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# Thermal expansion of the cubic (3C) polytype of SiC

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Thermal expansion of the cubic beta or (3C) polytype of SiC was measured from 20 to 1000° C by the X-ray diffraction technique. Over that temperature range, the coefficient of thermal expansion can be expressed as the second order polynominal: $\alpha_{11} = 3.19 \times 10^{-6} + 3.60 \times 10^{-9}T - 1.68 \times 10^{-12}T^2$ (1/° C). It increases continuously from about $3.2 \times 10^{-6}$/° C at room temperature to $5.1 \times 10^{-6}$/° C at 1000° C, with an average value of $4.45 \times 10^{-6}$/° C between room temperature and 1000° C. This trend is compared with other published results and is discussed in terms of structural contributions to the thermal expansion.

## 1. Introduction

Several papers have reported the thermal expansion of the cubic (3C) or beta polytype of SiC for the approximate temperature range from room temperature to 1400° C [1–7]. The results of these studies have been summarized by Touloukian [8]. Although Taylor and Jones [2] do consider the temperature dependence of the coefficient of thermal expansion, most studies only report mean or average values. These are summarized in Table I as values which were either taken directly from the publications or calculated from enlarged graphical presentations in the publications. The value of the mean coefficient of thermal expansion for $\beta$-SiC over those temperatures has been reported to range from the low value of $4.3 \times 10^{-6}$/° C after Suzuki *et al.* [7] to the high value of $6.2 \times 10^{-6}$/° C after Becker [1]. It is evident that considerable uncertainty exists regarding the coefficient of thermal expansion of $\beta$-SiC. In an attempt to clarify this unsatisfactory situation, this paper reports the high temperature X-ray diffraction measurements of the thermal expansion of $\beta$-SiC and expresses the coefficient of thermal expansion in polynomial form as a function of temperature. It also compares the results with values which have been previously published and discusses the structural effects on the thermal expansion of $\beta$-SiC.

## 2. Experimental details

Two $\beta$-SiC materials were obtained from PPG Industries (Barberton, Ohio) and General Electric Co. (Schenectady, New York) as submicron size powders which were a dark grey in colour. Each material yielded well defined X-ray diffraction peaks of relative intensities which were in good agreement with the current JCPDS standard [9]. The lattice parameters of the two silicon carbides were identical and equal to 0.43581 ± 0.000 05 nm, which compares satisfactorily with the reported JCPDS value. None of the other common polytypes of SiC, such as the 4H, 6H, or 15R were detected to be present in these pure beta polytype materials. The absence of other polytypes is probably related to the powder synthesis conditions.

Thermal expansion measurements were completed using nickel filtered CuK$\alpha$ radiation and a commercial high temperature diffractometer attachment with a platinum heating stage/sample holder which has been described in detail by Intrater and Hurwitt [10]. A helium atmosphere was used to prevent oxidation or nitridation of the sample SiC materials during the high temperature measurements. After alignment of the elevated temperature stage, the diffractometer was calibrated using a high purity $\alpha$-$Al_2O_3$ powder and then was checked by comparison with silicon and platinum standards. The *d*-spacing error was less than 0.000 008 nm over the $2\theta$ range between 100° and 150°. The precise temperature during the measurements was determined from the thermal expansion of the $\alpha$-$Al_2O_3$ which was used as an internal standard [11], although during the actual measurements the temperature was controlled using a Pt/Pt–10% Rh thermocouple. The (3 3 3) diffraction peak of the $\beta$-SiC which occurs at a $2\theta$ angle of about 133° at room temperature was monitored along with the (4 1 6) diffraction peak of the $\alpha$-$Al_2O_3$ internal standard which occurs at a $2\theta$ angle of about 136°. Measurements were made at temperature intervals of approximately 100° C for both heating and cooling between 20 and 1000° C. Further details of the experimental procedure are reported elsewhere [12].

## 3. Results

The lattice parameter *a* as it varies with temperature for the two $\beta$-SiC materials is illustrated in Fig. 1. Not only are the room temperature lattice parameters identical for these two materials, but their thermal expansions are also equivalent as is evident from Fig. 1. For this reason the data from the two materials were combined to determine a polynomial for the lattice parameter as a function of temperature using the least squares method. The lattice parameter *a* may

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TABLE I Summary of published thermal expansion coefficients of $\beta$-SiC

| Authors | Method | $\Delta T$(°C) | $\alpha$(1/°C) |
|---|---|---|---|
| Becker [1] | X-ray | RT–1200 | $6.2 \times 10^{-6}$ |
| Taylor & Jones [2] | X-ray | RT–1200 | $4.4 \times 10^{-6}$ |
| Popper & Mohyuddin [3] | Dilatometer | RT–1400 | $4.4 \times 10^{-6}$ |
| Clark & Knight [4] | X-ray | RT–1200 | $4.5 \times 10^{-6}$ |
| Kern *et al.* [5] | Dilatometer | RT–1000 | $4.8 \times 10^{-6}$ |
| Price [6] | Dilatometer | RT–1000 | $4.9 \times 10^{-6}$ |
| Suzuki *et al.* [7] | X-ray | RT–900 | $4.3 \times 10^{-6}$ |

be expressed by the third order polynomial:

$$a = 0.43577 + 1.3887 \times 10^{-6}T + 7.8494 \times 10^{-10}T^2 - 2.4434 \times 10^{-13}T^3 \text{ (nm)}, \quad (1)$$

where the temperature, $T$, is in °C. The standard deviation for the above equation is 0.000 026 nm [13].

The result in Fig. 1 illustrates that the lattice parameter $a$ of $\beta$-SiC is very nearly linear in temperature. On that basis, Equation 1 can be simplified for some engineering applications and the lattice parameter can be described by an equation which is linear in temperature:

$$a = 0.43570 + 1.9374 \times 10^{-6}T \text{ (nm)}. \quad (2)$$

However, for this linear approximation the standard deviation for the lattice parameter is about 0.000 043 nm, nearly twice that for the third order polynomial expression.

Thermal expansion coefficients are readily presented as second order polynomials in temperature [14, 15]. The coefficient of thermal expansion of $\beta$-SiC, $\alpha_{11} = (1/a)\,(\mathrm{d}a/\mathrm{d}T)$, can be obtained by differentiating Equation 1 and then dividing by the lattice parameter. That process yields the second order polynomial:

$$\alpha_{11} = 3.19 \times 10^{-6} + 3.60 \times 10^{-9}T - 1.68 \times 10^{-12}T^2 \text{ (1/°C)}. \quad (3)$$

The average coefficient of thermal expansion over the entire temperature range from 20 to 1000° C, $\bar{\alpha}$, calculated from $\bar{\alpha} = \int \alpha_{11}\mathrm{d}T/\int \mathrm{d}T$ is $4.45 \times 10^{-6}$/°C.

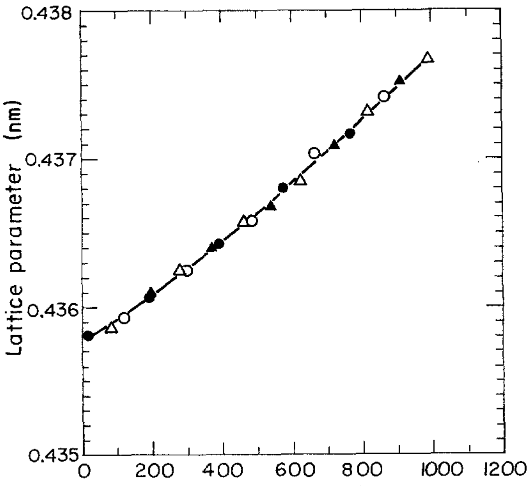

*Figure 1* Lattice parameter of the cubic $\beta$-SiC (3C) polytype as a function of temperature. PPG Industries: ○, heating; ●, cooling. General Electric: △, heating; ▲, cooling.

This value is similar to the reported values in the previous papers of Taylor and Jones [2], Popper and Mohyuddin [3], Clark and Knight [4] and Suzuki *et al.* [7].

## 4. Discussion

Fig. 2 illustrates the temperature variation of the coefficient of thermal expansion of $\beta$-SiC as determined in the present study. It also compares the result of this study with the results of Taylor and Jones [2], Popper and Mohyuddin [3] and Suzuki *et al.* [7]. The thermal expansion coefficients after Popper and Mohyuddin [3] and Suzuki *et al.* [7] which are plotted in Fig. 2 were graphically interpolated from enlargements of their original plots of $(\Delta L/L)$ against $T$ and $a$ against $T$, respectively. The room temperature coefficient of thermal expansion of $3.26 \times 10^{-6}$/° C that was determined in this study is only slightly larger than the value of $2.90 \times 10^{-6}$/° C reported by Taylor and Jones [2] and is nearly identical to the room temperature results of both Popper and Mohyuddin [3], and Suzuki *et al.* [7]. The coefficient of thermal expansion increases continuously to about $5.1 \times 10^{-6}$/° C at 1000° C which is also consistent with the results of both Popper and Mohyuddin [3], and Suzuki *et al.* [7], although only through 700° C for the latter. At 1000° C, it is again slightly larger than the value reported by Taylor and Jones [2] for that temperature, but slightly smaller than the other two reported values.

Taylor and Jones [2] have reported a distinctive temperature dependence for the coefficient of thermal expansion of $\beta$-SiC. Their results suggest that the thermal expansion coefficient of $\beta$-SiC increases rapidly below 200° C, but then remains constant at about $4.5 \times 10^{-6}$/° C over a wide temperature range from about 300 to 700° C, before increasing rapidly again above 700° C. A constant value for the coefficient of thermal expansion over the 300 to 700° C temperature

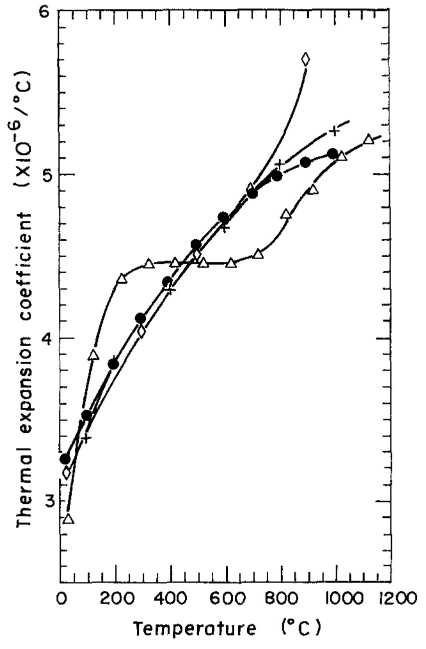

*Figure 2* Reported coefficients of thermal expansion for the cubic $\beta$-SiC (3C) polytype and their variations with temperature. ◇, Suzuki *et al.* [7]; +, Popper and Mohyuddin [3]; △, Taylor and Jones [2]; ●, this study.

range was not confirmed by the measurements of this study, rather the coefficients of thermal expansion gradually increased from about 4 × $10^{-6}/°$C to 5 × $10^{-6}/°$C over that particular temperature range. The thermal expansion results of Popper and Mohyuddin [3] and also those of Suzuki *et al.* [7] similarly agree with the continuously increasing coefficient of thermal expansion observed in this study in contrast to the constant value of the coefficient of thermal expansion between 300 and 700° C.

Hazen and Finger [16] present an equation for the mean thermal expansion coefficient which was derived on a basis related to the strengths of the interatomic bonds and which expresses the mean linear thermal expansion coefficients of crystal polyhedra between room temperature and 1000° C. That equation for the polyhedron thermal expansion is:

$$\alpha_{1000^\circ\mathrm{C}} = 4.0(4)\frac{N}{S^2 Z_a Z_c} \times 10^{-6}/^\circ\mathrm{C}, \qquad (4)$$

where $\alpha_{1000^\circ\mathrm{C}}$ is the mean thermal expansion coefficient between room temperature and 1000° C, $N$ is the particular structure's coordination number, $Z_a$ and $Z_c$ are the cation and anion valencies, respectively, and $S^2$ is a factor that is 0.20 for carbides and nitrides. Equation 4 has been demonstrated to quite adequately represent the coefficients of thermal expansion of many different structural polyhedra. For SiC the $N$, $Z_a$ and $Z_c$ are 4, 4 and 4, respectively. Substituting these values into Equation 4, yields an $\alpha_{1000^\circ\mathrm{C}}$ for the SiC polyhedron coefficient of thermal expansion coefficient of about 5 × $10^{-6}/°$C. That polyhedron coefficient of thermal expansion is in reasonable agreement with the average experimental value of 4.45 × $10^{-6}/°$C which was determined in this study. However, the theoretically calculated polyhedron value is slightly larger than the experimentally measured one and suggests that a bond angle tilting contribution to the thermal expansion exists [17] and that it reduces the overall coefficient of thermal expansion of the cubic (3C) $\beta$-SiC structure.

It is of interest to consider the elevated temperature trend of the thermal expansion coefficient as expressed by Equation 3 and illustrated in Fig. 2 as it relates to the Hazen and Finger polyhedron coefficient of thermal expansion calculated from Equation 4. Whereas the room temperature to 1000° C average coefficient of thermal expansion, $\bar{\alpha}$, is only slightly less than the theoretical value of the Hazen and Finger derivation, in fact, the value of $\alpha_{11}$ is much lower at room temperature and gradually increases with increasing temperature to the magnitude of the polyhedron value at about 1000° C. One interpretation which may be applied to explain this behaviour is that the bond angle tilting contribution which reduces the coefficient of thermal expansion continually decreases and becomes less important as the temperature increases. Whereas at lower temperatures the bond angle tilting contribution significantly reduces the overall thermal expansion of the cubic (3C) $\beta$-SiC structure, at elevated temperatures it is the polyhedron thermal expansion which dominates the overall thermal expansion of the structure. While these contributions are a plausible explanation, a detailed single crystal structure analysis at elevated temperatures is required to verify this interpretation.

## 5. Summary

The thermal expansion of the cubic beta polytype of SiC, the (3C) form, was determined by the high temperature X-ray diffraction technique from 20 to 1000° C. The coefficient of thermal expansion gradually increases from 3.2 × $10^{-6}/°$C at 20° C to 5.1 × $10^{-6}/°$C at 1000° C. The average coefficient of thermal expansion over that temperature range is 4.45 × $10^{-6}/°$C. This suggests that with increasing temperature the thermal expansion of $\beta$-SiC may become increasingly more dependent on the polyhedron thermal expansion, as the bond angle tilting contribution which reduces the overall expansion is diminishing.

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