Silicon Carbide Synthesis by Sublimation Method

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SiC properties change with grain size, orientation, and surface morphology. We prepared polycrystalline 3C–SiC on graphite by sublimation method using 6H–SiC powder compacts as source materials. The present study investigated the distance between the substrate and the SiC source, temperature gradients between the substrate and the SiC source = 20–40°C/mm, under the constant experimental conditions (argon pressure = 133 kPa, substrate temperature = 1580°C, deposition time = 30 minutes) Deposited crystals showed clear hexagonal habit and yellowish color. The maximum growth rate was obtained with preferred {111} orientation, and at 13 mm distance between the substrate and the SiC source. [Received August 6, 2003; Accepted November 27, 2003]

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1. Introduction

Silicon carbide (SiC) has been extensively used as high temperature structural material because of its excellent high temperature strength, good thermal conductivity and thermal shock resistance and excellent oxidation resistance. SiC is also being used as semiconductor material for high power and high frequency devices and other high temperature electronic applications.

At present, the biggest problem in the wide-scale use of these materials is difficulty in preparation of high quality single crystals. The difficulty comes from sublimation behavior without melting at atmospheric pressure. Since SiC sublimes above 2800°C at atmospheric pressure, it is difficult to have single crystals of SiC from melt. Another difficulty is controlling polytype and defects such as dislocation stacking faults, micro pipe, etc during the growth.

SiC was synthesized from silica and coke by the Acheson method for a wide variety of application. High purity and high quality SiC has been prepared by the Lely method, which is one of the sublimation methods. Modified Lely method has been developed by Tailor’s group using seed crystal and big SiC ingots were obtained by controlling polytype. Bigger ingots could be obtained using bigger crucibles.

Another modification of Lely method is Closed Space Technique (CST). The present letter reports the surface microstructure (such as crystal size and orientation) of SiC prepared on graphite substrate by sublimation CST method.

2. Experimental

Figure 1 shows a schematic drawing of a set-up arrangement for sublimation SiC growth. A graphite crucible was inductively heated up to 2200°C using a high frequency generator with 400 kHz frequency and 20 kW maximum out-put power. The sublimation experiment was carried out using graphite crucible. The distance between substrate and SiC source could be easily controlled in these experiments. Figure 2 shows the detailed from size and assembly of graphite crucible used in the present study. Substrates were cut a graphite bar to form pellets and surface was polished using #4000 abrasive. SiC source was α-SiC powder (Showa Denkou, DC grade) and formed to 6 mm pellets by uniaxial pressing at 60 kg/cm² for 1 min. Following experimental conditions were kept constant; Source temperature = 2200°C, substrate temperature = 1580°C, atmosphere = Ar, pressure = 133 kPa, and deposition duration = 30 min. The distance between substrate and source (ΔX) was changed from 10.14 mm to 17.14 mm using a crucible as shown in Fig. 2.

The deposited crystals were characterized by powder X-ray diffraction (XRD), and scanning electron microscopy (SEM). XRD pattern were obtained using CuKα (λ = 1.5406Å), with 30 kV of accelerating voltage, 40 mA of current, 0.01° of step scan, 0.5 deg of diversion slit width, 0.5
degree of scattering slit, 0.15 mm of receiving slit width, and filtering with monochromater and Ni filter.

3. Results and discussion

3.1 Identification and polytype

Deposited crystals on graphite substrates were examined by XRD in order to identify SiC polytype. Figure 3 and Fig. 4 show XRD patterns for the deposited crystals for ΔX < 12.5 mm and ΔX > 12.5 mm, respectively. All the peaks detected by XRD were identified to SiC and graphite substrate. No peak could be observed at 2θ = 34.2° and 38.2°, which means that 6H-SiC (2θ = 34.2°), 4H-SiC (2θ = 38.1°) and 2H-SiC (2θ = 38.1°) could not be identified. All the peaks in Fig. 3 and Fig. 4 identified with 3C-SiC and lattice constant was calculated to be 4.357 Å. The visual appearance also supported the presence of 3C-SiC since the deposited crystals were yellowish color. This result is reasonable, since the substrate temperature was kept constant at 1580°C. Thermal stability of SiC is as following, 1200–1400°C for 2H-SiC, 1400–1600°C for 3C-SiC, 1600–2100°C for 4H-SiC, and above 2100°C for 6H-SiC.[10]

3.2 Surface microstructure

Figure 5(a)–(h) show SEM micrographs of deposited SiC crystals on graphite substrate. According to these pictures in Fig. 5, ΔX was effective in controlling the surface morphology or surface microstructure. Mean crystal sizes were counted from SEM pictures using Cera Part[11] and resultant relationship between ΔX and crystal sizes is shown in Fig. 6. Crystal sizes increased as ΔX increased up to 12.97 mm and then decreased as ΔX increased according to Fig. 5 and Fig. 6.

This phenomenon can be explained by considering degree of supersaturation (Ss) from equilibrium vapor pressure. Since substrate temperature (Td) kept constant in the present experiments, increasing ΔX means increasing temperature of SiC source (Ts), which means increasing vapor pressure. Therefore at ΔX = 10.14 mm Ss is too low for enough nucleation and growth as shown in Fig. 5(a). As increasing ΔX, Ts increases, and at ΔX = 12.97 mm Ss is not enough to nucleate
but enough to growth as shown in Fig. 5(e). As $\Delta X$ increases further, $S_s$ becomes enough to nucleate a large number of nuclei and a large amount of small crystals grow as shown in Fig. 5(h).

4. Summary

SiC crystals were deposited on graphite substrate by the modified Lely method. Effect of distance between substrate and SiC source was investigated under constant conditions of substrate temperature, and deposition time in Ar atmosphere,

1) Deposited crystals were 3C-SiC in present experimental conditions.

2) Maximum crystal size could be obtained at $\Delta X=12.97$ mm.

References


11) Software for measurement grain size by Science Solutions International Laboratory.