Stochastic homogenization analysis of FIB-SEM image-based hierarchical model of sprayed porous ZrO$_2$

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Abstract

The stochastic prediction of apparent mechanical property of sprayed porous ZrO$_2$ film considering the scattering of Young’s modulus of ZrO$_2$ is the purpose of this study. To take the morphology at nanoscale into consideration, FIB (focused ion beam)-SEM image-based modeling technique was employed. Pores were categorized into dot-like, large slit-like, small slit-like and small miscellaneous pores. Their representative dimensions were statistically measured using FIB-SEM and 2D SEM observation in order to generate larger sized microstructure model for homogenization analysis than the observable region by FIB-SEM. The dot-like pores were homogenized first, and hierarchical model was generated. The first-order perturbation based stochastic homogenization method for hierarchical model of porous material was proposed. The stochastic homogenized Young’s modulus in the thickness direction, which is the spraying direction, was calculated and discussed.

Key words: Sprayed porous material, FIB-SEM, Image-based modeling, Stochastic homogenization method

1. Introduction

Porous materials have many attractive functions in engineering applications such as heat insulation and so on (Soneya, et al., 2015). There have been many published reports on the manufacturing and processing techniques of sprayed coatings (Hanaoka, et al., 2011). To achieve the desired function or to improve the properties, not only the porosity ratio but also microscopic morphology is an important design parameter (Gu, et al., 2001). Therefore, we can find many reports on the observation of microstructures and furthermore on correlation between properties and morphology (Kulkarni, et al., 2005). In addition to 2D observation by SEM, 3D observation has been performed by sequential 2D observation after grinding the specimen (Juzkova, et al., 2004), in which the resolution was 3 $\mu$m.

In this paper, sprayed porous ZrO$_2$ has been investigated by focused ion beam sectioning and SEM observation, FIB-SEM, to study three-dimensionally the nano-sized microstructure. The resolution in the cross-section is 0.0467 $\mu$m x 0.0396 $\mu$m, i.e., approximately 40 nm. Hence, the morphology could be observed very precisely.

The effects of pores on the mechanical and/or thermal properties have been investigated experimentally and numerically (Tan, et al., 2006, Altun, et al., 2011). Wang et al. categorized typical microstructure elements into pores, cracks and voids, and performed 2D finite element analysis (Wang, et al., 2003). Nakamura et al. modeled the pores by ellipse like shape whose major axis is almost perpendicular to the spraying direction in 2D cross-section, and microindentation test was simulated numerically (Nakamura, et al., 2000). This implies that morphology should be studied in association with manufacturing process (Ilavsky, et al., 1997). It should also be noted that Nakamura et al. pointed out that large enough region must be analyzed. This is an important point also in our study on homogenization, because it is a sort of averaging process. From the above literature review, 3D numerical analysis of sprayed porous
ceramic with nanoscale resolution is still a challenging theme.

In this study, the observation by FIB-SEM revealed the characteristic microstructure elements. Very small dot-like pores and various sized slit-like pores were clearly observed. Taking the real microstructure into account, 3D finite element model has been generated for homogenization analysis to predict the macroscopic mechanical property. Here, the following two main problems were studied.

To calculate the macroscopic homogenized property influenced by the microstructure, large enough region should be analyzed. However, FIB-SEM technique had a limitation to the size of observable region. Therefore, the representative dimensions were defined for categorized microstructure elements and measured statistically by 2D SEM images. Based on the statistic data, large sized 3D model was generated. The very small dot-like pore was modeled hierarchically to perform the numerical analysis on a desktop personal computer (PC).

Next, to take the uncertainty of the mechanical property of constituent material due to the manufacturing process into account, stochastic homogenization method was applied to the hierarchical microstructure model in this study.

The two-scale homogenization theory was firstly developed in early 1980s (Lions, 1981), and applied to fibrous composite materials (Guedes and Kikuchi, 1990). Validation has been done for porous ceramics, where the significance of the size of region to analyze was also described (Takano, et al., 2003). Based on this deterministic homogenization analysis, stochastic methodology has been studied. A first-order perturbation based stochastic homogenization (FPSH) method is a simple and easy-to-use technique (Kaminski and Kleiber, 2000, Kaminski, 2007, Sakata, et al., 2008). One of the authors has also successfully applied FPSH method to the prediction of apparent modulus of human vertebral trabecular bone and compared the prediction with many published experimental data (Basaruddin, et al., 2013). This paper presents that FPSH method can easily be applied to the hierarchical model of porous material.

2. SEM and FIB-SEM observation of sprayed porous ZrO₂

The target material is sprayed porous film of ZrO₂ with approximately 1 mm thickness as shown in Fig. 1. In the magnified view, numerous small pores were observed as in the literature review in Introduction. Interestingly, clustered very small dot-like pores were found in the most magnified view. From 2D SEM observation, the pores were supposed to be categorized roughly into dot-like pores and slit-like pores, which seemed to be natural in association with the spraying process as illustrated in Fig. 2. For further precise observation of pores, FIB-SEM was utilized.

In the use of FIB-SEM, there were some limitations. The most critical one was that only 30 μm cubic region was observable due to the limitation of sectioning by focused ion beam. It was also impossible to observe the neighboring 30 μm cubic region. The expense was another hurdle. To this end, 30 μm cubic region that includes both dot-like and slit-like pores as shown in Fig. 1 was selected for FIB-SEM observation and for detailed morphology analysis.

Since the goal of this study was to analyze the macroscopic mechanical property, especially the Young’s modulus in the thickness direction, i.e., z direction in Fig. 1, it was found that approximately 100 μm cubic region was required for homogenization analysis. The challenge in this paper is to generate 100 μm cubic microstructure model based on FIB-SEM observation of 30 μm cubic region. To compensate the incomplete information, statistic analysis was performed for 2D SEM images together with FIB-SEM observation. That is, pores were classified into typical microstructure elements first. Their shapes were defined, and representative dimensions were parameterized. The parameters were statistically measured as described in the next chapter.

3. Reconstruction of larger sized 3D microstructure model based on statistic morphology analysis

3.1 Morphology analysis using FIB-SEM images

Based on 100 sliced FIB-SEM images with resolution of 0.0467 μm x 0.0396 μm, 3D voxel model was reconstructed as shown in Fig. 3. Note that slicing direction is in x, and the thickness direction of sprayed film is in z.

Four typical microstructure elements were found. They were dot-like pores, large slit-like pores, small slit-like pores and miscellaneous small pores. The segmentation was done manually section by section as shown in Fig. 4(a). The segmented 3D model is shown in Fig. 4(b). The total porosity ratio in the 30 μm cubic region was 20.10 %. The proportion of each microstructure element is summarized in Fig. 4(c). The labeling technique, which is one of the image processing techniques, was applied as shown in Fig. 4(d), which revealed that most of the pores are connected. The connectivity was achieved by the contribution of small slit-like pores. In other words, small slit-like pores
connected the dot-like pores and large slit-like pores.

Fig. 1 SEM images of sprayed porous ZrO$_2$. Approximately 100 $\mu$m region should be considered to represent the complex heterogeneity, but 30 $\mu$m region was imaged by FIB-SEM.

Fig. 2 Schematic illustration of formation of pores during spraying process

Fig. 3 FIB-SEM images of 30 $\mu$m cubic region and 3D reconstruction

Resolution: 0.3 $\mu$m $\times$ 0.0467 $\mu$m $\times$ 0.0396 $\mu$m

Fig. 4 Morphology analysis of 30 $\mu$m cubic region where types of pores were segmented manually. The connectivity was investigated by the labeling image processing technique.

3.2 Statistic morphology analysis using 2D SEM images

Dot-like pore was supposed to be specific in the sprayed porous materials, which could be observed by high-resolution observation. The dot-like pore region was assumed to have simplified shape in Fig. 5(a). To characterize this shape, three representative parameters were defined. The correlation between $L$ and $H$ was plotted in Fig. 5(b). The mean value and standard deviation of $L$ were 68.3 $\mu$m and 26.2 $\mu$m, and those of $H$ were 17.6 $\mu$m and 9.5 $\mu$m, respectively. The angle $\theta$ was distributed as in the histogram shown in Fig. 5(c), whose mean value and standard deviation were 6.1 degree and 27.1 degree.

Next, large slit-like pores were assumed to be almost square. Its representative length was measured using 2D SEM images and summarized in a histogram shown in Fig. 6(a). The mean length was 48.0 $\mu$m, and standard deviation was 19.3 $\mu$m. Using a reconstructed large slit-like pore model by FIB-SEM with less than 30 $\mu$m size, variety of
models were generated according to the statistic data. Typical large sized model is shown in Fig. 6(b).

On the contrary, numerous small slit-like pores were found in the FIB-SEM images. Relatively larger sized pores in Fig. 7 were used in the later analysis.

Finally, miscellaneous small pores could be approximated fairly well by sphere. To determine the diameter, the volume of real miscellaneous small pores in FIB-SEM image-based 3D model was plotted in Fig. 8(a). Modeled spherical pores were assumed to be dispersed randomly as shown in Fig. 8(b).

3.3. Generation of 100 µm sized cubic model

With above statistic database of four microstructure elements based on FIB-SEM observation of 30 µm cubic region and 2D SEM images, a microstructure model of 100 µm cubic region was generated manually, so that the volume fraction of each microstructure element was almost the same with the value obtained by FIB-SEM observation.
in Fig. 4(c). Figure 9 shows the manually generated 3D model. The volume fraction of dot-like pore was 12.16 %, those of large and small slit-like pores were 2.57 % and 2.37 %, and that of miscellaneous small pore was 1.51 %.

In the generated 100 μm sized cubic model, the dot-like pore region was replaced by homogenized material model. In Fig. 9, representative volume element (RVE) model is also shown to calculate the homogenized properties. The porosity ratio in RVE is 37.8 %. It was subdivided into 0.0396 μm cubic voxel finite elements. The number of elements is 8,594,648. The Young’s modulus of ZrO$_2$ was $E_{ZrO_2} = 240$ GPa and the Poisson’s ratio was $\nu_{ZrO_2} = 0.3$ in this study. The homogenized properties of dot-like pore region were calculated as isotropic material model whose Young’s modulus, Poisson’s ratio and shear modulus were $E_{\text{dot}} = 50$ GPa, $\nu_{\text{dot}} = 0.22$ and $G_{\text{dot}} = 20.5$ GPa. We denote hereafter the homogenized stress-strain matrix for the dot-like porous region by $[\mathbf{D}]_{\text{dot}}$. The use of homogenized model for dot-like pore region allowed us to discretize the hierarchical model in Fig. 9 by larger sized voxel finite elements. In the next chapter, the scattering of Young’s modulus of ZrO$_2$ is considered to predict the apparent Young’s modulus by deriving the stochastic homogenization formulation for the hierarchical model.

Fig. 9 Generated 3D model of 100 μm cubic region using homogenized model for dot-like pore region

### 4. First-order perturbation based stochastic homogenization (FPSH) analysis

We suppose that the Young’s modulus of ZrO$_2$ is scattered in the form of normal distribution. Its expected value is $\text{Exp}[E_{ZrO_2}] = 240$ GPa and standard deviation is assumed to be $\text{SD}[E_{ZrO_2}] = 7.2$ GPa. It is denoted by the following relation using a random parameter $\alpha$ with $\text{Exp}[\alpha] = 0$.

$$
[D]_{ZrO_2} = [D]_{ZrO_2}^0 + [D]_{ZrO_2}^\alpha
$$

(1)

where $[D]_{ZrO_2}^0$ is the stress-strain matrix of ZrO$_2$ that is a function of $\alpha$, and $[D]_{\text{dot}}^0$ and $[D]_{\text{dot}}^\alpha$ are its zero-th and first order terms.

Since dot-like pore region is made of ZrO$_2$ whose properties are characterized by Eq. (1) with small $\alpha$, its homogenized stress-strain matrix can be expanded in the same way.

$$
[D]_{\text{dot}} = [D]_{\text{dot}}^0 + [D]_{\text{dot}}^\alpha
$$

(2)

In the right hand side model in Fig. 9, let the stress-strain matrix of each finite element be $[D]$, whose zero-th order term is $[D] = [D]_{\text{dot}}^0$ or $[D]_{\text{mix}}^0$ and first order term is $[D] = [D]_{\text{dot}}^\alpha$ or $[D]_{\text{mix}}^\alpha$. Then, the finally obtained homogenized stress-strain matrix can be expressed in the following equation.

$$
[D] = [D]^{0}([D]_{ZrO_2}^0(\alpha), [D]_{\text{dot}}^0(\alpha))] = [D]^{0} + [D]^{\alpha}\alpha
$$

(3)

It means that the same random parameter can characterize the homogenized property even when hierarchical model is used. This simple relation enables us to calculate the expected value and variance as follows.
The zero-th and first order terms of \([D^u]\) can be derived as follows. The microstructure model is expressed by 
\[ Y \] whose volume is \([\gamma]\). \([B]\) is the strain-displacement matrix of an element and \([\chi]\) is the collection of characteristic displacements \([\chi']\) where \(k'l'\) is the tensor form notation \((k'l' = 11, 22, 33, 23, 31, 12)\). The characteristic displacement is unique concept that is defined in the two-scale homogenization theory. It expresses the perturbed deformation model of the microstructure due to the heterogeneity. That is a periodic function at the microscale associated with the assumed periodicity of the microstructure model, and is used to split the perturbed displacement term in the asymptotic expansion into the microscopic characteristic displacement and the macroscopic strain. Please refer (Guedes and Kikuchi, 1990) for more detail. Now, zero-th order terms in Eq. (3) are calculated by the following equations.

\[
[D^u]^0 = \frac{1}{|\gamma|} \int \left( [D]^0 - [D]^0[B][\chi]^0 \right) dY
\]

\[
{\chi'}^0 = \left[ \int [B]^0[D]^0[B] dY \right]^{-1} \left[ \int [B][D]^0 dY \right]
\]

Here, \([D]_i^0\) is \(k'l'\) column vector of \([D]^0\). The first-order terms are calculated as follows.

\[
[D^u]^1 = \frac{1}{|\gamma|} \int \left( [D]^0 - [D]^0[B][\chi]^0 - [D]^0[B][\chi]^0 \right) dY
\]

\[
{\chi'}^1 = \left[ \int [B]^0[D]^0[B] dY \right]^{-1} \left[ \int [B][D]^0 dY - \int [B][B][D]^0 dY \right] \cdot {\chi'}^0
\]

The presented method was applied to the generated model in Fig. 9. Note that 75 \(\mu\)m cubic inner region was extracted for analysis considering the modeling error near the surface region and it was subdivided into 27 sub-models due to the computational capacity. From the calculated expected value and variance by Eqs. (4) and (5), probabilistic homogenized Young’s modulus in \(z\) direction (thickness direction) \(E_z^H\) was estimated. In detail, its expected value \(\text{Exp}[E_z^H]\) and standard deviation \(\text{SD}[E_z^H]\) were calculated. Considering the uncertainty, the distribution of \(\text{Exp}[E_z^H] - 1.96\text{SD}[E_z^H]\) was plotted in Fig. 10 to evaluate the 95% reliability level.

In the region with relatively higher Young’s modulus, no slit-like pores existed. It was found that large slit-like pores decreased significantly the apparent Young’s modulus in the thickness direction. That is reasonable because large slit-like pores are almost perpendicular to the thickness direction as in the literature review (Nakamura, et al., 2000).

![Probabilistic distribution of homogenized Young's modulus in z-direction. The higher valued region and lower valued region were highlighted, which showed the difference of the existence of slit-like pores perpendicular to z-direction. (95% lower reliability level; 1.96\sigma less than expected value)](attachment:fig10.png)
5. Discussion and concluding remark

This paper described two key technologies. The first one is the generation of large enough microstructure model suited for homogenization from FIB-SEM observation. The static data of categorized microstructure elements was used to generate the model. Here, hierarchical modeling technique was used for nanoscale dot-like pore region. The second point lies in the first-order perturbation based stochastic homogenization (FPSH) method for the hierarchical model. The limitation of the derived formulation is that it is applicable only to porous media. It means that new formulation is needed to solve multiphase composite materials, because multiple random parameters must be defined.

The main limitation of this study is that only one model in Fig. 9 was analyzed. Not only the scattering of constituent material but also the geometric morphology is another important random parameter. Based on the established quantitative statistic database, more number of microstructure models reflecting the morphology should be analyzed in the future. If more models were generated, the Gaussian mixture can provide more reliable scattering of apparent property (Basaruddin, et al., 2013).

The computational cost is another problem to solve the microstructure model including slit-like pores. The hierarchical model allowed us to discretize the model by 0.1 μm cubic voxel elements, but still only the distribution of apparent Young’s modulus was calculated in this paper. The use of a supercomputer is one of the possibilities, which will enable us to analyze much larger-sized model than 100 μm cubic region. To analyze the probabilistic behaviors for the hierarchical model, if the proposed FPSH method is compared with conventional Monte Carlo (MC) simulation, the present method has a remarkable merit in computational cost. Only one FPSH analysis by solving two equations (7) and (9) provides the probabilistic density function of the homogenized properties, while MC simulation requires huge number of random samples. In the near future, the comparison of the computational cost should be studied between FPSH and MC methods, where various sampling algorithms should be tested.

The validation of stochastic prediction is still an open problem, but experimental measurement of macroscopic property should follow the present research work.

Finally, using the stochastic prediction in Fig. 10, the bound analysis of deformation under uniform compression in \( z \) direction was carried out. The maximum displacement using the values in Fig. 10 was 6.25 % larger than that using expected value, which is equal to the deterministic prediction. Such bound analysis will be useful in the design of engineering applications. The probabilistic estimation of fracture is supposed to be the next challenging topic.

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References


