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In situ observation of grain evolution in ceramic sintering by SR-CT technique

XU Feng(许 峰)[1], HU Xiao-fang(胡小方)[1], NIU Yu(牛 玉)[1], ZHAO Jian-hua(赵建华)[1], YUAN Qing-xi(袁清习)[2]

1. Chinese Academy of Sciences Key Laboratory of Mechanical Behavior and Design of Materials, University of Science and Technology of China, Hefei 230026, China;
2. Institute of High Energy Physics, Chinese Academy of Sciences, Beijing 100049, China

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Abstract: Grain evolution of boron carbide ceramic powder during isothermal sintering process was in situ investigated by synchrotron radiation X-ray computed tomography (SR-CT) technique. The process of grain growth and material migration during three sintering stages was clearly distinguished from the 2-D and 3-D reconstructed images. The results show that from room temperature to 1 200 ℃ (0−270 min), grains gradually approach each other and form the sintering neck but grain growth does not start, which is indicated as the initial sintering stage. While the sintering time is between 270−390 min (temperature is 1 200 ℃), material migration between grains starts, while grains and sintering neck grow up, which is defined as the middle sintering stage. As the sintering time exceeds 390 min (temperature is 1 200 ℃), pores become isolated and spheroidized, which shows the final sintering stage. The double logarithm curve of mean grain radius and time logarithm during middle stage of isothermal sintering process is obtained from reconstructed images and the grain growth exponent is 0.364 03, falling in the predicted range of the traditional sintering theory. The experiment results are in accordance with those of the traditional sintering theory and provide effective experimental data for further analysis of the sintering process and the mechanical characteristics of ceramics.

Key words: sintering; grain evolution; non-destructive testing; synchrotron radiation X-ray computed tomography

1 Introduction

Ceramics has wide applications[1−4] for its outstanding physical and mechanical properties such as high strength and hardness, strong anti-corrosion and temperature resistance. Sintering is a widely applied method to prepare ceramics which has great impact on the properties of the final products. Sintering is a very complex thermal process which is affected by many sintering parameters such as grain-evolution. In situ observation of the grain evolution during sintering is therefore necessary for studying the thermally-activated mechanisms of sintering. However, it is difficult to realize the drawbacks of the traditional material testing method (such as SEM, TEM). Cutting and polishing of specimen before observation may destroy and change the internal microstructures.

Contrast to the traditional material testing methods, the new technique of the synchrotron radiation X-ray computed tomography (SR-CT)[5−6] makes it possible to in situ observe the grain-evolution during sintering. However, due to the technical limitations, there are few reports on this new method. Recently, LAME et al[7−8] introduced SR-CT technique to get in situ observation. LAME et al[7] observed the sintering process of Cu powder sintering at 1 050 ℃ and steel distaloy sintering at 1 130 ℃. BERNARD et al[8] constructed 3D visualization of microstructures evolutions of glass powder sintering at 700 ℃ and crystallized lithium borate powder sintering at 720 ℃. Recently, our research group[9] studied the porosity evolution of boron carbide bulk material during sintering process using SR-CT technique.

In order to further study the thermally-activated and material migration mechanism of sintering process, the work presented in this work focus on the grain growth kinetics during sintering. Grain evolution of loose boron carbide ceramic powder during isothermal sintering process was in situ investigated by SR-CT technique. The projection images of the sample were obtained during sintering process in real-time. Two-dimensional and three-dimensional reconstructed images were obtained by treating the projection images of different

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Corresponding author: XU Feng; Tel: +86-551-3600564; E-mail: xufeng3@ustc.edu.cn

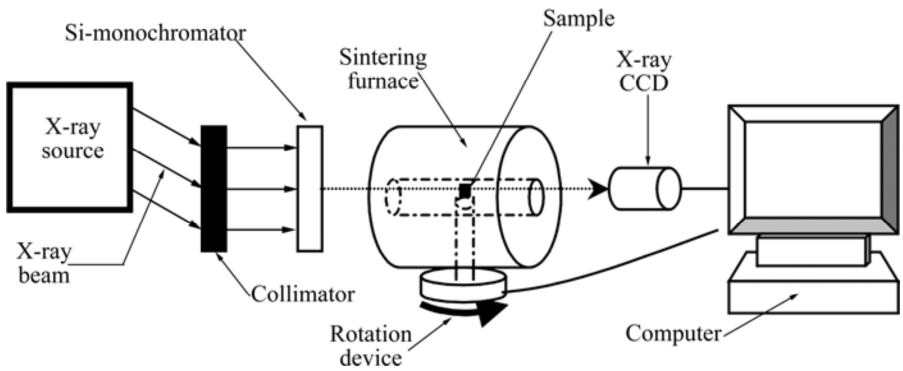

Fig.1 Schematic diagram of SR-CT projection imaging facility

sintering periods with filter back projection arithmetic and digital image processing method. From the reconstructed images, three sintering stages of the boron carbide ceramic sample were clearly distinguished and several sintering phenomenon during the sintering process such as grain contact, sintering neck growth and pore spheroidization were observed. The double logarithm curve of mean grain radius and time logarithm during middle stage of isothermal sintering process was obtained from reconstructed images and the grain growth exponent is 0.364 03, falling in the predicted value of the traditional sintering theory.

2 Experimental

The experiment was carried out on the 4W1A beam-line at BSRF, Beijing, China. A schematic diagram of this SR-CT projection imaging facility is given in Fig.1. A wide collimated synchrotron radiation X-ray (spot size up to 14 mm×10 mm) with energy range from 3 to 24 keV is available. The X-ray with 24 keV selected by silicon single-crystal monochromator was used in this test. The samples were heated in a sintering furnace specially designed for SR-CT. This furnace has a temperature range from room temperature to 1 600 ℃, even temperature region of 2 cm^3 and the highest heating rate of 250 ℃/h. There is a corundum cylinder connecting with a rotation device in the furnace. The MRS102 rotation device, which has angle resolution of 0.001 25° and repeatable positioning accuracy of 0.005°, was provided by Beijing Optical Instrument Factory. The samples were introduced on the top of the corundum cylinder. The synchrotron radiation X-ray passed through samples and reached an X-ray charge-coupled device (CCD) detector which recorded the intensity message of X-ray. The CCD including a 1 300×1 030 pixels chip with a unit pixel of 10.9×10.9 μm^2, offered an 8 bits dynamic range.

A packing of boron carbide ceramic powder with a diameter about 75 μm was investigated in this work. The loose powder was poured into a quartz capillary with 0.6 mm in diameter, 10 mm in height. The quartz capillary was heated in the sintering furnace at the heating rate of 240 ℃/h. The heating process and recorded points were shown in Fig.2.

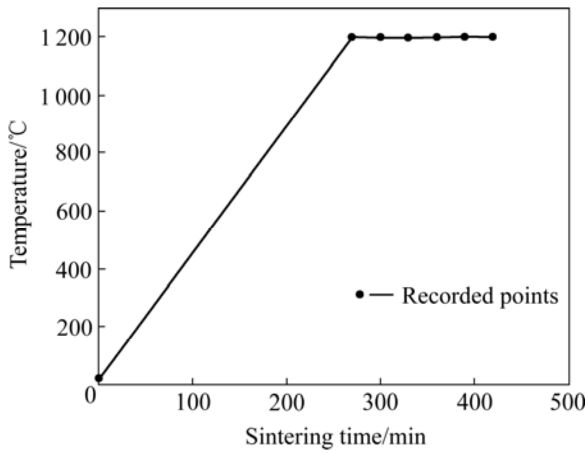

Fig.2 Heating process and recorded point

At each recorded point, the sample was imaged in different projection angles (in the range of 0°−180°). Typically, 180 shadow images of the specimen were acquired. The images were then processed by filtered back projection algorithm[10]. A number of two-dimensional (2D) slides (in serial order) representing the effective attenuation coefficients of the specimen in terms of gray levels were obtained. These two-dimensional specimen slices can be stacked to provide three-dimensional (3D) configuration scenery of the specimen.

A series of cross-section reconstructed images at different sintering time were successively obtained by SR-CT technique. Fig.3 shows seven reconstructed images representing nearly the same cross-section of the sample. The reconstructed images cover a gray value range of 0 (black) to 255 (white) where the high gray value means the high relative density. That is, the white area means grains and the black area means pores.

Three-dimensional reconstructed images can also be obtained by treating the cross-section images with digital image processing method, as shown in Fig.4.

3 Discussion

Sintering process has been studied a lot since 1960s and a series of sintering theories[11−15] were proposed. In these sintering theories, considering the differences in grain-evolution, sintering process contains three stages: initial, middle and final stage. During the initial stage, grains gradually contact with each other and form the sintering necks. Grain does not grow up in this stage. Grain growth starts at the middle stage. Grains grow more quickly together with pores and the pores remain connecting with each other as well as the sintering neck gradually grows up during this stage. During the final stage, the grain-evolution velocity of the material is gradually raised, the grain boundaries connect with each other, and the pores are isolated.

However, as stated in the introduction, these theories are hardly directly tested by experiments because of the drawbacks of the traditional material testing method: cutting and polishing of specimen before

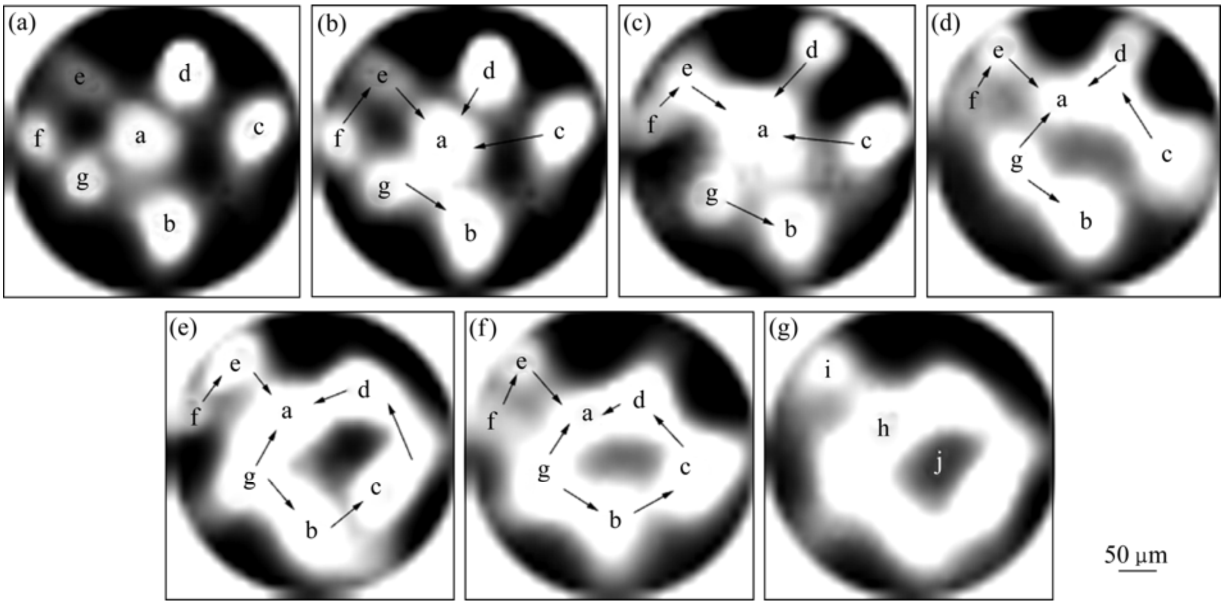

Fig.3 Reconstructed images of same cross-section of sample at different sintering periods (Letters a—i mark several different grains, arrow indicates material migration direction between grains, letter j represents final isolated pore): (a) 20 ℃, t=0 min; (b) 1 200 ℃, t=270 min; (c) 1 200 ℃, t=300 min; (d) 1 200 ℃, t=330 min; (e) 1 200 ℃, t=360 min; (f) 1 200 ℃, t=390 min; (g) 1 200 ℃, t=420 min

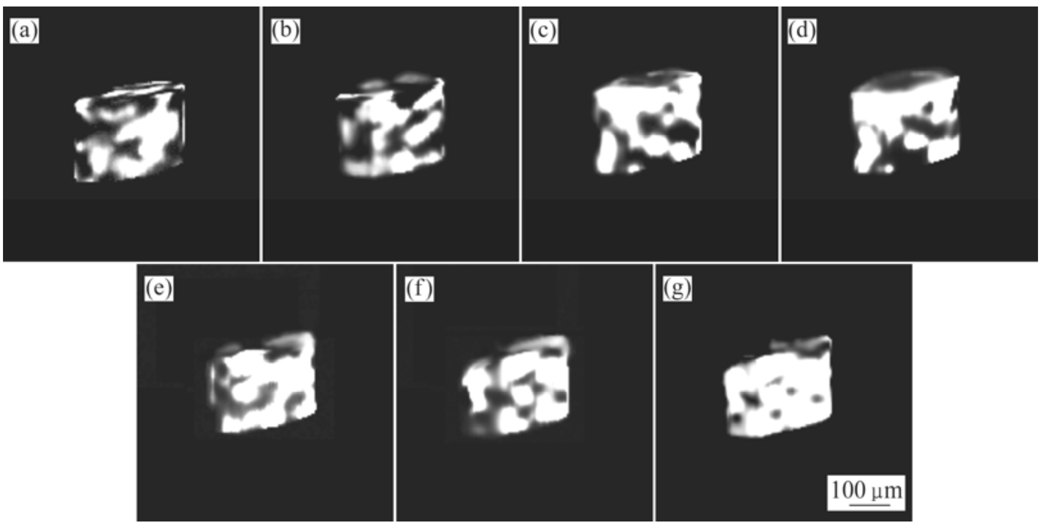

Fig.4 Three-dimensional reconstructed images of sample at different sintering periods: (a) 20 ℃, t=0 min; (b) 1 200 ℃, t=270 min; (c) 1 200 ℃, t=300 min; (d) 1 200 ℃, t=330 min; (e) 1 200 ℃, t=360 min; (f) 1 200 ℃, t=390 min; (g) 1 200 ℃, t=420 min

observation may destroy and change the internal microstructures.

Different from the traditional experimental methods, characteristics of grain-evolution during three sintering stages can be clearly distinguished and the grain growth exponent during middle sintering stage can also be directly obtained from the reconstructed images obtained by SR-CT experiment.

3.1 Grain evolution during sintering process

The microstructures evolution of the sintering process including grain contact, sintering neck growth and pore spheroidization can be observed from the SR-CT reconstructed images (as shown in Figs.3−4). Many sintering phenomena that are predicted in the traditional sintering theories are clearly observed.

1) From room temperature to 1 200 ℃ (0−270 min), grains gradually approach each other and form the sintering neck but grain-growth does not start (as shown in Fig.3(a) and (b)), which is indicated as the initial stage of sintering process.

2) While the sintering time is passing 270−390 min (temperature is 1 200 ℃), material migration between grains starts (material migration direction is indicated by the arrow), grains and sintering neck grow up and pores still connect each other (as shown in Fig.3(b)−(f)), which is defined as the middle stage of sintering process.

3) As the sintering time exceeds 390 min (the temperature is 1 200 ℃, as shown in Fig.3(f)−(g)), pores become isolated (shown as pore j) while the smaller grains coalesce to neighboring grains (shown as grain h and grain i), which shows the final stage of sintering process.

3.2 Grain growth exponent

In the traditional sintering theories, the characteristics of grain growth during the middle stage can be expressed as[14]:

$$R = K \cdot t^n \quad (1)$$

where R is the mean grain radius, K is a constant and n is the grain growth exponent in the range of 0.25 to 0.5 depended on the diffusion mechanism of sintering process. However, this expression was hardly directly tested by experiments for the drawbacks of the traditional material testing method.

The mean grain radius of the boron carbide ceramic powder during the middle sintering stage can be obtained from the SR-CT reconstructed images by digital image processing method (as shown in Table 1).

The relationship between the mean grain radius and sintering time in log-log coordinates is shown in Fig.5.

Table 1 Mean grain radius at different sintering time during middle stage of sintering

Sintering time/ min	Temperature/ ℃	Mean grain radius/μm
270	1 200	35.78
300	1 200	37.12
330	1 200	38.51
360	1 200	39.75
390	1 200	40.86

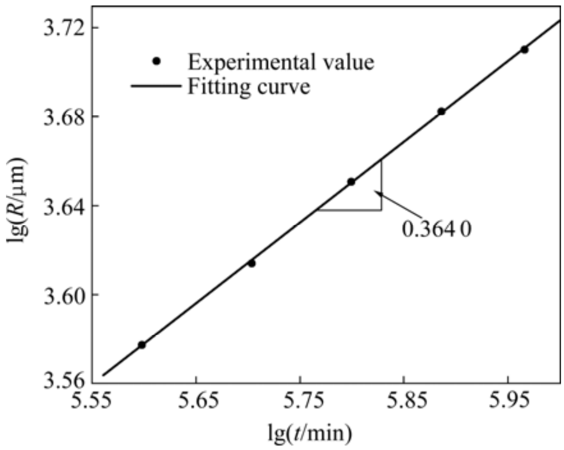

Fig.5 Double logarithm curve of mean grain radius and time during middle stage of sintering

A linear relationship is clearly shown and the grain growth exponent during the middle sintering stage is fitted as n=0.364 03, falling in the predicted range of the traditional sintering theory. Till then, characteristics of grain-growth during sintering were directly tested by experiments by the SR-CT technique. The experiment results are in accordance with the traditional sintering theory and provide effective experimental data for further analysis of the sintering process and the mechanical characteristics of ceramics.

4 Conclusions

1) A series of cross-section reconstruction images of boron carbide ceramic powder are obtained. Three dimensional reconstruction images of boron carbide ceramic powder are obtained by using digital image processing method. Grain evolution process of boron carbide ceramic powder during three sintering stages can be clearly observed from 2-D and 3-D reconstructed images.

2) Many sintering phenomena predicted in the traditional sintering theories such as grain growth, sintering neck formation, material migration and pores spheroidization, are clearly observed. The mean grain radius of the boron carbide ceramic powder during the

middle sintering stage is directly obtained from the SR-CT reconstructed images.

3) Analysis of grain growth characteristics during sintering process is done. A linear relationship is clearly shown and the grain growth exponent during the middle sintering stage is fitted as n=0.364 03, which is in the predicted range of the traditional sintering theory.

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