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Optimization of heating profile for densification of fuel pellets using Monte Carlo simulation

Tetsushi Matsuda [a,*], Hiroaki Muta [b], Kosuke Tanaka [c]

[a] Japan Fine Ceramics Center, 2-4-1 Mutsuno, Atsuta-ku, Nagoya, Aichi 456-8587, Japan
[b] Osaka University, 2-1 Yamadaoka, Suita, Osaka 565-0871, Japan
[c] Japan Atomic Energy Agency, 4002 Narita-cho, Oarai-machi, Ibaraki 311-1393, Japan

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ABSTRACT

Computational simulation based on the Monte Carlo model was performed for optimization of a sintering process to obtain UO_2 pellets with higher density, and as a guideline for design of the heating profile based on conventional two-step sintering (TSS). The optimized simulation parameters were obtained using the experimental shrinkage-curve for UO_2 in a reducing atmosphere. The calculation results sufficiently replicated shrinkage curves measured for different initial grain sizes. The optimized heating profile for more efficient densification than conventional heating included a faster heating rate, holding at a relatively low temperature as the second step in TSS, and raising the temperature again.

1. Introduction

Uranium dioxide (UO_2) is a material of significant interest due to its application in nuclear reactors. UO_2 fuel pellets for commercial reactors are fabricated by the sintering of powder compacts. The grain size and the density of UO_2 pellets must satisfy certain fuel specifications. Therefore, the sintering of fuel pellets has been studied for commercial applications. Grain coarsening techniques that could reduce fission gas release from fuel pellets during irradiation [1] have been studied with the use of additives or by control of the sintering atmosphere [2,3]. The sinterability of UO_2-Gd_2O_3 powder compacts has been studied by many researchers [4,5] because UO_2-Gd_2O_3 fuel pellets are widely used as a burnable absorber. The motion of the grain boundary and the densification of fuel pellets during irradiation have been studied to elucidate the in-pile behavior of fuel pellets [6]. The sintering behavior of (U,Pu)O_2 mixed oxide (MOX) fuel was studied after 1990, from which problems regarding the homogeneity and density of fuel pellets were recognized [7]. Thus, further studies on the sintering process of fuel pellets are required in the field of nuclear energy.

The sintering phenomena of metal or ceramic powders have been of significant interest since the 1950s [8,9]. The initial, intermediate, and final stages of sintering have been modeled by simplifying the geometrical configuration of particles. While such models are effective to understand the basis of sintering behavior, they are difficult to apply to actual sintering that involves various particle shapes with complex contact conditions among the particles. A study on the sintering of UO_2 was conducted around the same time, which led to clarification of the sintering conditions required to densify the fuel pellets [10]. The effect of the O/U ratio on the sintering kinetics of UO_2 was examined, and the activation energy and diffusion coefficients during the sintering of UO_2 were evaluated [11].

Various sintering processes have been studied by the ceramic industry to obtain sintered bodies with higher densities and smaller grains. In the 1960s, Palmour [12] reported on the importance of the heating profile and proposed the concept of rate-controlled sintering (RCS). Although many studies were subsequently reported, the theoretical background of RCS has yet to be clarified. Chen et al. [13] proposed the concept of two-step sintering (TSS), and demonstrated that TSS is effective to obtain smaller grains. The restriction of grain growth is generally known to lead to increases in the strength of a sintered body. Therefore, TSS is considered to be the most successful method to optimize the sintering process and improve the microstructure of ceramics [14]. Johnson et al. proposed an analytical method to determine the shrinkage curves of compact bodies [15]. They insisted that a master sintering curve (MSC) could be used to characterize the sintering behavior for a given green compact, regardless of the heating profiles [15]. However, their assumption that the rate controlling mechanism of

* Corresponding author.
E-mail address: t_matsuda@jfcc.or.jp (T. Matsuda).

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shrinkage remains the same throughout the entire sintering process was not considered appropriate. Therefore, a two-stage MSC approach was recently proposed to elucidate the sintering kinetics [16].

Aside from experimental approaches, computational methods to understand sintering behavior have been developed using various models over the last three decades. Fig. 1 shows the current progress and characteristics of these models. Various models such as the Monte Carlo (MC) model [17,18], the discrete element model (DEM) [19,20], the phase field model (PFM) [21,22], and the finite element model (FEM) [23] have been developed and utilized. However, the available models have not yet sufficiently simulated the entire sintering process. Therefore, a specific and appropriate simulation method should be selected for a particular purpose.

Although the DEM, PFM, and MC models require the initial microstructure in a compact, it is too difficult to reconstruct a microstructure that is perfectly equivalent to that in an actual compact because this requires precise information on the contact, size and shape of the particles. The initial microstructure typically used in a simulation takes into consideration only the relative density and average particle size of the compact. For this reason, acquisition of a correlation between computational and experimental results for sintering is required. Such a type of tuning method that correlates computational results with experimental results would thus be a key technique to effectively utilize computational simulations.

Among these models, the MC model and PFM can simulate the entire sintering process, with the microstructure taking into consideration. The PFM simulates a sintering process using thermodynamic parameters, and it treats the stress and solid solution during the sintering process. Although the PFM has excellent advantages and potential to progress, it seems to take time for engineering applications because exact thermodynamic data and a powerful computer ability are required to treat a large number of particles in an actual compact. On the other hand, the MC model does not handle stress, and the simulation parameters must be tuned using experimental results. However, it can be said that the MC method is one of the engineering-applicable simulation tools at the present time.

In this study, a computational simulation based on the MC model was conducted for optimization of a sintering process to obtain fuel pellets with higher density, and simulations were performed to obtain a guideline for design of the heating profile based on conventional TSS.

2. Simulation

2.1. MC model

In this study, a three-dimensional MC procedure [24,25] based on the Potts model was used to simulate sintering. The initial microstructure is represented by mapping grains and pores onto a discrete triangle lattice; Fig. 2 shows a cross-section of the three-dimensional microstructure. Each lattice site is assigned a number of state, which represents the grain orientations and pores. A number between 1 and Q is assigned for the grain state, where Q represents possible grain orientations; $Q = 64$ was selected for this study.

Neighboring lattice sites with the same grain orientation are defined as the same grain. The interfaces of neighboring sites with different orientations are defined as having a grain-boundary energy. The interfaces of neighboring sites that consist of grain and pore lattices are defined as having a surface energy. The grain-boundary energy and surface energy of materials affects

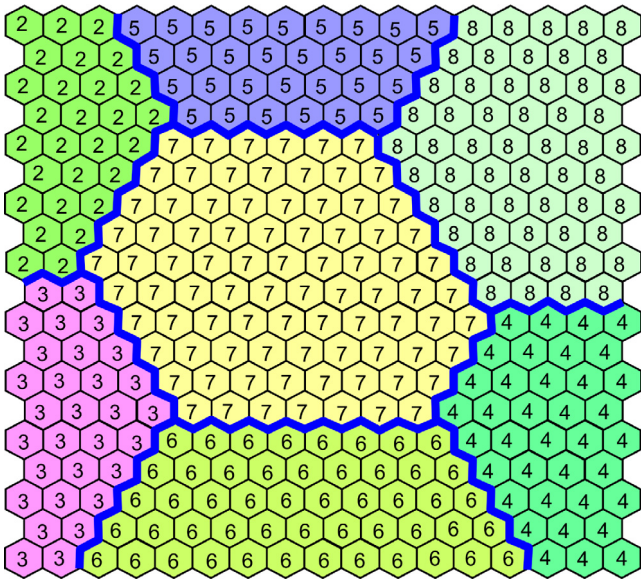

Fig. 2. Schematic of a MC lattice in two dimensions.

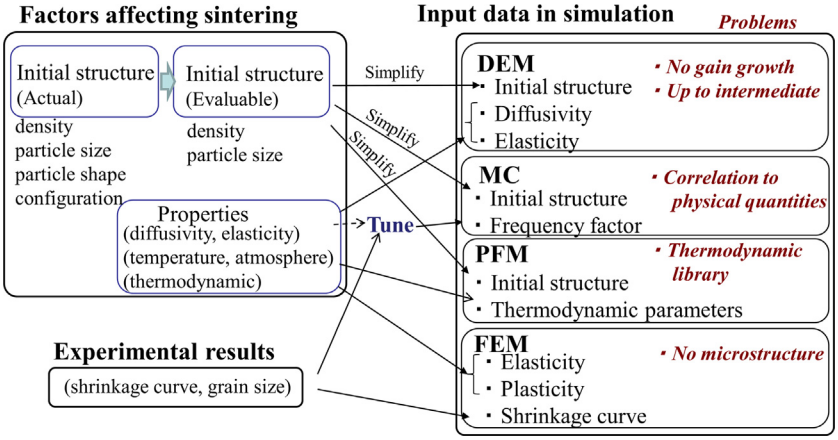

Fig. 1. Research progress of sintering simulations in recent years.