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Development of a mesoscopic particle model for synthesis of uranium-ceramic nuclear fuel

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ABSTRACT

A novel technique has been developed to fabricate uranium-ceramic nuclear fuel using the depleted uranium (DU), U_3O_8 powder, and allylhydridopolycarbosilane (AHPCS) polymer precursor. This process involves a continuous change of the composition, porosity, and material properties. To fabricate nuclear fuel with a uniform structure/volume ratio, it is important to understand the transport phenomena during high temperature processing and at different length scales. In our prior work, a system-level model based on the reactive porous medium theory was developed to account for pyrolysis process during the uranium-ceramic fuel fabrication. In this paper, a mesoscopic model based on the smoothed particle hydrodynamics (SPH) is developed to simulate the synthesis of filler U_3O_8 particles and SiC matrix. The system-level model provides the necessary thermal boundary conditions for the mesoscopic simulation. The evolution of the particle concentration and the structure as well as the composition of the composite produced is investigated. Since the process heat flux plays an important role in the material quality and uniformity, the effect of the heating rate, filler particle size and distribution on the uniformity and the structure of the final product are investigated. The uncertainty issue is also discussed.

1. Introduction

A novel preparation technique for the nuclear fuels starting from the depleted uranium (DU), U_3O_8 powder, and allylhydridopolycarbosilane (AHPCS) polymer precursor has been developed in our prior study [1]. This processing cycle involves the curing of the polymer precursor mixed with the U_3O_8 particles at 150–250 °C, in which the polymer undergoes cross-linking to form a green body, followed by a pyrolysis stage which involves the formation of the amorphous SiC between 400 °C and 900 °C, the crystallization of SiC and the synthesis of SiC and U_3O_8 at 1000–1500 °C, UC can be produced in the synthesis. This processing technique allows the uniform distribution of the uranium fuel source along with a high ceramic yield of the parent matrix. The processing technique offers an inexpensive method to obtain a dense nuclear fuel. The resulting ceramic composite has the uranium oxide well dispersed in *a*-SiC matrix. The thermal conductivity of the composite can be improved through the crystallization owing to the high conductivity of SiC [2]. Further mechanical consolidation was achieved using the polymer infiltration process (PIP). Among various advanced ceramics, the silicon carbide has been proven to be a promising candidate for use in the nuclear applications as an inert-matrix material due to its characteristics in the following aspects. First, SiC displays a number of superior characteristics as a structural material, such as its thermal and mechanical properties, chemical stability and low radioactivation. Second, the irradiation effects on swelling and mechanical properties of high purity SiC have been evaluated, and the excellent high temperature irradiation resistance was revealed by several researchers [3,4]. Third, the SiC matrix composites have been widely developed for use in the advanced energy systems [5–7]. It has also been recognized that the uranium carbide (UC) is an attractive fuel for certain generation IV reactors such as the gas-cooled fast reactor due to its high thermal conductivity and high melting point. Therefore, the ceramic composite pellets consisting of the UC particles in the SiC matrix can be used as a high-temperature refractory ceramic fuel due to the superior characteristics of UC and SiC.

The pyrolysis and synthesis processing technique allows the fabrication of various ceramic materials at a relative low temperature. Also, it allows the fabrication of the net-shape components without suffering from the size limitation [8]. During the powder synthesis, the powder binding mechanism strongly depends on the local temperature and heat flux. Meanwhile, the morphological changes at the particle scale will influence the thermal transport and in turn, affect the local temperature. Several numerical models have been developed, which can be generally categorized into two approaches: the porous media based approach and the particle

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Nomenclature

C_p	Specific heat at constant pressure (J/Kg K)
D	Diffusivity (m^2/s)
D	Particle diameter (m)
F	Volume ratio
k_s	Thermal conductivity (W/m K)
r_{ij}	Distance between particle i and j (m)
m	Mass (kg)
$\dot{m}$	Mass generation rate (kg/s)
N	Number of particles at the neighboring
P	Gases products
q''	Heat flux (W/m^2)
$\dot{Q}$	Reaction heat (kg/s)
T	Temperature (K)
t	Time (s)
$\vec{u}$	Velocity vector (m/s)
W	Smoothing function
Y	Mass fraction
$\dot{Y}$	Generation rate of mass fraction

Greek symbols

α	Coefficient
ρ	Density (kg/m^3)
μ	Viscosity (kg/m s)
ν	Kinematics viscosity (m^2/s)

Subscripts

1	SiC
2	Filler particles
g	Gas phase
i	Particle i
j	Particle j
s	Solid phase
β	Coordinate direction

based approach. The porous media based approach is based on the porous media theory to describe the macroscopic property evaluation during the synthesis or sintering process. Tarafdar and Bergman [9] developed a sintering model to investigate the variations of the local morphologies on the effective transport properties. They found that the penetration depth of the sintering was strongly related to the thermal penetration depth and boundary temperatures. Chen and Zhang [10] developed a partial shrinkage model for the selective laser sintering (SLS) of two-component metal powder and they discovered that the sintering depth and volume of the HAZ was significantly increased with the decrease of the gas volume fraction. Xiao and Zhang [11] studied the melting of an alloy powder bed. It was concluded that the increase of the initial porosity of the powder bed can shorten the duration of the preheating and increase the surface temperature and interfacial velocity. He et al. [12] presented a model to study the effects of the particle convection, gas convection and radiation. They concluded that the radiative heat transfer provided a significant contribution to the total heat transfer, and the particle and gas convections show an opposite tendency to the total heat transfer. Kaushal et al. [13] developed a model for the biomass char combustion in a fluidized bed. It is shown that a smaller size of wood or char benefits the overall performance of the gasifier.

Several particle based models are developed for mesoscopic phenomena in the materials processing or underground water flow focusing on the local behaviors around the particles and studying the evolution of the structure and the heat and mass transfer on the particle level. Jagota and Dawson [14] and Martinez-Herrera and Derby [15] used the finite element methods (FEM) to study the shape evolution of the adjacent sintering particles. The effect of particle geometry on the sintering rate is also investigated. It was concluded that the assumption of spherical particle shape is reasonable to predict sintering dynamics for many different particle geometries. He et al. [12] proposed a one dimensional char combustion model considering the pore structure to investigate the effect of the particle size on the transport phenomena in the single char combustion. The simulation results show that the larger particles generally have higher temperatures at the exterior surface, and the smaller particles have the better combustion efficiency in a limited period. Gao et al. [16] presented a double particle-layer and porous medium model to simulate the gas flow and the heat transfer near the surface of an immersed object in a fluidized bed. They concluded that the heat transfer rate is determined by the gas velocity. The heat transfer is conduction dominated in the stifling region since the gas is almost stagnant there. Tartakovsky and Meakin developed a numerical model to simulate the multiphase flows and the miscible flows in the fractured porous media using the meshless particle SPH method [17]. The effects of the pore level heterogeneity and anisotropy on the immiscible and miscible flows were studied [17]. Tartakovsky et al. [18] further improved their model to study the reactive transport and the mineral precipitation in the porous media. They concluded that the initial perturbation of the solid surface leaded to the unstable growth and the increase of the reaction length parameter can stabilize the non-uniform surface growth.

A macroscopic model for the pyrolysis processing was developed in our prior work [19] in which the Darcy's law and the convection–diffusion equations were used to model the volatile flow in the porous media. The system-level or global model describes the polymer pyrolysis processing, including heat transfer, polymer pyrolysis, SiC crystallization, chemical reactions, and volatiles transport. However, the modeling the particle level structure evolution of the composite produced during synthesis of the uranium oxide and SiC is needed for the process optimization. In this paper, a mesoscopic model is developed based on the meshless particle method to study the structure and species evolution at the mesoscopic or local level. The mesoscopic model is applied to a repetitive unit cell and describes the mass transport, the composition change and the movement of the particles. The unit cell represents a unit of the source material that consists of several U_3O_8 particles, the SiC matrix and other components produced from synthesis. The smoothed particle hydrodynamics (SPH) technique is employed to solve the solid–solid reaction between the SiC matrix and the filler particles, the composition changes, the uniformity of the resulting products, and the shrinkage and the motion of the U_3O_8 particles in the SiC matrix. The results from the macroscale simulation are used to provide the necessary thermal boundary conditions for the particle level simulations. The evolution of the structure and the composition of the produced composite due to the heat fluxes are monitored. The effects of the heating rate and the U_3O_8 particle size and volume on the species uniformity and the structure formation are investigated. The uncertainty due to the random distribution of the filler particles is also investigated.

2. Physical and mathematical model

Fig. 1 shows the key elements of the SPH local model within a macro computation domain that is constituted by many