# Compaction of the SiC granule by cyclic CIP

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Green compacts by cyclic CIP procedure and their sintered bodies were made with SiC granules which exhibited typical elasto plasticity in the compression test on individual granules. For all the compacts of repeating times of 1, 10, 100 and 1000, central parts of the compact had lower Vickers hardness than the peripheral parts. This result implied that there was the presence of density gradients in all the compacts and central parts had lower density than the peripheral parts. It was considered that this was caused by the pitch in the carbon powder added as sintering aid. Bulk density of sintered bodies increased with CIP repeating times and the density of specimens of CIP repeating times of 100 and 1000 was 99% of theoretical. From measurement results of Vickers hardness in sintered bodies and SEM observation for microstructure, it was considered that the density gradient in cyclic-CIP compacts hardly leaves in the sintered body and the homogeneous sintered body with high density was obtained by the increase of CIP repeating times. 

## 1. Introduction

It seems likely that there are relation between the process of compaction or sintering and deformation or cracks at sintering ceramics products [1–3]. It is desirable to keep density homogeneity and, simultaneously, to make fully dense compacts in order to prevent from cracks and deformation.

Cyclic-CIP procedure is as follows; First of all, a certain degree of bias pressure $P_0$ is applied. Then, a cyclicly varying pressure is superimposed (maximum pressure: $P_{max}$). After repeating for a certain times, superimposed cyclic pressure is stopped and finally the bias pressure is released. Using this technique Nishimura *et al.* studied that in case of $Al_2O_3$ powder, the green density increased remarkably as the number of repeating times of pressure increased. In addition, the rate of increasing density against the repeating times became larger as the maximum pressure became higher [4, 5]. On the other hand, Kitamura *et al.* investigated that using cyclic-CIP procedure on $Al_2O_3$ powder, to increase repeating times helped the homogeneity of density distribution. That meant to increase the number of repeating time was more effective than to increase the uniformity of the compacts [6, 7].

However, in case of the cyclic-CIP procedure on carbon powder, the density of compacted body did not change very much in spite of the increasing number of repeating times. That was caused by the pitch consisted in the carbon powder [8]. Therefore, it is considered that in case of preparing compacts with cyclic-CIP procedure, powder characteristics has a significant influence on the CIP compaction behavior.

The granules are generally used for preparing ceramic products and it is expected to investigate mechanical property of individual granules in obtaining new knowledge about compaction behavior. The authors measured the load displacement curves in compression test of individual ceramic granules and reported the relation between compression behavior of granules and property of compacted bodies [9–11].

In the present report, the bodies were compacted through cyclic-CIP of SiC granules which exhibited typical elasto plasticity in the test on individual granules. And,

(1) the relation between repeating times of CIP and density homogeneity,
(2) the relation between homogeneity in compacted body and that in sintering body,

were discussed.

## 2. Experimental

### 2.1. Characterization of granules

The granule composed of $\alpha$-SiC primary particle was used as a raw material. In the granulation of raw powder, total amount of 6 wt % of PVA and fatty acid-base binders was added.

Histogram of granule size distribution measured is showed in Fig. 1. The average diameter is about 60 $\mu$m and the size extends over a region from 20 to 100 $\mu$m. Fig. 2 shows the scanning electron microscope (SEM) photographs of SiC granules. They have coarsely-packed structures and extremely rough surfaces.

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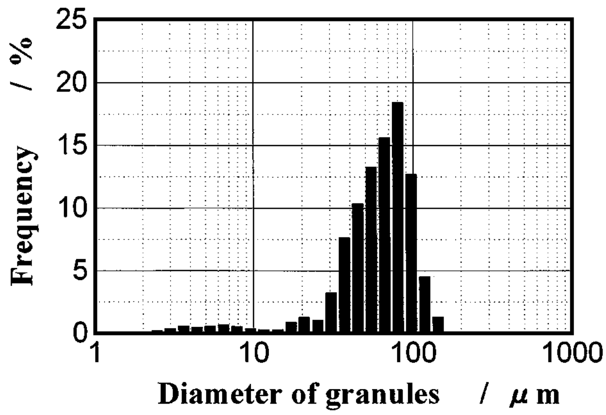


*Figure 1* Histogram of diameter of granules used in this experiment.

Fig. 3 illustrates the load-displacement curves observed after compressive tests for individual granules. It was recorded by micro compression testing machine (MCTM-500, Shimadzu Co. Ltd.) with a constant load of 0.05 mm/s. The load increases linearly with displacement up to approximately 3 $\mu$m and the yield starts at the maximum load point (1) at this region. After this, the load gradually increases due to the gradual fracture of SiC granule up to approximately 26 $\mu$m (2). Then the load increases again and this additional increase in load is thought to be attributed to the compression of primary particles decomposed the ceramic granule.

### 2.2. Preparation of cyclic-CIP compacted body and measurement of characterization

The granules were pre-formed as cylinders of 15 mm in diameter and 30 mm in height by uni-axial pressing under a pressure of 5 MPa for 60 seconds.

After that, cyclic-CIP forming was performed under the conditions summarized in Table I. Ordinary CIP was performed to compare with the date of cyclic-CIP under the condition in Table I. For the cyclic and ordinary CIP compacted bodies, the bulk density was calculated from the dimensions and weight.

TABLE I CIP conditions

| | |
|---|---|
| Cycle-CIP condition | |
| Maximum pressure | 100 MPa |
| Bias pressure | 0 MPa |
| Frequency | 0.5 Hz |
| Number of cycles | 10, 100, 1000 cycles |
| Static CIP condition | |
| Maximum pressure | 100 MPa |
| Holding time | 60 s |

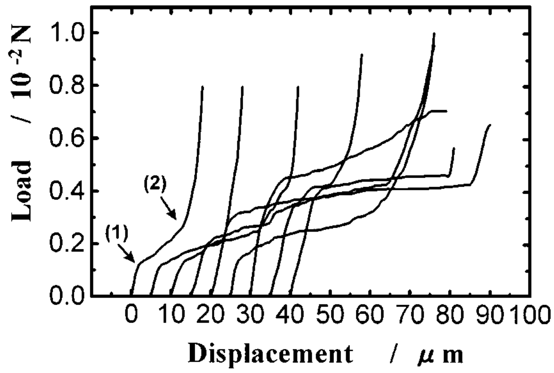


*Figure 3* Load-displacement curves of SiC granules.

The specimens were broken into two pieces at around the center part along longitudinal direction, polished the abrasive paper # 600, then the Vickers hardness of the compact was measured at intervals of 1 mm in the radial directions. Another broken surface were subjected to granule morphology observation by SEM.

### 2.3. Sintering and measurement of characterization of sintered bodies

Compacted bodies of both cyclic and ordinary CIP were sintered in a vacuum up to 1800 °C and then at 2075 °C for 3 hours in an argon atmosphere. The bulk density of the obtained samples after sintering was measured by the Archimedes method.

The specimens were cut into two pieces at around the center part along longitudinal direction, polished with

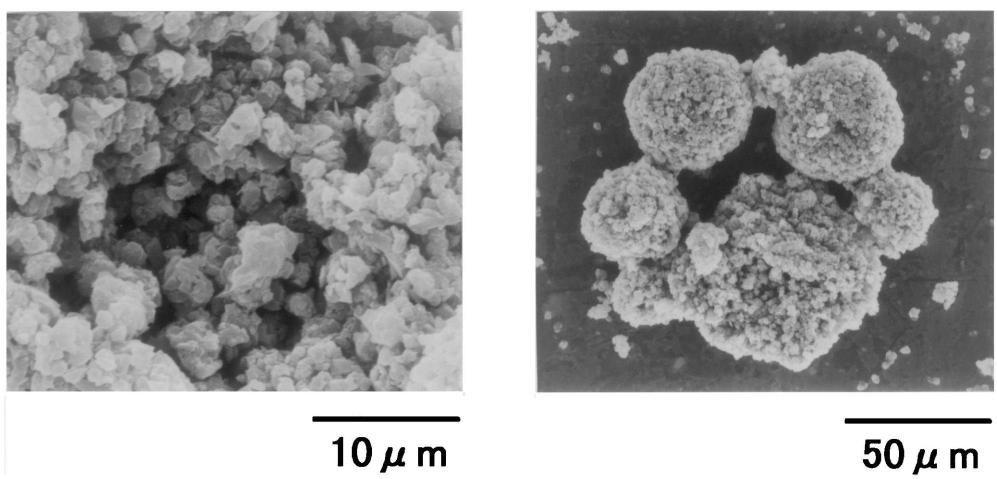


*Figure 2* SEM photographs of granules.

diamond pastes of 1.0 $\mu$m. For the polished surface, the pores distribution was observed by SEM and the Vickers hardness was measured at intervals of 1 mm in the radial directions.

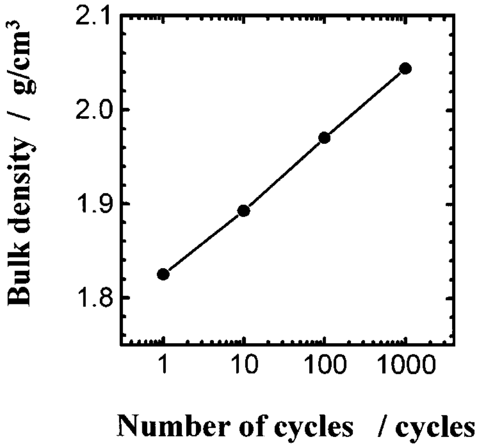


*Figure 4* Effect of number of cycles on the bulk density of SiC compacts.

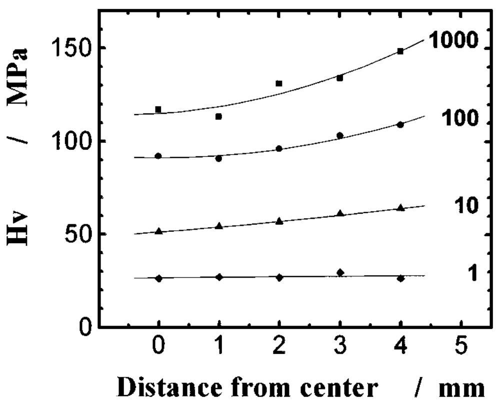


*Figure 5* Vickers hardness distribution in compacts.

## 3. Results and discussions

### 3.1. The density distribution in cyclic CIP compacted bodies

The relative density of the specimen after pre-forming was about 48%. Fig. 4 shows the effect of repeating times on the density of the green compacts formed by cyclic-CIP procedure. The value that the repeating time of CIP is one shows the relative density of compact obtained from the ordinary CIP procedure. Since the green density increases linearly against the log scale of repeating times, it is clear that for the granule used in this experiment the cyclic CIP procedure has also the effect of accelerating the densification of compacts. The green density has no tendency to saturate at the repeating times of 1000 yet.

Fig. 5 shows Vickers hardness distributions of the compacts in the radial directions. For all the compacts of repeating times of 1, 10, 100 and 1000, central parts in the compact have lower Vickers hardness than the peripheral parts. There is a good correlation between Vickers hardness on the surface of compact and the green density with those previously reported [9]. Therefore, it is obvious that there is the presence of density gradients in all the compacts of repeating times of 1, 10, 100 and 1000, and central parts in the compact have lower density than the peripheral parts.

The differences of Vickers hardness between the central parts and peripheral parts in the compact increase with repeating times and it means that the density gradient in the compact with higher repeating times of CIP is higher than that with lower repeating times. It was reported that the cyclic-CIP procedure would have the effect of accelerating the homogeneity of density distribution in compact, but the result of the density distribution in this research showed reversal tendency.

Nishimura *et al.* reported that, in case of the cyclic-CIP procedure on carbon powder which contained the pitch, the density of compacted body did not increase

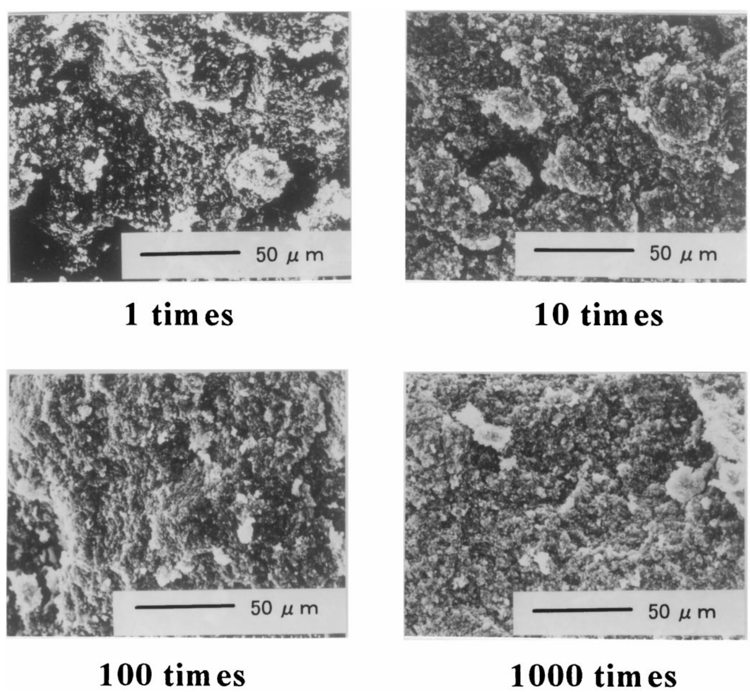


*Figure 6* Fracture surface of SiC compacts.

very much in spite of the increasing number of repeating times and this unchange of density was due to the viscoelasticity of pitch in carbon powder [8]. It is likely from previous work that the pitch prevent the compact from densifying. SiC granule used in this experiment contains total amount of about 10 wt % of pitch as a sintering aid. Therefore, the reason why peripheral parts in the compact showed higher density in comparison to the central parts is that the peripheral part which the pressure smoothly transferred during short period easily densified compared to the central part.

Fig. 6 shows the SEM photographs of granule morphology in the central part of compact. The green bodies of repeating times of 1 and 10 have traces of uncrushed granules remaining, but granule frameworks were not encountered in those of repeating times of 100 and 1000. On the other hand, in the case of peripheral part, the granules were completely crushed in the green bodies at all the green bodies. As for the large density distribution in the compacts of repeating times of 100 and 1000, however, no reason was considered from the SEM observation.

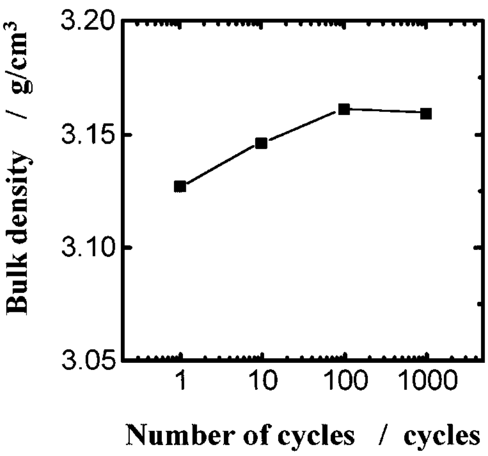


*Figure 7* Effect of number of cycles on the bulk density after sintering.

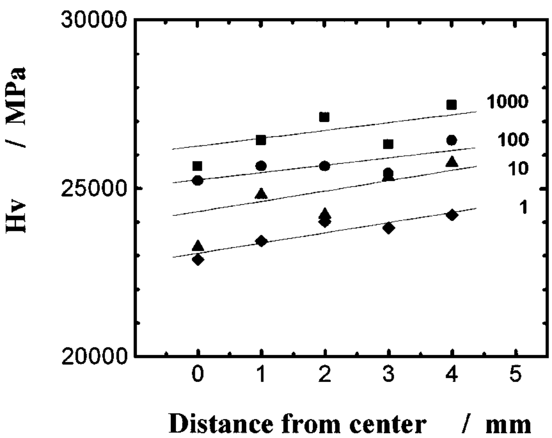


*Figure 8* Vickers hardness distribution in sintered bodies.

## 3.2. Effect of homogeneity in compacts on homogeneity in sintered bodies

Fig. 7 plots the results of density of sintered bodies. Bulk density increases with increasing the CIP repeating times. However, the density of sintered bodies obtained from compacts of repeating times of 1000 does not change in comparison to that of repeating times of 100. Those values are 3.16 g/cm$^3$ and both almost reach the density of 99% of theoretical. Therefore, as for SiC granule used in this experiment, sintered body with almost theoretical density can be obtained from the compact of CIP repeating times of 100.

Fig. 8 shows Vickers hardness distributions of the compacts in the radial directions. A load of 5 kg was used to carry out tests. For all the sintered bodies of CIP repeating times of 1, 10, 100 and 1000, Vickers hardness decreases slightly toward the center in the radial directions. The differences of hardness between the central

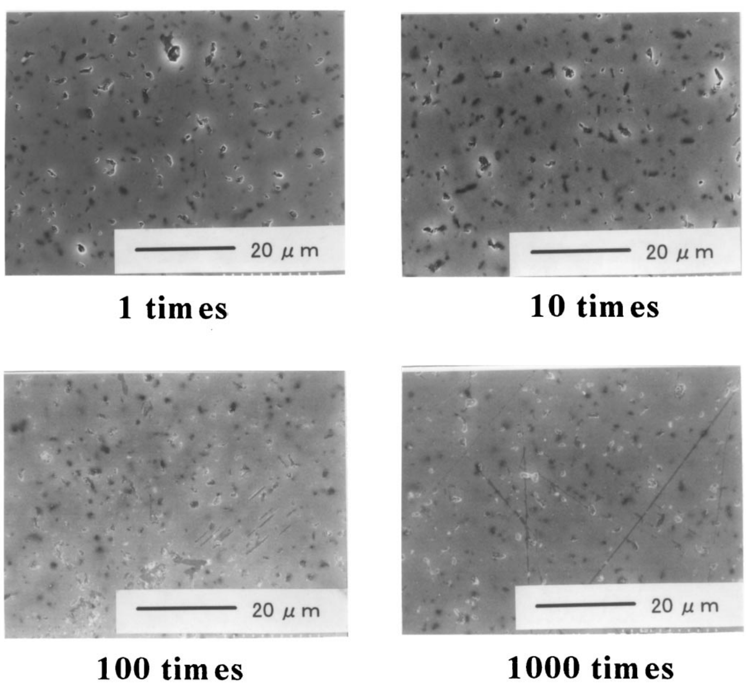


*Figure 9* Pores distribution in the sintered bodies.

parts and peripheral parts were almost constant on all the specimens. However, Vickers hardness had a tendency to increase with increasing CIP repeating times. The average value of each sintered body is 23.8 GPa for repeating time of 1, 24.8 GPa for 10, 25.8 GPa for 100 and 26.7 GPa for 1000, and it became clear that SiC sintered body with high hardness value was gained by being sintered the compact of CIP repeating times of 1000.

Fig. 9 shows SEM photographs of the pores distribution in the central parts of the sintered bodies. In the case of all the specimens of CIP repeating times of 1, 10, 100 and 1000, slight amount of pores and unreacted carbon were observed in the central parts. Moreover, the pore located in the central part had a tendency to change smaller with increasing the CIP repeating times. Therefore, it is concluded that the increase of CIP repeating times has the effect on formation of sintered body with a fine-pored structure.

The inhomogeneity observed in the compacts of CIP repeating times of 100 and 1000 becomes rather small in the sintered bodies from the results of Figs 8 and 9. Therefore, it became clearly from this experiment that the density gradient in cyclic-CIP compacts hardly leaves in the sintered body and the homogeneous sintered body with high density is obtained by the increase of CIP repeating times.

## 4. Summary

After the cyclic-CIP compaction using SiC granules which exhibited typical elasto-plastic in the compression test on individual granules, the green body was sintered. The relation between repeating times of CIP and density homogeneity in compacted body, in addition, the relation between the repeating times and the homogeneity in sintered body were discussed. The following results were obtained.

(1) The numbers of repeating times made the density in compacted body higher but did not affect density homogeneity in it. The differences between density of surface and that of center part tended to increase.

(2) Sintering made the density inhomogeneity in green body decreased. It was considered that to increase repeating times of CIP made it possible to make highly homogenized sintering body.

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