Evaluation of joined silicon nitride by X-ray computed tomography (X-ray CT)

Naoki KONDO,† Yoshihiro NISHIMURA, Takayuki SUZUKI and Hideki KITA

Advanced Manufacturing Research Institute, National Institute of Advanced Industrial Science and Technology (AIST), 2266–98 Shimo-shidami, Moriyama-ku, Nagoya 463–8560

1. Introduction

Silicon nitride ceramics are often used as industrial components, such as stalk tubes, ladles, and heater element protection tubes in the aluminum casting industry, and transfer rolls in the steel industry. These components are often large, sometimes exceeding 1 m in length. Various industries now require components with improved properties, such as larger size, lighter weight, and/or complex shapes.

One of techniques to make such components is joining. By making small ceramic parts and joining them, large, lightweight, and/or complex-shaped components may be actualized. However, ceramics including silicon nitride are difficult to be machined, welded, or shaped using plastic forming techniques, since they behave quite differently from metals and plastics. Moreover, ceramic components are brittle and sensitive to defects. Therefore, the existence of defects at joints reduces their reliability.

To ensure the reliability of the joined ceramic components, efficient detection and elimination of defects are required. Non-destructive evaluation (NDE) is useful to evaluate the internal defects, which exist inside of the joined components and cannot be found by observing their surfaces. One promising technique for NDE is X-ray computed tomography (X-ray CT), which has been widely applied for defect evaluation.† 1Received May 26, 2010; Accepted September 27, 2010

2. Experimental

A previously reported joined silicon nitride sample5) was examined by X-ray CT. The sample fabrication procedure is briefly summarized below.

Mixed powder of silicon (Si), zirconia (ZrO₂), and spinel (MgAl₂O₄) was compacted, and then reaction bonded (RB) in a nitrogen atmosphere at 1450°C. By this procedure, Si was nitrided, and porous silicon nitride body with the composition of Si₃N₄=5 mass % ZrO₂=5 mass % MgAl₂O₄ was formed. This porous body later comprised the bulk region of the joined sample.

Slurry was also prepared from the same mixed powder by adding water and dispersant. Joining was conducted by a slip-cast-like procedure. Two porous RB silicon nitride bodies were placed side-by-side, and the slurry was poured into the gap between them. The porous bodies soaked up water from the slurry, and a consolidated Si joint in the gap was formed. This Si joint later comprised the joint region. After drying, the joined body was subjected to RB at 1450°C for nitridation of the silicon, followed by post-sintering (PS) at 1800°C for densification. By this procedure, a dense, joined silicon nitride body was obtained.

A 3 × 5 × 7.5 mm sample for X-ray CT observation was cut from the joined silicon nitride body. The position of the sample in the joined body and X–Y–Z directions are defined in Fig. 1. As the sample was cut from the edge of the joined plate, defects such as segregation of additives, particles, cracks, and pores were more frequent than in the center part.

Microfocus X-ray CT equipment, Scan X mate-A100S40 by Comscantecno Co. Ltd., was used. Investigation was conducted under voltage of 100kV and current of 20μA. Resolution was about 10μm. Technique used for this investigation was a popular one, and detail was reported previously.5) The surface of the sample was also examined by scanning electron microscopy (SEM) and energy dispersive X-ray spectrometry (EDX).

3. Results and discussion

By X-ray CT, the existence of pores and cracks, the segregation of elements, impurities, and particles, and the density

©2010 The Ceramic Society of Japan
and crystal structure within the ceramic body are expected to be detected from the differences in the absorption of X-rays. Heavy elements, which have a higher absorption ratio, are shown as bright in the images, while pores and cracks, which have a lower absorption ratio, are shown as dark in the images. A three-dimensional image can be obtained by scanning the ceramic body.

**Figure 2** shows typical sectional images of $X$-$Y$, $X$-$Z$, and $Y$-$Z$ planes around a joint region by X-ray CT. $X$-$Y$ and $X$-$Z$ sections have a bulk-joint-bulk configuration, while $Y$-$Z$ sections generate images from the joint region only. A $Y$-$Z$ image from the bulk region is shown in **Fig. 3** for comparison. SEM-EDX observation was performed on the exterior surface of the $X$-$Z$ plane, and the result is shown in **Fig. 4**.

The upper, outer area of the joint region in **Fig. 2** is bright, indicating heavy element segregation in this region. Gradation in brightness corresponds to distribution of the heavy elements. No such bright area existed in the bulk region, as shown in **Fig. 3**. Note that the bright outline at the edge of the sample was not due to segregation of the heavy element, but due to a problem from the imaging technique. The bulk and joint regions were fabricated from a powder compaction and a slurry, respectively. Therefore, the heavy element segregation was caused by the slurry process. By SEM-EDX, the heavy element was identified as Zr, which was added to the mixed raw powder. The amount of Zr in the bright area was roughly 1.1 times higher than in the dark area, according to SEM-EDX. The joined sample contains 5 mass% ZrO$_2$ (i.e., ~3.7 mass% Zr) as additive. Therefore,
difference in amount of Zr between the bright and dark areas, which should be less than ~0.4 mass %, was clearly visualized by X-ray CT.

In the X–Z plane in Fig. 2, bright spots were observable at the interface between the bulk and joint regions. These spots were distributed like dotted lines, which were confirmed in 3-D visualization by X-ray CT. The spots exposed on the surface are shown in Fig. 4. The spots were Zr-rich and Si-poor, according to SEM-EDX. In the previous report, tetragonal zirconia (t-ZrO₂) was detected by X-ray diffraction analysis. Therefore, the bright spots are most likely t-ZrO₂ particles. The spots at the interface were formed by the separation of ZrO₂ in the slurry. Separated ZrO₂ concentrated at the interface, which resulted in their dotted-line-like distribution.

Bright spots were also found in the bulk and joint regions, as shown in Fig. 2, but these large spots were isolated. The spots, occasionally found on the surface, were also examined by SEM-EDX, which indicated that they were also Zr-rich and Si-poor. These isolated ZrO₂ particles were due to insufficient milling of the raw powder. These particles are usually located inside of the silicon nitride body. X-ray CT can reveal the existence of the particles, which do not expose on the surface.

Dark lines were observed in the X–Y and Y–Z planes, as shown in Fig. 2. These lines were determined to actually be a plane by 3-D visualization, and were therefore considered to be a crack. This crack did not expose on the surface. Therefore, it was hardly found by surface observation. X-ray CT can visualize the crack locating inside of the joint. This crack seemed to form during the drying of the consolidated joint. Cracks were also observed at the edge of the X–Z plane. These cracks were formed due to separation of the bulk and joint regions at the edge, and also seemed to form during the drying.

As mentioned above, X-ray CT successfully revealed internal defects, such as the segregation of Zr, the ZrO₂ particles and their distributions, and the internal crack. The information was obtained by observing from outside of the joined silicon nitride. The combinational use with SEM-EDX, which has higher resolution and ability of element analysis, but is a destructive method, gives detailed information such as particle shape, identification of element and its distribution, etc.

4. Conclusion

X-ray computed tomography (X-ray CT) was performed to evaluate defects in a joined silicon nitride sample. The resolution of the X-ray CT was about 10 μm. The bulk and joint regions of the joined silicon nitride body were fabricated by powder compaction and slip-casting, respectively, followed by sintering. Several internal defects were observed by X-ray CT.

1) Segregation of Zr, due to separation of ZrO₂ in the slurry, was observed at the upper, outer region of the joint.
2) ZrO₂ particles, due to separation of ZrO₂ in the slurry, were found at the interface between the bulk and joint regions.
3) Large isolated ZrO₂ particles, due to insufficient milling of ZrO₂, were found in both the bulk and joint regions.
4) Cracks, formed during the drying after slip-casting, were found in the joint region and at the edges.

These defects were mostly introduced during the fabrication procedure, i.e., insufficient milling of the ZrO₂, separation of ZrO₂ in the slurry, and cracking during drying after slip-casting. X-ray CT can obtain the internal information from outside of the specimen, and is a useful evaluation method to examine defects, which are introduced not only by joining but also by usual ceramics processing.

Acknowledgement This research was supported by METI and NEDO, Japan, as part of the project: “Development of innovative ceramics manufacturing technologies for energy saving.”

References