EFFECT OF PROCESSING ON FRACTURE TOUGHNESS OF SILICON CARBIDE AS DETERMINED BY VICKERS INDENTATIONS

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ABSTRACT

Several α-SiC materials were processed by hot isostatic pressing (HIPing) and by sintering an α-SiC powder containing boron and carbon. Several β-SiC materials were processed by HIPing a β-SiC powder with boron and carbon additions. The fracture toughnesses K_{Ic} of these β- and α-SiC materials were estimated from measurements of Vickers indentations. The three formulas used to estimate K_{Ic} from the indentation fracture patterns resulted in three ranges of K_{Ic} estimates. Furthermore, each formula measured the effects of processing differently. All three estimates indicated that fine-grained HIPed α-SiC has a higher K_{Ic} than coarsed-grained sintered α-SiC. Hot isostatically pressed β-SiC, which had an ultrafine grain structure, exhibited a K_{Ic} comparable to that of HIPed α-SiC.

INTRODUCTION

Extensive investigations have been made to improve the strength and high-temperature properties of polycrystalline silicon carbide for heat engine applications. One factor greatly limiting the structural application of SiC is its poor fracture toughness. Fracture toughness K_{Ic} is the resistance to crack initiation and propagation. Therefore, improving silicon carbide's K_{Ic} is critical to application of this material.

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A variety of methods have been employed to evaluate the fracture toughness of SiC materials, including the chevron-notched beam[1,2], the double cantilevered beam[3], and controlled surface flaw tests.[1,4] The indentation method was chosen for this assessment of SiC because it can be performed quickly on a minimal amount of material. Unlike other test methods it does not require machining of a test bar. The test involves loading the material's surface with a Vickers diamond indenter. The sharp indenter causes cracks to emanate radially from the corners of the resultant impression.

Various approaches have been taken[5-9] to relate the indentation fracture pattern to the fracture toughness K_{1c}. However, each formula that has been derived as a solution to the indentation stress field produces different results. Breval et al.[10] noted the disparity between values obtained by various methods in an evaluation of several materials derived from sol-gels. Furthermore, for a given formula K_{1c} values may be load dependent.[4] For sintered α-SiC specifically, previously reported indentation K_{1c} values range from 2.3 to 3.8 MPa $m^{1/2}$.[1-4,11,12] Each experimenter has often used different relations for his K_{1c} determination as well as a different applied load. These factors make it difficult to make comparisons across the literature.

In this work three formulas chosen from the literature[1-3] have been used to make comparative fracture toughness determinations. The indentation test was employed to investigate the degree of toughness obtained in SiC materials produced by powder consolidation techniques such as sintering and hot isostatic pressing. The results are discussed in terms of density, grain size, and grain morphology, as influenced by processing methods.

EXPERIMENTAL PROCEDURE

Green Forming

The α-SiC* powder contained premixed boron and carbon additions. The β-SiC** powder was mixed with 0.6 wt % boron and 2 wt % carbon and with 0.6 wt % boron and 3 wt % carbon. A detailed characterization of each powder has been given in earlier studies.[13,14] Each powder was sieved through a 100-mesh screen and then milled in a solution of water and ammonium hydroxide (pH 11) by using SiC grinding media and a polyethylene bottle. The slurry was milled for 48 hr, and the pH was readjusted to 11. The slurry then was mechanically pressed under 14 MPa for 1 min in a procedure developed by Freedman and Millard.[15] Resulting disks were about 4.7 cm in diameter with a thickness of about 0.6 cm. Each disk was slowly dried in a desiccator and then cold isostatically pressed in vacuum-sealed bags under a pressure of 413 MPa. Samples were thoroughly dried in an oven at 200 °C.

Sintering

The α-SiC disks were sintered at temperatures of 2100, 2150, or 2200 °C for 4 hr. Specimens were also sintered at 2150 °C for 1/2, 1, or 2 hr. The density of the final samples was greater than 96 percent of theoretical.

Hot Isostatic Pressing

Green disks of α- and β-SiC were encapsulated in tantalum and hot isostatically pressed (HIPed). The disks were wrapped in boron-nitride-coated Grafoil*** and then placed in tantalum cans. The cans were outgassed for 6 to 8 hr at 1100 °C and vacuum sealed. The cans were then placed in a HIP unit, where an initial pressure of 14 MPa was applied. The temperature and pressure

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*Type II α-Sic - H.C. Starck, West Berlin, West Germany.

**HSC-100GL β-SiC - Superior Graphite Co., Chicago, IL.

***Union Carbide Corp., Cleveland, OH.

were increased simultaneously to the desired values. The α-SiC specimens were HIPed at 137 MPa and 1900 or 2000 °C for hold times of 1/2, 1, or 2 hr. The α-SiC samples with and without sintering aids were HIPed at 137 MPa and 1900 °C for 1 hr. After the pressure was released and the furnace cooled, the disks were removed from the tantalum cans. A more detailed description of the process is given in an earlier report.

Some α-SiC disks were encapsulated in glass and HIPed by an outside source.* The details of this process are undisclosed.

Fracture Toughness Determination

Test specimens were machined to 8-rms surface finish and then polished with 1-μm diamond paste. An indenter load of 24.5 N (2.5 kg) was applied at a rate of 1 mm/min and held for about 15 sec. Indentation radii and crack lengths, labeled a and C in Fig. 1, were measured with an optical microscope at a magnification of 400. Indents were not used if more than one crack extended from a corner or if the cracks did not extend parallel to the indent's diagonals. A total of 10 acceptable indents on each sample were used to obtain average K_{1c} values. The Young's modulus was approximated as 400 GPa and used in the following formulas. Evans and Charles[6] used a least-squares fit to relate $(K_{1c}\Phi/Ha^{1/2})$ to C/a, where Φ is a constraint factor (~3) and H is the hardness. This resulted in

$$\frac{K_{1c}\Phi}{Ha^{1/2}} = 0.15k\left(\frac{C}{a}\right)^{-3/2} \qquad (1)$$

where $k = 3.2$ for large C/a values. When $H = 0.47P/a^2$ for a load of P, the equation is simply $K_{1c} = 0.0753(P/C)^{3/2}$.

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*ASEA, Robertsford, Sweden.

Further modifications were made after the importance of residual stress effects had been recognized, for much of crack extension occurs during the load removal. Evans[7] introduced a factor of $(E/H)^{2/5}$, where E is the Young's modulus, and fit existing date with a polynomial curve to give

$$\log\left(\frac{K_{1c}}{Ha^{1/2}}\right)\frac{H^{2/5}}{E} = 16.32x^5 - 24.97x^4 + 11.23x^3 - 2.02x^2 - 0.34x - 1.59 \qquad (2)$$

where $x = \log(C/a)$

Anstis et al.[8] more recently developed a relation based on the work of Lawn et al.[9] to obtain the elastic/plastic stress field. They superimposed the residual stress field of the unloaded material onto the stress field due to an ideally elastic contact, yielding

$$K_{1c} = 0.014\left(\frac{E}{H}\right)^{1/2}\left(\frac{P}{C}\right)^{3/2} \qquad (3)$$

As in Eq. (1) the relation is proportional to $(P/C)^{3/2}$, but the factor E/H is also included.

RESULTS AND DISCUSSION

Microstructure

Nearly full-density (~96 percent of theoretical*) sintered α-SiC materials were produced at temperatures ranging from 2100 to 2200 °C. Higher density generally imparts higher strength in ceramic materials. However, under the high sintering temperatures and times required to achieve full density, grain growth occurs. For example, Fig. 2 shows an increase in average grain size as the sintering time at 2150 °C was extended from 1/2 to 4 hr. Similarly Fig. 3 shows the grain growth in α-SiC sintered for 4 hr at 2100, 2150 and 2200 °C. After a 4-hr hold at 2200 °C the grains were greatly elongated, with some longer than 80 μm (Fig. 3).

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*Theoretical density taken as 3.201 g/cm^3.

Hot isostatic pressing provided near full-density (~98 percent of theoretical) materials at lower temperatures. The applied pressure, in addition to temperature, acted as a driving force for densification. Small grains (0.2 to 3 μm in diameter) resulted from lower processing temperatures (Fig. 4). No significant difference was evident with regard to temperature or time. The grain morphology of HIPed α-SiC consisted primarily of equiaxed grains with carbon at the grain boundaries.[16] The density of glass-encapsulated HIPed α-SiC was ~98 percent of theoretical. The grains in the glass-encapsulated HIPed material were slightly larger than the tantalum-encapsulated α-SiC grains.

The slurry-pressed β-SiC material with 0.6 wt % boron and 2 or 3 wt % carbon was HIPed to 96 percent of theoretical density. The HIPed β-SiC materials had submicrometer-sized grains, which could not be resolved with the optical microscope (Fig. 4).

Fracture Toughness

The three K_{1c} values calculated for each sample indicated significant differences in the three relations. Using Eq. (1) to determine K_{1c} produced values typically 12 percent higher (3.4 to 5.0 MPa $m^{1/2}$) than the values produced by Eq. (2) (3.0 to 4.4 MPa $m^{1/2}$) Equation (3) produced values as much as 36 percent lower (1.9 to 3.0 MPa $m^{1/2}$) than those produced by means of Eq. (2).

As shown in Figs. 5 and 6 the indentation results indicated decreasing K_{1c} values with gradual increase in grain size. This is shown in Fig. 8. The effect of grain size on fracture toughness K_{1c} was reported earlier by Kruse and Hausner,[17] who observed a decrease in K_{1c} with grain coarsening due to excessive sintering temperature or increasing boron to carbon ratio.

The data for sintered α-SiC determined by Eq. (2) developed by Evans[7] (3.0 to 3.6 MPa $m^{1/2}$) compare well with data for sintered α-SiC in the existing literature. A comparison of K_{Ic} test methods by Orange et al.[1] reported indentation results for sintered α-SiC ranging between 2.6 and 3.5 MPa $m^{1/2}$ (Table I). These values agreed with chevron-notched beam test results for the same material (2.8 to 3.6 MPa $m^{1/2}$, Eq. (3)). They also reported results from controlled surface flaw (CSF) tests for sintered α-SiC within this range of data (2.8 MPa $m^{1/2}$) and noted that the straight-edge-notched beam (SENB) test method gave much higher results (4.1 to 4.5 MPa $m^{1/2}$). Srinivasan and Seshadri[4] reported K_{Ic} of sintered α-SiC by indentations (3.6 MPa $m^{1/2}$, Eq. (2)) and by CSF (3.3 MPa $m^{1/2}$). Here, reported values for the SENB test were again significantly higher. Lower values (2.2 and 2.3 MPa $m^{1/2}$) have also been reported, however, for sintered α-SiC when the indentation method was used.[2,3] These lower estimates are closer to the data obtained in this report by using Eq. (3).

HIPing of the α-SiC material produced higher indentation K_{Ic} values (3.6 to 4.0 MPa $m^{1/2}$, by Eq. (2), Fig. 7) than did the sintering process (3.0 to 3.6 MPa $m^{1/2}$). The high fracture toughness of the HIPed α-SiC material can be attributed to its smaller grain size in comparison with pressureless sintered material (Fig. 8). No K_{Ic} values for HIPed SiC were found in the literature, but values for hot-pressed α-SiC are generally higher than sintered α-SiC values. Seshadri et al.[11], using the indentation method, reported a K_{Ic} of 4.6 MPa $m^{1/2}$ for hot-pressed α-SiC, compared with 3.8 for sintered α-SiC. Zdaniewski and Kirchner[12], again using the indentation method, reported values between 3.8 and 4.7 MPa $m^{1/2}$ for hot-pressed α-SiC. The glass-encapsulated HIPed samples had a slightly lower fracture toughness (3.5 MPa $m^{1/2}$, by Eq. (2)) than the tantalum-encapsulated samples. This

lower K_{Ic} for higher grain size is the same trend that was displayed in the sintered materials.

The K_{Ic} values for HIPed β-Sic (3.7 to 4.1 MPa $m^{1/2}$) (Fig. 7) were comparable to those for HIPed α-SiC (3.6 to 4.0 MPa $m^{1/2}$). Statistical analyses such as the F-test indicated significant difference in variance, and Student's t-test indicated no significant difference in K_{Ic} between HIPed α- and β-SiC specimens. However, K_{Ic} values for HIPed α- and β-SiC were higher than those observed for pressureless sintered α-SiC (3 to 3.6 MPa $m^{1/2}$). For HIPed α- and β-Sic and pressureless sintered α-SiC, the F-test indicated no significant differences among the variances, but Student's t-test indicated statistically significant differences in K_{Ic} values.

CONCLUSIONS

The indentation test method allowed a rapid investigation of the fracture toughness K_{Ic} of sintered and hot isostatically pressed silicon carbide. Hot isostatic pressing of α- and β-SiC yielded materials with higher fracture toughness than did the sintering process applied to α-SiC. High K_{Ic} corresponded with small grain size.

Aspects of the indentation test method are not fully understood. Of the three formulas used, the relation derived by Evans gave results closest to existing data obtained in chevron and controlled surface flaw tests. This may not always be the case if there are variations in the test procedure. For example, further investigations should be performed to examine load effects on these materials.

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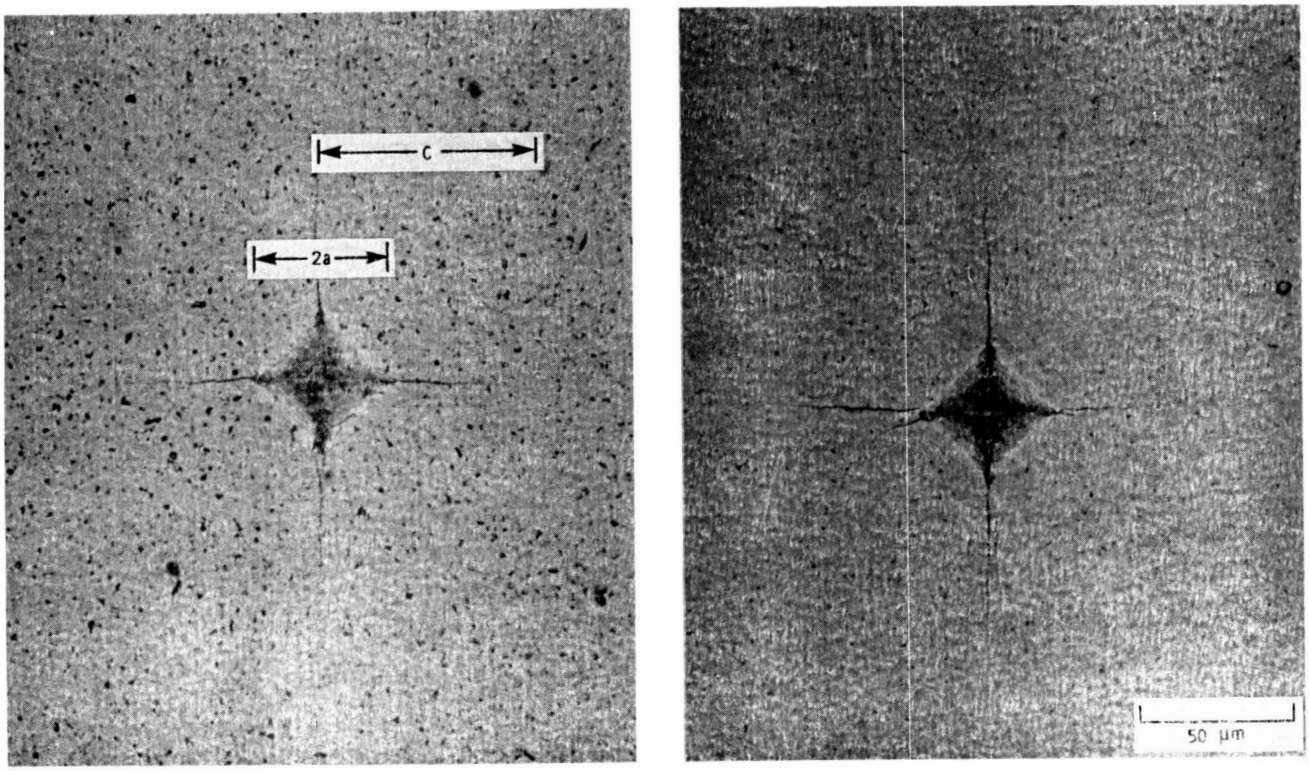
TABLE I. - INDENTATION TEST RESULTS FOR α-SiC COMPARED WITH OTHER TEST METHOD RESULTS

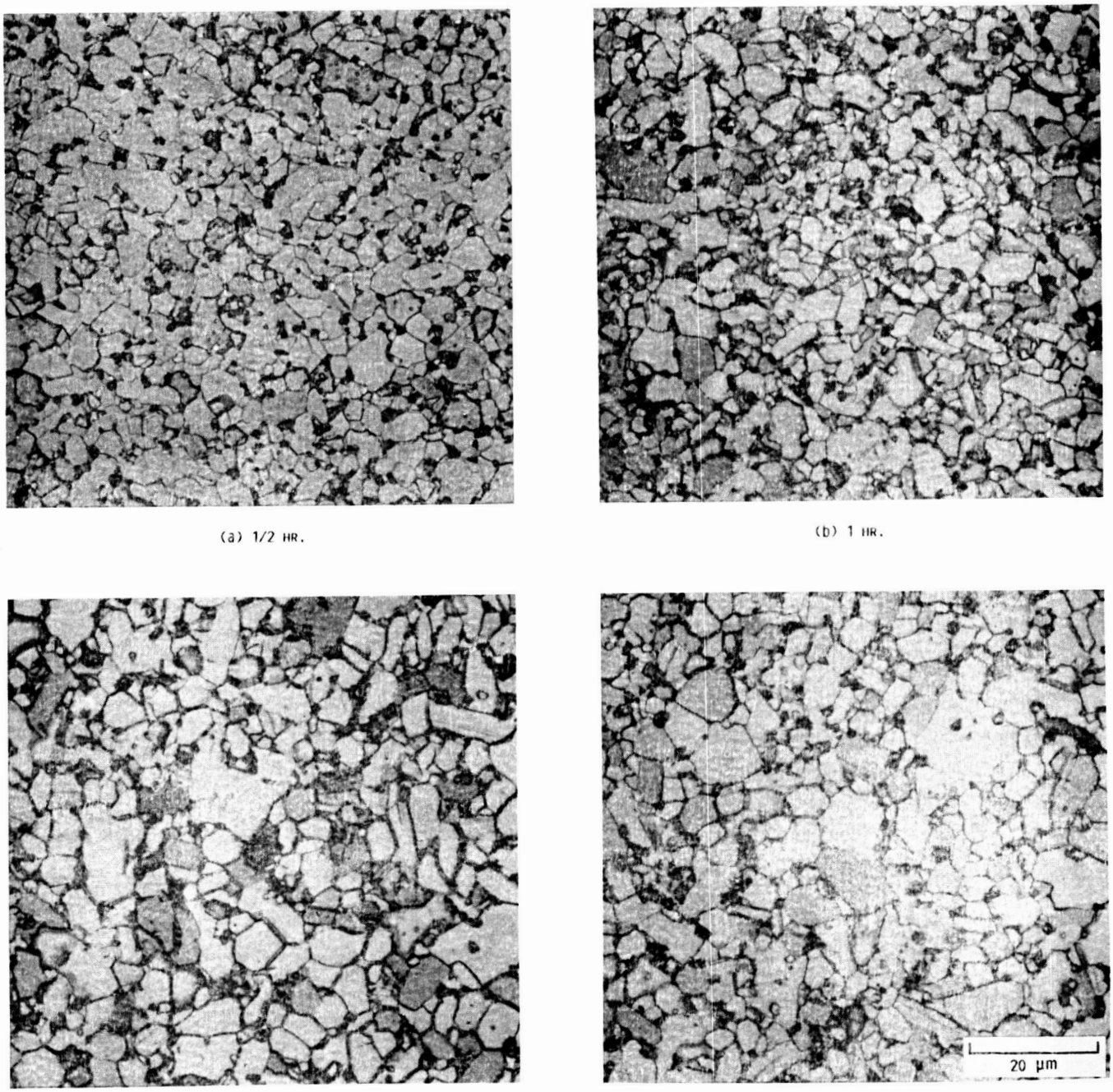
Reference	Material	Test method				
		Indentation	Chevron-notched beam	Straight-edge-notched beam	Controlled surface flaw	Double cantilevered beam
		Fracture toughness, K_{Ic}, MPA $m^{1/2}$				
1	Sintered	2.6-3.5	2.8-3.6	4.1-4.5	2.8	---
2	Sintered[a]	2.2	3.1	--------	---	---
3	Sintered[a]	2.3	--------	--------	---	2.5
4	Sintered[a]	3.6	--------	4.8	3.3	---
11	Hot pressed[b]	4.6	--------	--------	---	---
	Sintered[a]	3.8	--------	--------	---	---
12	Hot pressed[b]	3.8-4.7	--------	--------	---	

[a]Carborundum Co.
[b]NC203, Norton Co.



(a) 1900 °C. (b) 2000 °C.

FIGURE 1. - TYPICAL MICROINDENTATIONS IN α-SiC HIPed FOR 1 HOUR AT DIFFERENT TEMPERATURES.



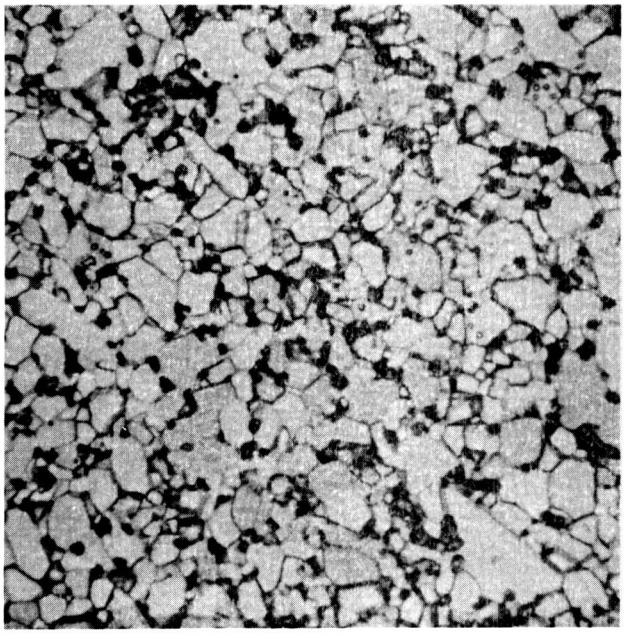
(a) 1/2 HR.

(b) 1 HR.

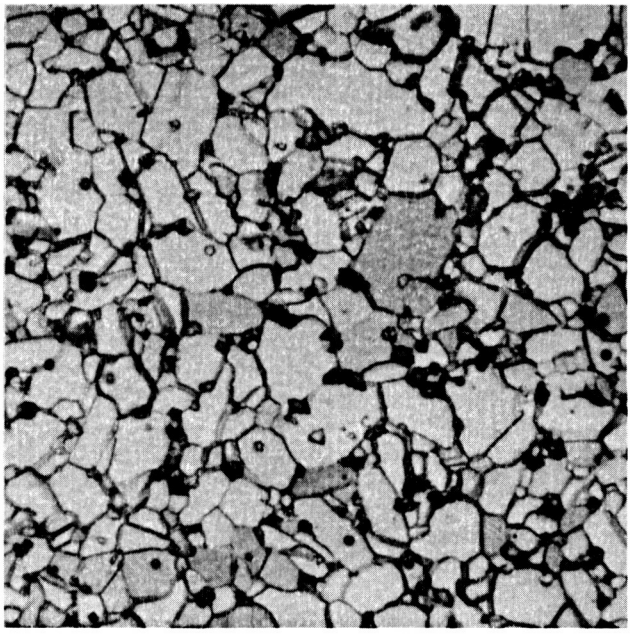
(c) 2 HR.

(d) 4 HR.

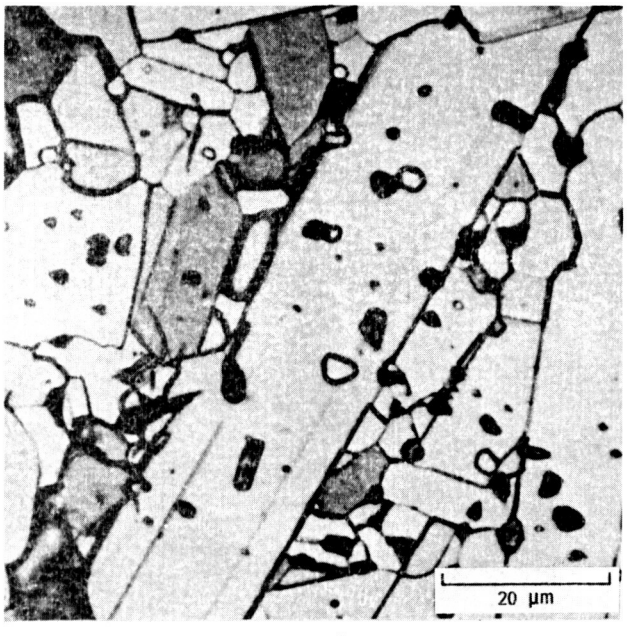
FIGURE 2. - MICROSTRUCTURE DEVELOPMENT IN SLURRY-PRESSED α-SiC SINTERED AT 2150 ^{o}C FOR DIFFERENT TIMES.



(a) 2100 °C.

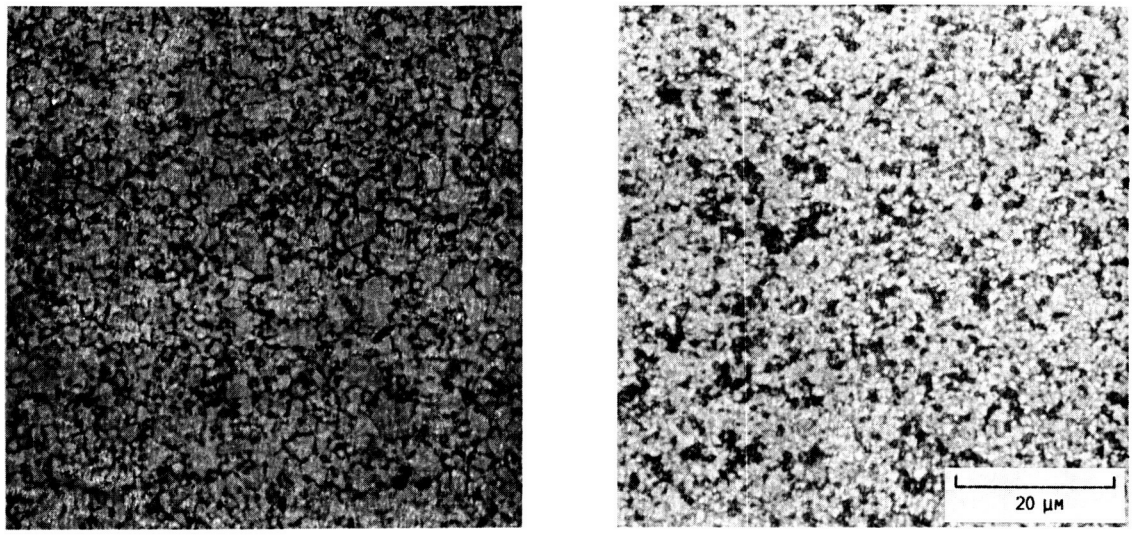


(b) 2150 °C.



(c) 2200 °C.

FIGURE 3. - MICROSTRUCTURE DEVELOPMENT IN α-SiC SINTERED FOR 4 HOURS AT DIFFERENT TEMPERATURES.



(a) α-SiC. (b) β-SiC.

FIGURE 4. - MICROSTRUCTURES OF α- AND β-SiC HIPed AT 1900 °C FOR 1 HOUR.

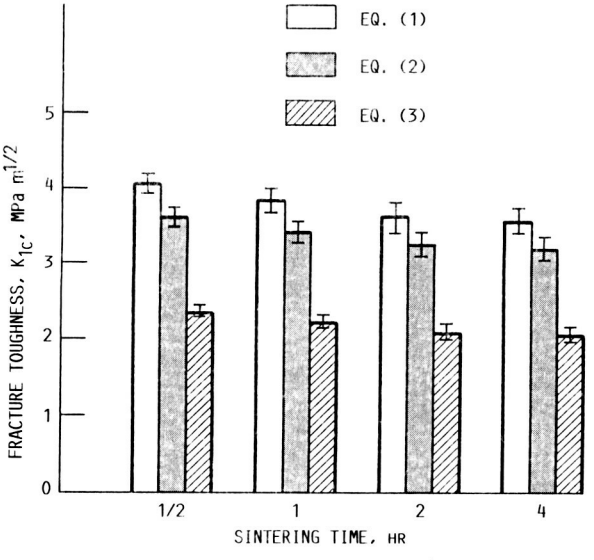


FIGURE 5. - FRACTURE TOUGHNESS OF α-SiC SINTERED AT 2150 °C FOR DIFFERENT TIMES.

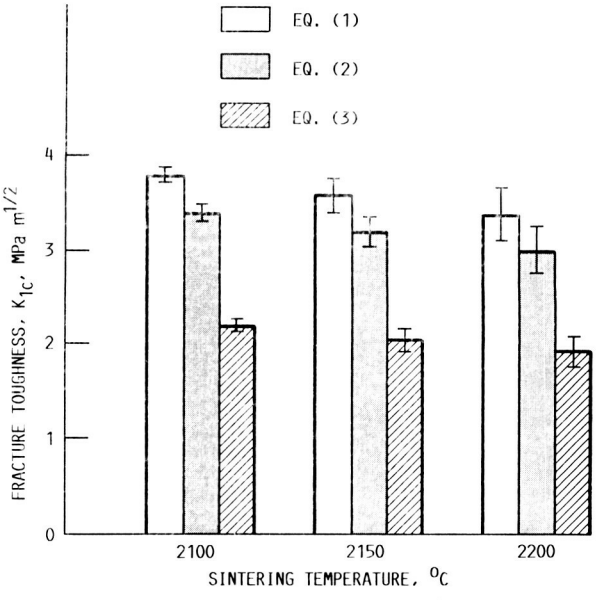


FIGURE 6. - FRACTURE TOUGHNESS OF α-SiC SINTERED FOR 4 HOURS AT DIFFERENT TEMPERATURES.

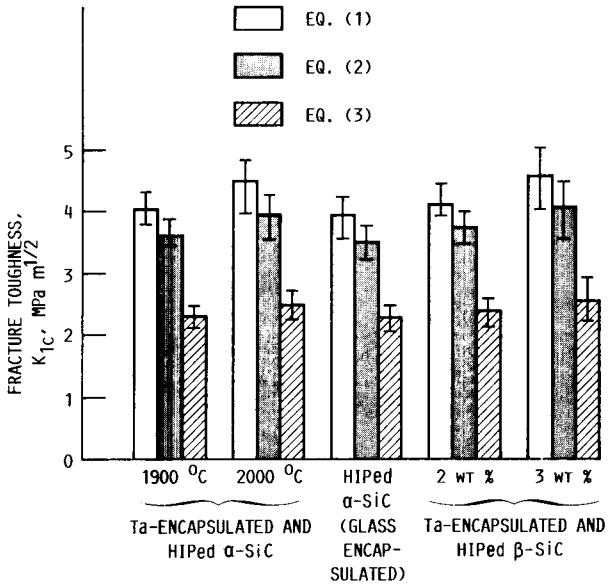


FIGURE 7. - FRACTURE TOUGHNESS OF HIPed α- AND β-SiC MATERIALS. EACH BAR REPRESENTS 10 TESTS.

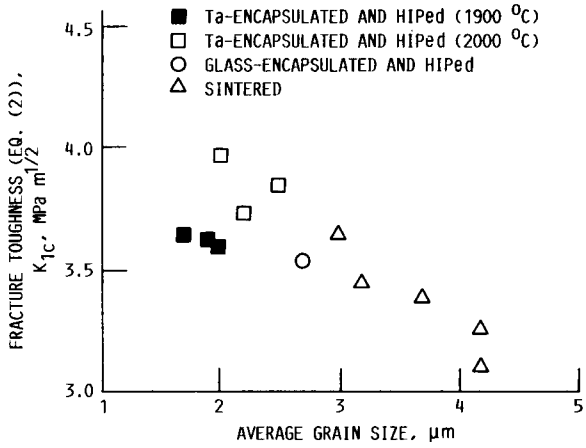


FIGURE 8. - EFFECT OF PROCESSING ON GRAIN SIZE AND FRACTURE TOUGHNESS OF α-SiC.

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15. Supplementary Notes

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16. Abstract

Several α-SiC materials were processed by hot isostatic pressing (HIPing) and by sintering an α-SiC powder containing boron and carbon. Several β-SiC materials were processed by HIPing a β-SiC powder with boron and carbon additions. The fracture toughnesses K_{Ic} of these β- and α-SiC materials were estimated from measurements of Vickers indentations. The three formulas used to estimate K_{Ic} from the indentation fracture patterns resulted in three ranges of K_{Ic} estimates. Furthermore, each formula measured the effects of processing differently. All three estimates indicated that fine-grained HIPed α-SiC has a higher K_{Ic} than coarsed-grained sintered α-SiC. Hot isostatically pressed β-SiC, which had an ultrafine grain structure, exhibited a K_{Ic} comparable to that of HIPed α-SiC.

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