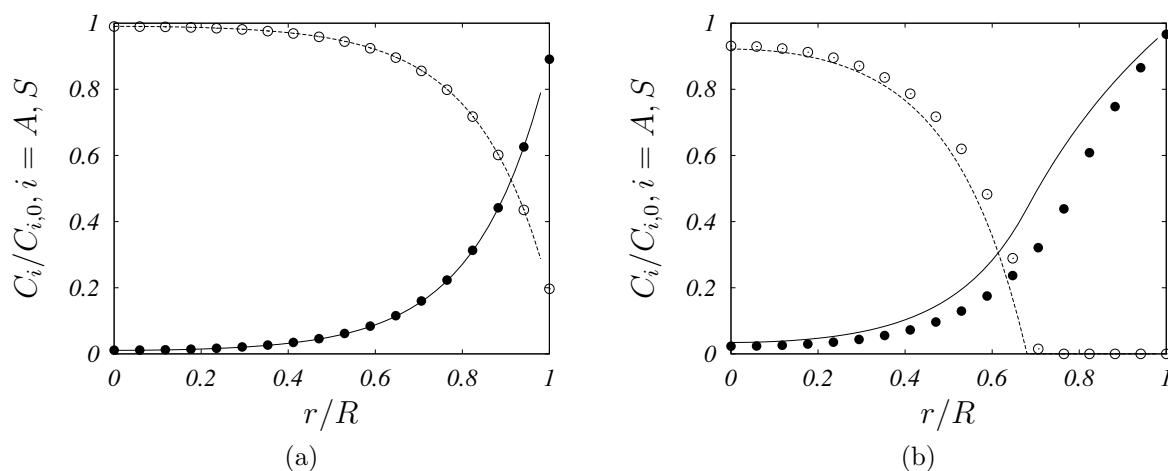
(a)   (b)

Figure 3: Solid and gas-phase concentration profiles inside a porous copper particle for the early stage (a) and the late stage (b) of a heterogeneous reaction (lines: analytical solution, symbols: predictions by ParScale).

by Wen [8]. Figure 4 shows the overall conversion, again indicating only minor differences that can be explained by the shortcomings of the analytical solution. The comparison of the analytical and numerical solution shows good agreement, and the mean difference is below 2 %. We note here that previous work [6] came to a similar conclusion.
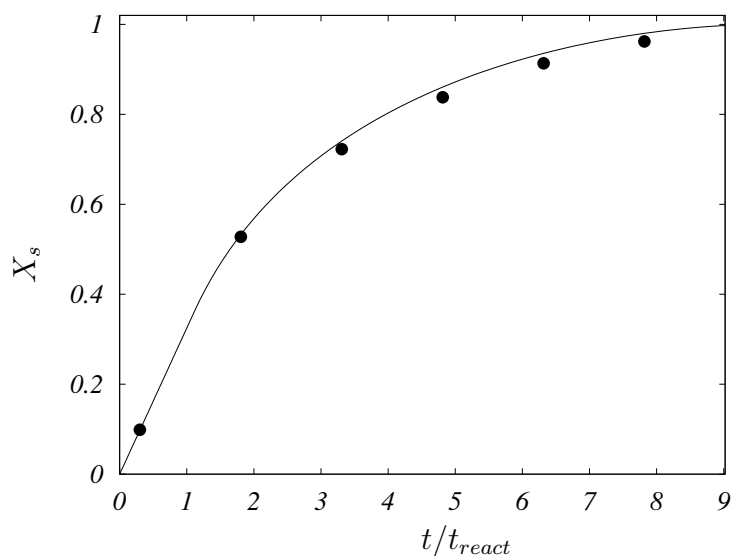

Figure 4: Conversion characteristics during a typical oxidation cycle of a porous copper particle (line: analytical solution, dots: prediction by ParScale).

4 Simple shear flow

We now investigate the influence of `ParScale` under well-controlled flow conditions. Therefore, particles are placed in a cubic periodic box ($H/d_p = 15$) at various particle volume fractions $\phi_p = (0.3 - 0.64)$. LeesEdwards boundary conditions are applied to drive a homogeneous shear flow, which is typically used in studies of granular rheology [9]. In the current contribution, the shear gradient is pointing in the y-direction, and we analyze all quantities of interest (e.g., the conductive flux) only in this direction. Along with the shear gradient, a temperature gradient is applied. We determine the Biot (Eqn. 8) and the Peclet (Eqn. 9) number as the two main non-dimensional influence parameters as already mentioned by [10]. With the usage of `ParScale` at low Biot numbers, we expect that our results agree with previous work [10]. However, for higher Biot numbers the transferred flux to the ambient fluid is much larger than that sustained by conduction inside the particle. Consequently, the influence on the heat transfer rate on the particles' shell temperature, and hence the conductive flux becomes important. Therefore, we expect that intra particle temperature profiles should be considered above some critical Biot number Bi_{crit}.

$$Bi_t = \frac{\alpha \, d_p}{\lambda_p} \tag{8}$$

where α is the heat transfer coefficient that characterized the rate of cooling by the ambient fluid.

$$Pe = \frac{(d_p/2)^2}{\lambda_p \, / \, \rho_c \, c_p} \cdot \dot{\gamma} \tag{9}$$

where $\dot{\gamma}$ is the shear rate. The conductive reference flux is expressed as [9]

$$\dot{q}_{cond,ref} = -\lambda_p \, (\partial_y \, T)_{middle}. \tag{10}$$

where $\dot{q}_{cond,ref}$ is the reference conductive heat flux and y_{middle} is the length of the region over which the temperature gradient is applied. Table 2 shows the main non-dimensional parameters of the sheared bed simulation. All other parameters, e.g., the particle stiffness, the coefficient of restitution and time step are in agreement with our earlier simulations [10].

Table 2: Dimensionless properties of the sheared bed particle case.

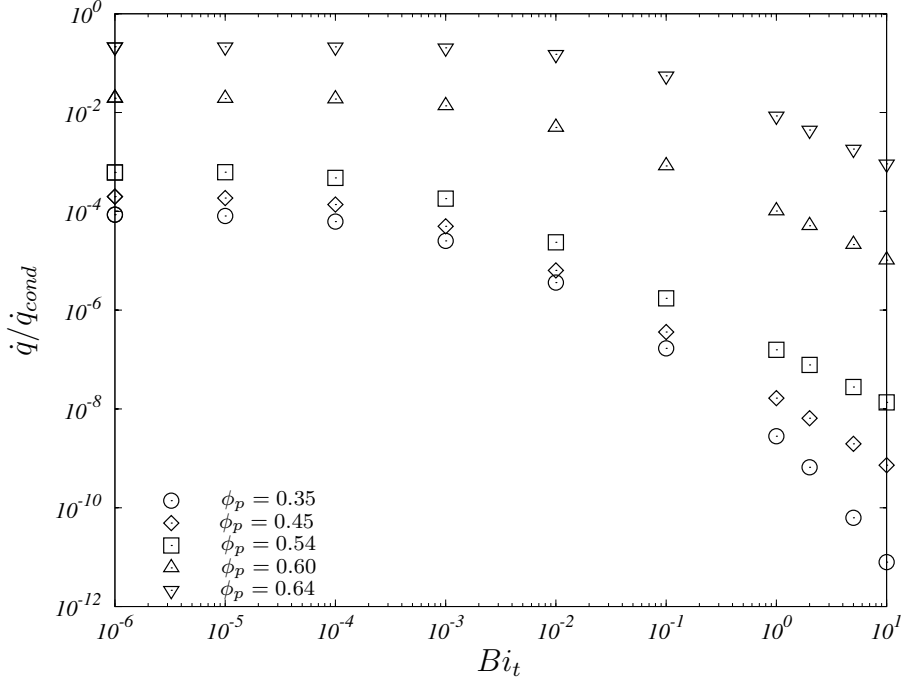|	
ϕ_p	0.3...0.64
Bi_t	$10^{-6}...10$
Pe	0.25

Figure 5: Effect of the Biot number on the conductive heat flux in the gradient direction for various particle volume fractions.

It can be seen in Figure 5 that the influence of the Biot number on the overall conductive flux needs to be considered above a certain value for the Biot number, which depends on ϕ_p. Even at low Biot number regimes (i.e., $Bi_t \approx 10^{-3}$) the conductive flux is under-predicted by up to 10 %. For the highest particle volume fractions considered, for which conductive fluxes are most relevant since they are comparable to the particle-convective flux, the critical Biot number is $Bi_{crit} \approx 10^{-2}$. Above this Biot number intra particle profiles should be taken into account for the moderately fast sheared particle bed that we considered. It is also shown that the influence of the Biot number on the conductive heat flux is becoming more important in case the particle volume fraction is decreasing. Thus, the slope of the curve indicating the relationship between Bi and q_{cond} is becoming larger with decreasing ϕ_p for the high Bi regime. However, in the rather dilute flow at which these extreme dependency is observed, the conductive flux is negligibly small compared to the particle-convective flux.

5   Conclusions

We presented a novel open-source simulation tool `ParScale` which is published under LGPL licence and can be linked to any particle-based solver. We outlined the coupling to the open-source DEM solver `LIGGGHTS`® and demonstrated the usage of `ParScale` with selected verification cases. A good agreement is found between numerical results produced

by `ParScale` and analytical solutions available in literature. We extended the analysis of the well-known sheared particle bed by taking intra-particle temperature gradients into account. A key result is that even at comparably low Biot numbers the intra-particle property profiles have a substantial influence on the conductive flux. The physical reason is that the particle surface temperature is lower than the particle-average temperature in case cooling by the ambient fluid is taken into account. This leads to smaller surface temperature differences in the event of a particle-particle collisions. Thus, the transferred heat flux to the environment should be considered when predicting the particle-particle conductive fluxes. This is especially true for high particle concentrations and fast cooling conditions, since the conductive flux for low particle concentrations is anyhow very low. The current study was limited to selected particle volume fractions and Peclet numbers. Future work will consider wider ranges of these parameters.

6   Aknowledgement

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