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# Pore-resolving simulation of char particle gasification using micro-CT

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ABSTRACT

Understanding the interaction between transport, reaction and morphology at the scale of individual char particles is important for optimizing solid fuel gasification and combustion processes. However, most particle-scale models treat porous char particles as an effective porous continuum, even though the presence of large, irregular macropores, voids and fractures render such upscaled treatments mathematically invalid, and the models non-predictive. A new modeling framework is therefore proposed to elucidate the impact of morphology on char particle gasification and combustion. A pore-resolving, transient, three-dimensional simulation for gasification of a realistic coal char particle is developed based on X-ray micro-computed tomography (micro-CT). The large macropores and voids resolved by micro-CT are explicitly represented in the particle's geometry and conservation equations based on first principles are solved in those regions. Upscaled, effective-continuum equations are applied only within the micro- and meso-porous grains surrounding the voids, where such equations are mathematically appropriate. To assess the impact of the realistic particle morphology, a second model which employs effective-continuum equations everywhere and assumes spherical symmetry is also developed for a particle having the same initial mass, volume, porosity, surface area and equivalent diameter as the pore-resolving model. The results indicate that large, irregular voids enhance mass transport throughout the particle and affect its overall conversion behavior when reactions occur under intra-particle diffusion control.

## 1. Introduction

The gasification and combustion behavior of individual coal char particles affects important outputs at the reactor-scale, such as carbon conversion [1] and temperature [2]. In entrained flow gasification [3] and combustion [4] processes, char particles often react under zone II conditions, in which both reaction and transport through the particle's pore structure influence the char consumption rate. For zone II conditions, it has been convincingly demonstrated that particle morphology has a strong impact on conversion rates and cannot be accurately simulated assuming spherical, homogeneous particles [5]. Therefore, strategies to optimize thermochemical conversion processes via improved reactor design and operating conditions require a fundamental understanding of the interplay between reaction, transport and morphology at the scale of the individual porous char particles.

Since it is impossible to solve conservation equations for reaction and transport within the actual char structure and its myriad pores, physics and chemistry at the scale of these geometrical heterogeneities are typically averaged, or "upscaled". Upscaling (e.g. via homogenization [6] or volume averaging [7]) transforms equations based on first principles and pore-scale geometries into computationally tractable, effective-continuum equations, which contain effective properties that are dependent on the porous medium's characteristics.

For mathematical validity and accuracy, upscaling methods require a separation of length scales [8,9]. This implies that the characteristic length-scale of the geometrical heterogeneities (e.g. pores) must be significantly smaller than that of the system (e.g. particle), as well as the characteristic length of the physical processes being studied (e.g. concentration gradients) [9]:

$$L_{pores} << L_{particle} \tag{1a}$$

$$L_{pores} << L_{gradient} \tag{1b}$$

Inequality (1a) implies that upscaling is meaningful only if there exists a statistically significant sample of heterogeneities over which to average (a representative volume element), but treating heterogeneities with sizes approaching that of the macroscopic domain as an effective-continuum is not valid. Similarly, if the characteristic length-scale of the concentration or temperature gradients becomes nearly as small as the length-scale of the geometric heterogeneities, local averaging is impossible and inequality (1b) is violated.

For char particle gasification and combustion, the length-scale constraints are particularly acute [8,10]. Across a range of coals and devolatilization conditions, char particles contain large macro-pores,

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**Nomenclature**

| | |
|---|---|
| $A$ | pre-factor exponential |
| $B$ | permeability |
| $D$ | diffusion coefficient |
| $d$ | diameter |
| $E_a$ | activation energy |
| $h$ | enthalpy |
| $\vec{J}$ | diffusion flux |
| $k$ | thermal conductivity |
| $MW$ | molecular weight |
| $m$ | mass |
| $P$ | pressure |
| $R_i$ | rate of production of $i^{th}$ Species |
| $R_u$ | universal gas constant |
| $r$ | radius |
| $S$ | surface area per unit volume |
| $S_g$ | specific surface area (per unit mass) |
| $S_f^h$ | fluid enthalpy source |
| $S_m$ | mass source term |
| $T$ | temperature |
| $t$ | time |
| $\vec{v}$ | velocity |
| $\nu$ | atomic diffusion volume |
| $V$ | volume |
| $X$ | conversion |
| $x$ | mole fraction |
| $Y$ | mass fraction |
| $\varepsilon$ | porosity |
| $\vartheta$ | stoichiometric coefficient |
| $\mu$ | dynamic viscosity |
| $\rho$ | density |
| $\tau$ | tortuosity |
| $\bar{\bar{\tau}}$ | stress tensor |
| $\psi$ | pore structure parameter |
| $\eta$ | effectiveness factor |
| $\mathfrak{R}$ | reaction rate (intrinsic) |

*Subscripts*

| | |
|---|---|
| $2D$ | effective continuum model |
| $3D$ | pore-resolved model |
| $c$ | carbon |
| $eff$ | effective |
| $equiv$ | equivalent (diameter) |
| $i$ | $\mathrm{i}^{\mathrm{th}}$ Species |
| $j$ | $\mathrm{j}^{\mathrm{th}}$ Species or Reaction |
| $knud$ | Knudsen |
| $m$ | mixture |
| MPS | microporous solid |
| $n$ | reaction order |
| $o$ | initial |
| $p$ | pore |
| $rxn$ | reaction |
| $s$ | solid |
| $scale$ | scale factor |
| $t$ | true (density) |
| $T$ | thermal |
| $tot$ | total |

voids and fractures with dimensions that approach the scale of the particles themselves [10,11]. This results in violation of the length-scale constraints and renders an effective-continuum treatment invalid and non-predictive. Application of an effective-continuum model when the length-scale constraints are not satisfied can result in the failure to predict the extent of heterogeneous reactions [12], mixing [13] and localized phenomena such as hotspots [14].

Despite their general inapplicability, the vast majority of spatially-resolved char particle gasification and combustion models simply ignore the length-scale constraints and employ spherically symmetric, effective-continuum models. Although such an approach might be applicable to synthetic char particles, which are spherical, uniformly porous and lack large voids [15], or to hypothetical spherical particles with a single spherical void [16], the effective-continuum approach is not applicable to realistic void and large macropore morphologies. For this reason, effective-continuum simulations often require adjustable parameters to match experimental data.

This issue has been recognized by several investigators and has led to discrete representations of porous char particles, using Monte Carlo or random walk methods to analyze char conversion [8,10,17,18]. However, the fidelity with which discrete networks represent actual char particles is questionable, and the discrete approach is difficult to combine with models of other physical and chemical processes, such as mass transfer, heat transfer and reaction in the surrounding gas.

Two recent studies have developed simulations of char particle combustion that resolve large pores/voids, but both are based on idealized pore structures and treat the char surrounding the voids as completely non-porous. Richter et al. simulated oxy-combustion of a porous coal char particle by treating the particle as an agglomerate of 185 monodisperse, non-porous spheres, each with a diameter of 25 µm [19]. The void-space between the spheres was meshed and the reacting flow problem was solved using ANSYS FLUENT. Calculations for the entire agglomerate were compared to a non-porous particle, and enhanced reaction rates were observed for the agglomerate. The agglomerate model was later used to calculate drag coefficients and Nusselt numbers for porous particles [20]. Xue et al. used a very similar approach to study oxy-combustion of particles with cone-shaped pores extending from the surface toward the particle center [21]. The char surrounding the idealized pores was also treated as non-porous.

In the context of non-reacting simulations, micro-CT has been used to image coal macro-pores larger than 13.85 µm for flow simulations using COMSOL [22]. Ciesielski and coworkers used confocal scanning laser microscopy on sections of wood particles to obtain microstructural parameters characteristic of the pore structure [23]. The data was used in a constructive solid geometry algorithm to generate three particle geometries which were used in simulations of heat transfer and mass transfer using COMSOL [23,24]. It was concluded that intra-particle transport is not adequately represented using effective-continuum models.

This paper presents the first pore-resolving, reacting flow simulation for char particle conversion based on a realistic char particle. The approach resolves large pores and voids, circumventing difficulties associated with upscaling large heterogeneities, which have prevented previous effective-continuum treatments from being truly predictive. A second novelty of the current approach is the combination of the void-resolving simulation with an effective-porous-continuum treatment of the micro/meso-porous grains surrounding the resolved pores, in contrast to [19,21], where the char grains surrounding the idealized resolved pores were assumed to be completely non-porous. In the transient, 3-D simulation approach outlined below, the large macropores and voids are explicitly represented in the particle's geometry using micro-CT, and conservation equations based on first principles are solved in those regions, as shown in Fig. 1. Effective-continuum equations are employed only within the micro/meso-porous grains (referred to hereafter as "microporous", although these regions contain micro-, meso- and macro-pores), where they are mathematically appropriate