1. Introduction

The fabrication of highly densified polycrystalline diamond (PCD) compact requires high pressure and temperature conditions, because diamond is a high-pressure phase material. Two fabrication methods of PCD have been developed, namely, the static high pressure sintering technique and the shock compaction technique. One of the static sintering techniques using Co metal or Fe-Ni-Co alloy as a sintering catalyst is a commercial process developed by Wendorf and Rocco\(^1\) more than 25 years ago. Although the PCD synthesized by this process has superior physical properties,\(^2\) the presence of a large amount of metallic catalyst in it causes deterioration such as graphitization or cracking.\(^3\) On the other hand, by the shock compaction method it is not difficult to generate exceedingly high temperature and pressure conditions, so that diamond powder can be densified without metal catalysts that promote graphitization of diamond.\(^4\) Thus, the shock compaction technique seems to have a greater potential for the fabrication of PCD. However, few studies have been performed to determine the heat-resistance property of shock-compacted PCDs. The heat resistance of PCD is one of the most important properties under the conditions of the expected applications. Regarding the shock compaction of diamond powder, it was reported that the addition of silicon resulted in a reduction of the number of cracks and an improvement of the mechanical properties of the recovered compacts.\(^1\) In the present study, we investigated the heat resistance of shock-compacted diamond powders with and without silicon additive, and compared it with that of commercial PCD obtained by static sintering with metal catalysts. From the results, we investigated the thermal stability of PCDs and the influence of the silicon additive on these properties of shock-compacted PCDs.

2. Experimental procedure

Synthetic diamond powder provided by General Electric Co., Ltd. was used as the starting material powder for shock compaction. The grain size was 2 to 4 μm grade of GE-300 series, as indicated in our previous work.\(^3\) Fine silicon powder as an additive was mixed with starting diamond powder in ethyl alcohol using an agate mortar. The added silicon powder of grain size under 0.1 μm grade was provided by Vacuum Metallurgical Co., Ltd. The content of silicon in the composite powder and the pure diamond powder were pressed into SUS-304 capsules to form a disk 5 mm thick and 12 mm in diameter.\(^5\) The densities of the pressed powders were 60% of the theoretical density. Shock treatment was carried out with a mouse-trap-type plane wave generator and a momentum trap recovery system.\(^5\) An iron flyer plate of 3.2 mm thickness was impacted at a velocity of 2.3 km/s against the capsules containing powder.

The shock-treated PCDs in the form of disks were carefully removed from the recovered capsules using a lathe. Both the upper and bottom faces of each disk were ground and polished with a diamond wheel for characterization, and then subjected to X-ray diffraction (XRD) with nickel-filtered Cu Kα radiation, optical microscopy, and Vickers hardness test (normal load, 9.8 N, 15 s). To evaluate the heat resistance of PCDs, the shock-compactcd PCDs and a commercially available PCD (SYNDITE 002 provided by DeBeers Co., Ltd.) were treated at high temperatures of 1000 and 1200°C for 30 min in a vacuum of (1–1.5) × 10⁻³ Pa. After the heat treatments, XRD patterns were investigated to analyze the transformation of PCD, optical microscopic images to check microcracking and transformation of the sample surface, and the results of Vickers hardness test were investigated. Only the shock-compactcd PCDs treated at 1000°C were treated again at an elevated temperature of...
1200°C for 30 min in vacuum. After the heat treatment, the same characterization was performed.

3. Results and discussion

Shock-compacted PCDs were recovered as a whole disk from capsules, as shown in Fig. 1. Figure 1 shows the bottom view of a shock-compacted PCD with silicon. No clear visible cracks are observed, while the shock-compacted pure PCD has some cracks. Figure 2 shows the XRD patterns obtained before and after heat treatment of commercial PCD, pure PCD, and silicon-added PCD. It is found that commercial PCD is considerably graphitized by the sintering metal catalyst when heat-treated at 1000°C. Figure 3 is an enlargement of the XRD patterns in Fig. 2 (20–80° 2θ region). As shown in this figure, pure PCD graphitizes gradually with increasing heat-treatment temperature. On the other hand, shock-compacted PCD with silicon has no graphite after heat treatment at 1000°C, but slightly graphitizes at 1200°C. It is confirmed that shock-compacted PCD has better resistance against graphitization than commercial PCD having metal catalysts, and that the addition of silicon contributes to the increase in resistance against graphitization of the shock-compacted PCD.

Figures 4, 5 and 6, respectively, show the polished surfaces of the commercial PCD, pure PCD, and silicon-added PCD before (A) and after (B) heat treatment. As shown in Fig. 4 (B), many cracks were observed on the polished surface of commercial PCD after heat treatment. In the shock-
compacted PCDs, in contrast, there was little difference in the polished surfaces before and after heat treatment, as shown in Figs. 5(B) and 6(B). As mentioned above, the shock-compacted PCD hardly transformed to graphite by this heat treatment, so that it showed no significant changes of its polished surface.

Table 1 compares the hardness values of commercial and shock-compacted PCDs before and after heat treatment; it is evident that hardness deterioration due to heat treatment is marked in the commercial PCD and less distinct in the shock-compacted PCDs. Especially, for the shock-compacted PCD with silicon, the deterioration of hardness is hardly seen after the heat treatment at 1000°C and only slightly seen by increasing temperature to 1200°C. Thus, the addition of silicon contributes to decrease the deterioration of hardness by heat treatment. The deterioration of hardness is related to the change in the XRD patterns: i.e., graphitization.

Therefore, it is concluded that the thermal stability of shock-compacted PCD is superior to that of commercial PCD with metal catalysts. In particular, neither graphitization, cracking nor change of the hardness was observed after the heat treatment at 1000°C in the shock-compacted PCD with silicon. The shock-compacted PCDs heat-treated at 1000°C were partly graphitized by a second heat treatment at 1200°C, but the amount of graphitization as observed from the XRD pattern and the deterioration of hardness were suppressed by the addition of silicon, suggesting that the addition of silicon contributes to improve the thermal stability of shock-compacted PCD.

4. Conclusion

Pure and fine-silicon-added composite diamond powders, ranging from 2 to 4 μm, were shock-compacted at a velocity of 2.3 km/s by an impacting iron flyer plate of 3.2 mm thickness, and the heat resistance of the shock-compacted pure and silicon composite diamond powder was studied. No graphitization or cracking was observed in the shock-compacted PCD after heat treatment at 1000°C for 30 min in a vacuum of (1-1.5) × 10⁻³ Pa. Although a temperature increase of up to 1200°C caused a slight increase in the amount of graphite as shown in the XRD pattern and a decrease in the hardness value, no significant difference was found in the microstructure. Thus, the addition of silicon resulted in a high thermal stability against graphitization. These results suggest that silicon is a desirable additive for improving the thermal stability of shock-compacted diamond powder.

References