

Microstructural Characterization and Influence of Ceramography Method on the Microhardness of Sintering Agents Added Silicon Carbide

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This study carried out the microstructural characterization, by light microscopy, of sintered SiC in the presence of liquid phase at temperatures of 1800, 1850 and 1900 °C, added with Al₂O₃ and Y₂O₃ as well with Al₂O₃-YAG composite in the proportions of 5, 10 and 15% by weight. It was possible to observe the formation of microstructural patterns resulting from the heterogeneous segregation of the additives, such observation is associated to the pulling of additives during the initial stage of ceramography. Two ceramographic paths were followed: method I - surface grinding followed by manual polishing and method II - automatic grinding and polishing. In order to compare the two techniques the analysis of 3D roughness by light interferometry and the Vickers microhardness test were used. It was concluded by the analysis of variance with 95% confidence that the degree of finishing obtained by both methods allowed the adequate microstructural characterization, as well as, to meet the requirements established by the ASTM C1327-15 standard for hardness test.

Keywords: *Ceramography, Surface grinding, SiC, Hardness, 3D roughness*

1. Introduction

The SiC is present in several applications requiring high hardness, resistance to ballistic impact, abrasion, corrosion and chemical inertness. It is used in the manufacture of abrasives, cutting tools, mechanical seals and heat exchangers¹. In particular, SiC may be used in the preparation of samples for microscopy characterization. This preparation, generally called materialography, is specifically named ceramography in the case of SiC. For materialography, diamond is the most widely used abrasive, followed by SiC, whose granule is used as a coating for grinding paper for roughing stage of sample preparation. In view of the impossibility of performing ceramography SiC therewith, the literature indicates the use of diamond disks for the preparation of advanced ceramics².

Similar to grinding, the rectification is a process of abrasion machining, in which material is removed by the action of ceramic abrasive grains of extreme hardness, without orientation and irregular shape. Rectification deserves attention in the mechanical manufacturing for its capacity to ensure a great finish and high dimensional and geometric accuracy, compared to other machining processes³.

To evaluate the hardness of SiC by Vickers and Knoop techniques it is required a specular surface with 0.1 μm roughness^{4,5}. Noting the high hardness and brittle behavior of ceramic materials, the occurrence of pull-out is frequent, which may result in an incorrect microstructural analysis due to the larger difficulty in identifying pores, measuring the grain size and to determine the hardness. Changing grinding and polishing parameters, such as time and pressure, these effects can be minimized. Therefore, it is recommended the use of automated equipment, which provides full control of the operating conditions and becomes possible to obtain a suitable surface in a reasonable time^{6,7}.

The objective of this research is perform to microstructural characterization of α-SiC through mechanical preparation by grinding and polishing, as well as to study automatic methods of preparation samples for the Vickers microhardness test and roughness measurements.

2. Material and Methods

In this study a green type α-SiC, manufactured by Saint Gobain, Brazil, was supplied with 97.72% purity and particle size distribution d_{90} of 2 μm. The Al₂O₃, a Calcined Alumina APC-2011 SG, Alcoa Alumínio S/A, Brazil, was supplied

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with d_{90} of 5.80 μm and 99.40% purity. The Y_2O_3 , a REO type, by Alfa Aesar, Brazil, was supplied with 99.9% pure and d_{50} of 4.40 μm .

One mixture of Al_2O_3 and Y_2O_3 (63.65 and 36.35% by weight) and other mixture of Al_2O_3 -YAG composite (36.30 and 63.70% by weight) were incorporated to the SiC as additives for sintering in presence of liquid phase in the proportion of 5, 10 and 15% by weight. The composite was produced by solid state reaction at 1400 °C as eutectic composition, in a ratio of 36.30 to 63.70 wt% of Al_2O_3 and YAG, and then crushed in a planetary mill^{8,9}. Sintering of SiC occurred in tube furnace with an inner lining of graphite, without powder bed, at 1atm in argon atmosphere for 30 min, at temperatures of 1800, 1850 and 1900 °C. Table 1 shows the schematic denomination of the groups formed according to the percentage of additives and sintering temperature.

The apparent porosity (P) of the sintered samples was calculated using ABNT NBR 6220, based on the Archimedes principle. The masses were measured with an accuracy of 10^{-4} g, at constant temperature, with a Mettler balance, model AE 200.

For microstructural characterization of the material presented in Table 1 it was employed the ceramographic route referred to as method I, which used a flat surface grinder with cylinder wheel, RC-310 model, by Machinery Industry Chinelatto Ltda. The grinding steps were 0.2 to 0.3 mm, ending with 0,005 mm, for a total operation time 20 to 30 min per sample. Subsequent manual polishing was carried out in a polishing Arotec APL-4 with diamond paste in grit sizes of 15, 9, 6, 3, 1 and 0.25 μm using DP-Blue lubricating fluid, from Struers, diluted in equal parts of water distilled water and ethanol. The best polishing conditions was obtained at minimum speed and 3,3 N per sample. The polishing average time was 24 hours. The route that proved to be efficient for the commercial sintered SiC, called method II¹⁰, was also applied to the SO2B group using the Presi Mecatech 334 automatic polishing machine. The grinding was done with diamond discs of 54 and 18 μm applying a force of 4 N and angular speed of 150 rpm, at the base and head in opposite directions, for approximately 5 min. The polishing operation used diamond grits in suspension of 6 and 1 μm with a solution of water and alcohol in 50% as a lubricant, with 300 rpm for base and 150 rpm for the head in opposite directions, and forces of 10 N. The time for polishing was 30 and 110 min, respectively.

Materials chemically stable as SiC, are difficult to etch¹¹. Since in this case, the etching produces no significant contrast gain, this step can be suppressed, and the microstructure of the material can be perfectly observed by optical microscope after suitable mechanical preparation, but without observation of grain boundaries¹¹.

The microstructure was observed with the aid of a reflection optical microscope Olympus BX 60M. Topographic surface features and the level of finish of both methods could be

obtained in a Zygo[®] NewView[™] 7100 white light profiler by 3D roughness analysis. The data were collected from a surface area of 0.47 x 0.35 mm with 20X magnification, in five different points.

The Vickers microhardness (HV) test was conducted to evaluate reliability of the methods I and II in accordance with ASTM C1327-15, with ten indentations, on the durometer Micromet Buhler with 1000 gf load. By the standard, the root mean square roughness (R_{ms}) must not exceed 0.1 μm . The R_{ms} value represents the standard deviation of the distribution of the peaks on the surface, which is a more sensitive parameter than the average roughness¹². For the roughness analysis and hardness testing only the SO2B group was investigated. The statistical analysis of variance was done with the aid of Microsoft Excel[®], considering the values of hardness in HV and roughness in μm .

3. Results and Discussion

Both ceramography procedures by methods I and II provided similar reflection and specular morphology in the range of 50 to 1000X magnification. For this reason it was decided to only show micrographs based on the method I.

Figure 1 shows a sequence of images obtained by light microscopy of SO1A sample with successive increased magnification (50 to 1000X) on the same region. The white dotted area square of a micrograph is enlarged in the next figure. The analysis from increases up to 500X revealed the presence of white phase, light gray (matrix), black and dark gray. The dark gray phase had a morphology patterns consisting of continuous lines, like rivers branching, which is visible from 50X magnification as indicated in Figures 1. The phases are indicated by arrows in Figure 2a, which also perceives patterns size difference between the groups.

Figure 3 shows some examples of the SO group under 100X, in which it is observed the dark gray phase. The patterns of this phase, depending on the samples, had either a finer line width (approximately 10 μm), thicker (about 30 μm) or degenerated in alveolar format. Some samples, because of pull-out and presence of porosities, did not show the formation of patterns, while possessing all the aforementioned described phases. Table 2 summarizes the structure of the patterns samples provided by the SO and SE. From this table, it can be seen that the increase in the thickness is directly proportional to the amount of additives and temperature.

Magnifications of 1000X allowed one inferring that both the dark and dark gray phases have the same shape as the light phase, which are connected, as shown in Figure 2. These observations indicated that the dark gray and dark phases are produced by pull-out of the light phase, all being corresponding additives. This condition justifies the increase in thickness of the patterns with the amount of additives and temperature. A rounded dark phase, as indicated by arrows in

Table 1. Formation groups as a function of temperature and percentage by weight of additives.

Sample	Temperature (°C)	Al ₂ O ₃ e Y ₂ O ₃ (wt%)	Al ₂ O ₃ -YAG (wt%)
SO1A	1800	5	-
SO1B	1850	5	-
SO1C	1900	5	-
SE1A	1800	-	5
SE1B	1850	-	5
SE1C	1900	-	5
SO2A	1800	10	-
SO2B	1850	10	-
SO2C	1900	10	-
SE2A	1800	-	10
SE2B	1850	-	10
SE2C	1900	-	10
SO3A	1800	15	-
SO3B	1850	15	-
SO3C	1900	15	-
SE3A	1800	-	15
SE3B	1850	-	15
SE3C	1900	-	15

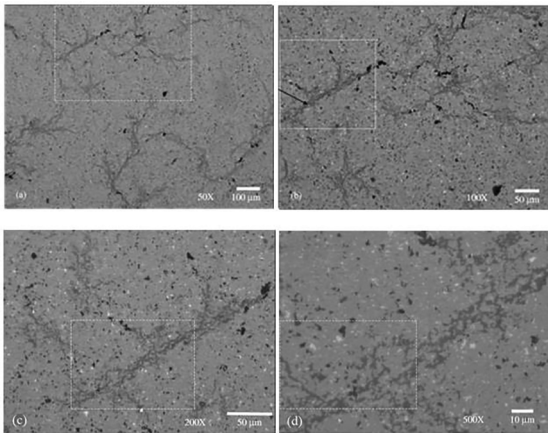
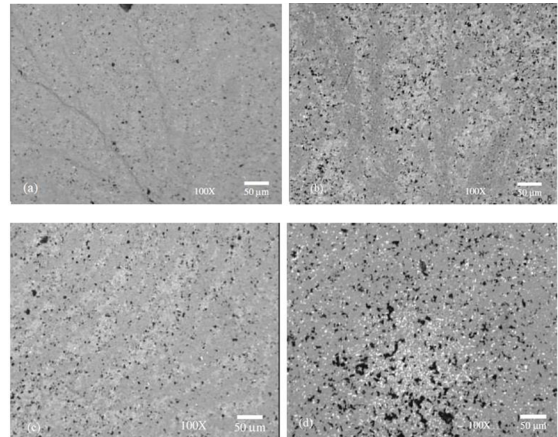
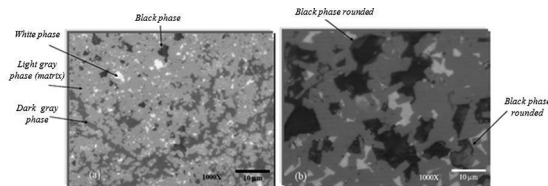
**Figure 1.** Optical Micrograph of SO1A sample with increases of (a) 50X (b) 100X (c) 200X and (d) 500X.**Figure 3.** Optical micrograph at 100X showing patterns of: Fine lines (a) - SO1B; Thick lines: (b) - SO2B; Alveolar: (c) - SO3A; Without pattern: (d) - SO2C.**Figure 2.** Optical micrographs with increased 1000X: (a) SO1A and (b) SO2C.

Figure 2b (SO2C sample at 1000x magnification) indicates the occurrence of more severe pull-out that includes SiC grains.

Also in Table 2 are presented the apparent porosity values for each condition. It was found that the porosity for SO1A, SO2A and SO3A, are respectively 0.37, 0.32 and 0.43%,

that is, one order of magnitude lower than the amount of additives. This is an indication that porosity did not influence this microstructural characterization. The formation of SiC microstructure patterns sintered with Al₂O₃ and Y₂O₃, as shown in Figure 3, has been previously observed¹³. These patterns are derived from heterogeneous segregation of the second phase¹³.

Through the statistical analysis it was verified that there is no significant difference between the methods I and II for the hardness and roughness tests with 95% confidence, as the value of F calculated is smaller than the table, as shown in Table 3. The value of F is calculated by division between the

Table 2. Values of Porosity (P) with its Standard Deviation (SD) and the illustrative pattern scheme of the SO and SE samples.

Sample	P (%)	SD	Fine	Thick	Alveolar	Whitout pattern
SO1A	0,37	0,23	■			
SO1B	1,84	0,60	■			
SO1C	0,32	0,01	■			
SE1A	1,54	0,20	■			
SE1B	3,61	0,32	■			
SE1C	3,73	0,28	■			
SO2A	0,33	0,11	■			
SO2B	3,62	0,29		■		
SO2C	2,94	1,61				■
SE2A	2,67	0,27				■
SE2B	4,96	0,73		■		
SE2C	3,22	0,81		■		
SO3A	0,43	0,17			■	
SO3B	1,24	0,25			■	
SO3C	1,02	0,33				■
SE3A	2,78	0,14				■
SE3B	4,24	0,15			■	
SE3C	1,23	0,39		■		

Table 3. Results of the R_{ms} roughness analysis and the HV1 hardness test with the respective analysis of variance.

Hardness Vickers (GPa)					
Method I			Method II		
SO2B-1	SO2B-2		SO2B-1	SO2B-2	
19,2 ± 0,8	19,8 ± 0,4		23,9 ± 1,4	22,6 ± 1,0	
Causes of variation	DF	SS	MS	F	F table
Treatment	3	1569667,015	523222,3	0,0947	2,84
Residual	36	198993759,7	5527604,4		
Total	39	200563426,7			
Roughness R_{ms} (μm)					
0,077 ± 0,002	0,091 ± 0,004		0,120 ± 0,015	0,086 ± 0,004	
Causes of variation	DF	SS	MS	F	F table
Treatment	3	0,004215	0,001405	0,122	3,24
Residual	16	0,184833	0,01155		
Total	19	0,189048			

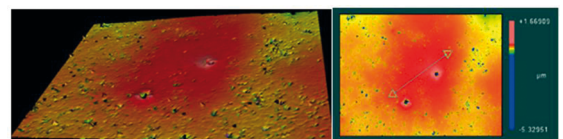
DF: Degrees of freedom; SS: Sum of squares; MS: Middle square.

mean squares for the treatment and residual mean squares. The indentations were made at points that seemed to be denser because of the greater light reflectivity. However, it was not possible to clearly distinguish different phases by optical microscopy.

Further analysis of roughness in the indentation region by linear scan revealed that at such points, the R_{ms} value is even lower, 0,027 μm. The topographic 3D map indicated it is a more homogeneous area with fewer pull-outs (Figure 4).

4. Summary and Conclusion

Two analyzed ceramography procedures, although different, pointed to the same result in the hardness test,


Figure 4. Topographic Map surface and linear scanning near indented area in SO2B group.

meeting the requirements of ASTM C1327-15, and revealed the microstructure without artifacts. In fact, method II was able to reduce total process time and give high productivity, since several samples could be prepared simultaneously under the same parameters. However, roughness analysis showed that methods I and II provided the same degree of surface finish that met the requirements of the hardness test. From the

3D topographic map, it was observed that the indented areas corresponded to free points of pull-out, which is confirmed that it did not significantly interfere in the hardness test. In both methods there was pull-out, whose severity increased with the increase of the amount of additives (5, 10 and 15%) and could not be avoided under the conditions of preparation employed. Even in the groups of samples where the apparent porosity was less than 0.5%, there was the presence of the segregation patterns, which is an indication that these are formed only by pull-out of additives.

5. References

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