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Effect of Acid Etching Time and Concentration on Oxygen Content of Powder on the Microstructure and Elastic Properties of Silicon Carbide Densified by SPS

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Abstract: In this current paper, oxygen content of a fine particle size SiC (H. C. Starck UF 25 Silicon Carbide) and coarser particle size SiC (Saint Gobain Silicon Carbide) were modified by using different concentrations of HF for etching. Fully dense silicon carbide ceramics (>99% th. density) were produced by the spark plasma sintering technique at 1950 °C under an applied pressure of 50 MPa for 5 min hold with boron carbide and carbon addition. Archimedes method, scanning electron microscopy, and the ultrasound analysis were used to examined density, microstructure, elastic (E), shear (G), and bulk (K) moduli of dense silicon carbide ceramics to investigate the effect of oxygen impurities on the densification and the properties of silicon carbide. The results showed that high oxygen content is detrimental to the final density of SPS silicon carbide. When the oxygen content increased from 0.60 to 5.92 wt.%, the relative density decreased from 99.99% to 96%. For both SiC powders, by increasing the etching time, the grain size of SiC decreased. It means that the high oxygen caused grain growth. Ultrasound analysis results showed that the high oxygen content affected the elastic properties. SiC samples with the high oxygen content had a lower elastic moduli, shear moduli and bulk moduli. It was clear that increasing the oxygen content decreased the elastic properties.

Keywords: Silicon Carbide, Spark Plasma Sintering, Microstructure

1. Introduction

Silicon carbide (SiC) has some outstanding physical and chemical properties, such as low theoretical density (3.21 g/cm^3), a high hardness, a high elastic modulus, high thermal conductivity, good wear and oxidation resistance, and low coefficient of thermal expansion [1-8]. Due to those properties, dense silicon carbide is suitable to use in high temperature, corrosion resistant applications and aerospace applications, automotive, wear components and armor [3-4, 9-10]. However, it is difficult to obtain a sintered body with high density without applied pressure or sintering aids, due to high covalent bonds between silicon and carbon atoms and low self diffusion coefficient [1, 10-11].

The other issue with the non-oxide high temperature ceramics such as SiC, B_4C and TiB_2 is that they tend to have an oxide layer on their surfaces. It depends on particle size, moisture in the air, and additives. This oxide layer causes large grain coarsening and inhibits densification [12-15].

There are a number of methods to sinter SiC, for instance, pressureless sintering, hot pressing (HP), spark plasma sintering (SPS) with solid state or liquid phase sintering [10, 16]. In liquid phase sintering of silicon carbide, Al_2O_3, Y_2O_3, and other rare earth oxides are commonly used as additivies to form liquid phase [17, 18]. When using oxide sintering aids in liquid phase sintering, the silica layer (SiO_2) on the SiC particles reacts with the other oxides to help form of the liquid phase. Since the Gibbs free energy of SiO_2 is strongly negative, the oxygen layer is on the SiC particle and the oxide layer will inhibit the densification [1, 17].

In solid state sintering of silicon carbide, boron containing species and carbon are used as sintering aids. Boron base

additives increase the rate of self-diffusion coefficients of Si and C. Carbon removes the SiO_2 layer on the silicon carbide surface and also inhibits the grain growth by forming inclusions [19-23].

Magmami et. al. sintered SiC with boton carbide and carbon addition at 1980°C for 7 hours with pressureless sintering, and density reached to 98.4% [24].

Sajgalık et. al. produced >99% dense additive free SiC with the hot press technique at 1850°C for 1 hour holding time under 30 MPa pressure [11].

SPS is a novel method that can produce dense silicon carbide a within short time with high heating rate and pressure. Due to the shorter holding times it can inhibit large grain growth [1-2, 10, 25-26].

Hayun et. al. densified SiC without additive at 2050°C for 10 min with an applied pressure of 69 MPa via SPS. 99% of theoretical was achieved with this method [27].

Guillard et. al. have also reported that SiC was densified only 92% at 1850°C with a 5 min holding time under 75 MPa pressure by SPS method without additives [26].

Thus far, researchers have investigated different methods to produce fully dense silicon carbide. However, the effect of oxygen content of silicon carbide on propertiesof dense ceramic has not been addressed as necessary.

The aim of this work was to address the influence of the acid etching process on oxygen content of starting powders on the microstructure and elastic properties of dense silicon carbide. To achieve this goal, two different silicon carbide powders' acids were etched using varying HF concentration, etching time, and neutralizing media and the change in oxygen contents were observed. The spark plasma technique was used to densify silicon carbide powders with boron carbide and carbon additives.

2. Experimental

Four different raw materials were used for this research including H. C. Starck silicon carbide (UF-25, H. C. Starck GmbH&Co., Germany), Saint Gobain silicon carbide (Niagara Falls, New York), HC. Starck boron carbide (HD-20, H. C. Starck GmbH&Co., Germany), and carbon (Lamp black from Fisher Scientific) as starting materials. The Saint Gobain powder was not commercially available; it was obtained directly from the Saint Gobain, Niagara Falls, New York plant. The main differences between these two powders were their particle sizes and oxygen contents. The Saint Gobain (SG) powder had larger average particle size of 1.5 μm. H. C. Starck (HC) had a smaller average particle size of 0.5 μm. In the content, HC-SiC will refer fine SiC, and SG-SiC will refer coarse SiC. A LECO TC600 oxygen/nitrogen analyzer was used to determine the oxygen content of silicon carbides. To reduce oxygen content of powders both powders were acid etched.

To modify the oxygen content of powder fine silicon carbide was etched with 20%, 40%, 50% HF, and HF&NHO3 acid for 1, 4, and 24 hours. The acidic slurry was neutralized with ethanol and ammonium hydroxide.

Table 1. Acid etching conditions.

Acid	Concentration (%)	Etching Time (Hour)	Neutralized with
HF	20	24	Ethanol
HF	40	1-4-24	Ammonium Hydroxide
HF	50	1-4-24	Ammonium Hydroxide
HF & HNO_3	40-65	1-4-24	Ammonium Hydroxide

60 g SiC powder was mixed with 200 ml different concentrations of HF (Acros Organics) and stirred for 1, 4, or 24 hours in a Nalgene HDPE beaker, and neutralized until it reached pH 7, then centrifuged to separate the liquid from the silicon carbide powder. Afterwards, wet silicon carbide was washed with deionized water three times and silicon carbide powder dried in a drying oven. Using a mortar and pestle the dry powder cake was ground to fine powder.

As a result of these experiments, 1 hour etching with 50% HF and neutralization with ammonium hydroxide was chosen. Coarse SiC powder was etched only with this method.

The matrix of samples consist of B_4C, C, and SiC. 0.5 wt.%B_4C, 1.5 wt.%C, and SiC were weighed and put in a Nalgene bottle with SiC ball and ball milled for 24 hours in ethanol. After milling, the mixture was sieved to separate media from the liquid mixture using a mesh sieve (1.4mm), dried on a hot plate at 275°C, and ground to uniformity with mortar and pestle.

All samples were then sintered by Spark plasma sintering (Thermal Technology). 5 grams of each powder mixture was loaded into a graphite die (20 mm inner diameter) with graphite punches. The inside of the graphite die was lined with graphite foil. Samples were sintered using a two stage sintering. SPS was heated up to 1400°C with a 200°C/min heating rate under vacuum with an applied pressure of 50MPa and it was held at 1400°C for 1 minute. After intermediate holding, the samples were heated up to 1950°C at 200°C/min, under 50MPa applied pressure and held for 5 min. After that, the SPS was shut down and the samples were allowed to cool.

After sintering, to remove excess graph foil from the surface, the dense samples were sandblasted. Surface grinding was necessary after sand blasting to produce a smooth and flat surface for ultrasound analysis to measure elastic properties. The density was measured using the Archimedes method after surface grinding. Then, samples were cut close to the center of the sample using a LECO Vari/Cut 50 diamond saw. A small piece was chosen and mounted with epoxy using a Buehler SimpliMet 1000 mounting press. Samples were then polished down to 0.25 μm finish using the Buehler- Ecomet 250-Grinder-Polisher. To highlight SiC grain boundaries, a modified Murakami method (20g KOH and 20g K3Fe (CN) 6 in 60 ml DI water) was used to etch (for 4.5 min) the pieces of the polished samples. Etched pieces were washed with acetone and ethanol to remove residual salt. The Zeiss Sigma field emission scanning electron microscope was used to define grain size, and shape of grains. Grain sizes were measured by

the linear intercepts using Lince 2.4.2. image analysis software. The ultrasound analysis method was used to measure the elastic properties.

3. Results and Discussion

LECO TC 600 oxygen/nitrogen analyzer was used to measured oxygen content of both SiC powders. Each oxygen content result represents the mean of three analyses. The oxygen content of fine SiC was 1.69±0.04 wt% before the acid etching process. The acid etching conditions and oxygen content of fine SiC after etching process is shown in Table 2. While increasing the concentration of HF, it also increased the efficiency of acid etching to remove the oxide layer. The oxygen content of SiC powder for 20% HF, 40%HF, and 50% HF for 24-hours etching was 1.09±0.04 wt%, 0.75±0.03 wt%, and 0.48±0.03 wt% respectively. When the longer etching time was used, the oxygen content of the etched silicon carbide powder slightly decreased. Nevertheless, increasing the etching time of powder did not decrease the oxygen content as much as expected. The oxygen content of powder for 40% HF and 50% HF for 1 hour, 4 hours, and 24 hours was 0.82±0.01 wt%, 0.81±0.01 wt%, 0.75±0.03 wt%, 0.60±0.02 wt%, 0.56±0.02 wt%, and 0.48±0.03 wt%, respectively. In the time frame, 1 hour etching was more effective thus SiC powder etched for 1 hour with 50% HF was chosen despite the lower oxygen content in 50% HF for 4 and 24-hour etching. Etching silicon carbide powder with 75% HF & 25% HNO_3 did not decrease oxygen content; in contrast, it increased the oxygen content of powder because HNO_3 acted as a strong oxidizing agent. The oxygen content of powder for 1 hour, 4 hours and 24 hours was 1.45±0.02 wt%, 5.07 ±0.06 wt%, and 5.92±0.04 wt%, respectively.

Table 2. Acid etching conditions and oxygen content of fine SiC powder.

Acid	Concentration (%)	Etching Time (Hour)	Neutralized with	Oxygen Content (wt.%)
HF	20	24	Ethanol	1.09±0.04
HF	40	1	Ammonium Hydroxide	0.82 ±0.01
		4		0.81 ±0.01
		24		0.75±0.03
HF	50	1	Ammonium Hydroxide	0.60±0.02
		4		0.56±0.02
		24		0.48±0.03
HF & HNO_3	40-65	1	Ammonium Hydroxide	1.45±0.02
		4		5.07±0.06
		24		5.92±0.04

After acid etching, oxygen content of fine SiC powder's dramatically decreased to 0.60±0.02 wt% from 1.69±0.04 wt%. When the acid etching process removed the oxide layer from powder, it has also created new surfaces on the powders which tend to oxidize. The oxide layer forms on the surface of freshly etched powders as soon as it is exposed to the air. For that reason, the oxygen content of powders cannot be lowered to 0%.

Coarse SiC had 0.38±0.01 wt% oxygen content before the acid etching treatment. The powder was etched with 50% HF

for 1 hour, and oxygen content was measured as 0.20±0.01 wt%. To determine the lifetime of the powder, acid etched dry powder was allowed to age at ambient conditions. The oxygen growth curve of acid etched coarse SiC can be seen in Figure 1. The oxygen content of acid etched SiC powders was measured at different times, and after 14 days oxygen content increased to 0.31±0.01 wt% from 0.20±0.01 wt%.

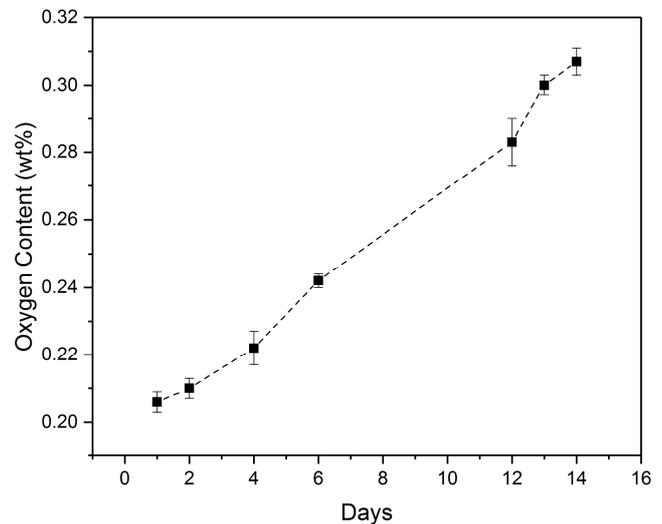*Figure 1. Oxygen growth curve for Saint Gobain SiC.*

Fine SiC (H. C. Starck)

Figure 2 showed microstructure images of 40% HF, 50%HF, and HF and HNO_3 etched fine silicon carbide. Density, grain size and elastic properties of dense fine SiC results can be seen in Table 3. Each value represents the mean of five measurements. SEM images showed that samples made with fine silicon carbide powder had mainly elongated grains. As the etching time increased, the grain size decreased. For 40%HF etched samples, the grain size changed from 3.57μm to 2.80μm as the etching time increased. The 1 hour (3.57μm) and 4 hour (3.02μm) 40% HF etched samples showed some exaggerated grain growth with several micron grains, while the 24 hour (2.80μm) 40% HF etched sample shows smaller grains. The 50% HF etched samples showed little difference in grain size with decreased oxygen content. The 1 hour, 4 hour, and 24 hours samples had a 2.89μm, 2.71μm, and 2.63μm grain size respectively. The 40%HF etched samples had bigger grain sizes than the 50% HF etched samples since the oxygen content of the starting powder was higher. The HF and HNO_3 etched samples showed different morphologies since their oxygen content was higher than the 40% HF and 50% HF etched samples. The 1 hour HF and HNO_3 etched sample had mostly elongated grains, the 4 hour and 24 hour HF and HNO_3 etched samples had small grains with visible porosity. 1 hour, 4 hour and 24 hour HF and HNO_3 etched samples had a 6.68μm, 1.75μm, and 1.57μm average grain size respectively. To be consistent, 1.5 wt.% carbon was added to all the samples' matrix. The 4-hour and 24-hour HF and HNO_3 etched SiC powders had 5.07 wt.% and 5.92 wt.%

oxygen content respectively. Since these two powders had a high oxygen content, the amount of additional carbon was not enough to remove residual oxygen; thus, the samples could not achieve high density.
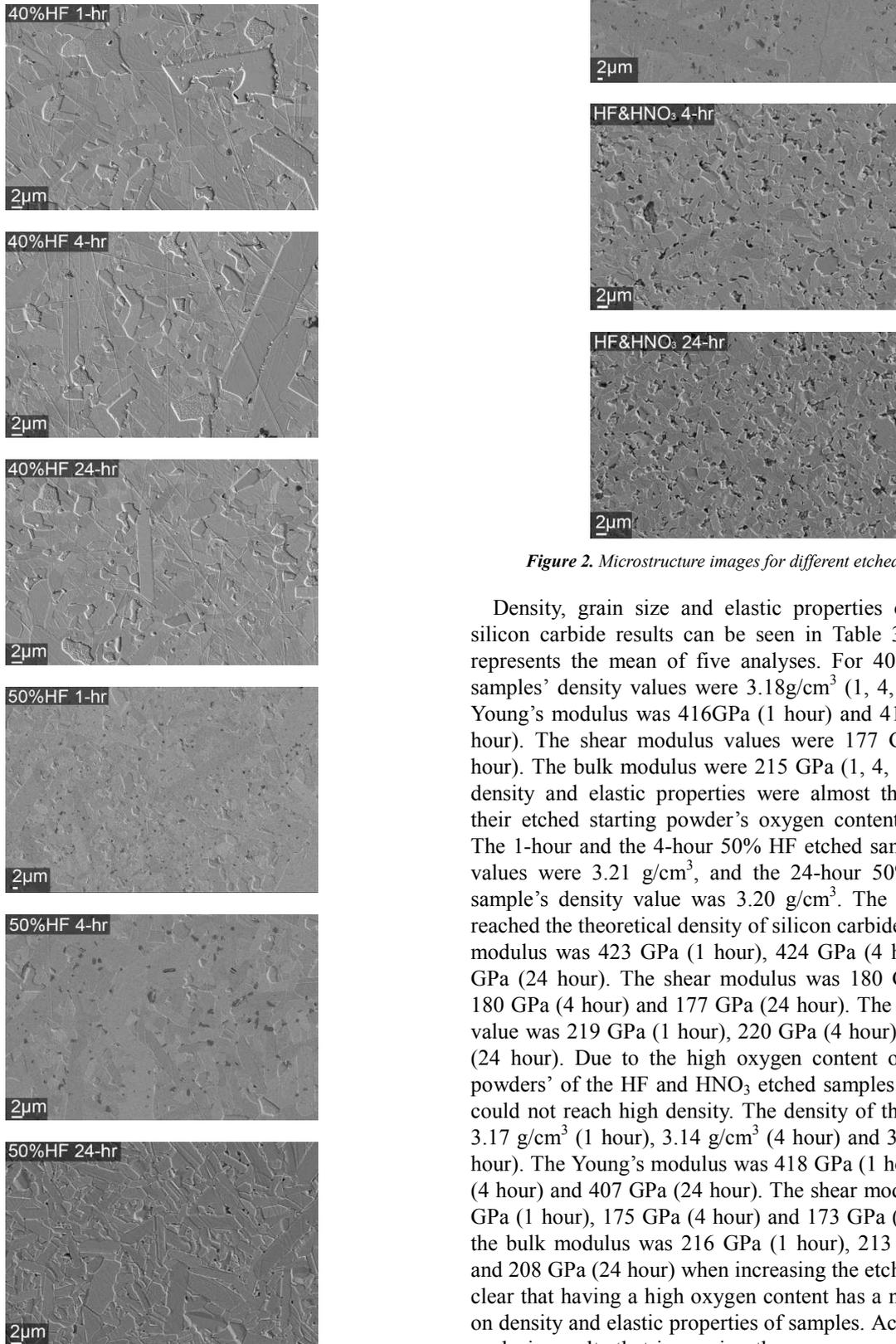

Figure 2. Microstructure images for different etched fine SiC.

Density, grain size and elastic properties of dense fine silicon carbide results can be seen in Table 3. Each value represents the mean of five analyses. For 40% HF etched samples' density values were 3.18g/cm^3 (1, 4, 24 hour), the Young's modulus was 416GPa (1 hour) and 417 GPa (4, 24 hour). The shear modulus values were 177 GPa (1, 4, 24 hour). The bulk modulus were 215 GPa (1, 4, 24 hour). The density and elastic properties were almost the same since their etched starting powder's oxygen content was similar. The 1-hour and the 4-hour 50% HF etched samples' density values were 3.21 g/cm^3, and the 24-hour 50% HF etched sample's density value was 3.20 g/cm^3. The samples have reached the theoretical density of silicon carbide. The Youngs modulus was 423 GPa (1 hour), 424 GPa (4 hour) and 417 GPa (24 hour). The shear modulus was 180 GPa (1 hour), 180 GPa (4 hour) and 177 GPa (24 hour). The bulk modulus value was 219 GPa (1 hour), 220 GPa (4 hour) and 216 GPa (24 hour). Due to the high oxygen content of the starting powders' of the HF and HNO$_3$ etched samples', the samples could not reach high density. The density of the sample was 3.17 g/cm^3 (1 hour), 3.14 g/cm^3 (4 hour) and 3.09 g/cm^3 (24 hour). The Young's modulus was 418 GPa (1 hour), 412 GPa (4 hour) and 407 GPa (24 hour). The shear modulus was 178 GPa (1 hour), 175 GPa (4 hour) and 173 GPa (24 hour), and the bulk modulus was 216 GPa (1 hour), 213 GPa (4 hour) and 208 GPa (24 hour) when increasing the etching time. It is clear that having a high oxygen content has a negative effect on density and elastic properties of samples. Accordingly, the analysis results that increasing the oxygen content decreases the density, and consequently, the samples that have a lower

density will have greater porosity as shown in literature [8]. Having porosity in samples reduces the elastic properties of samples. Wilhelm et al. also mention that oxygen content of silicon carbide was one of the variables to affect mechanical properties [28].

Table 3. *Density, grain size and elastic properties of dense fine SiC.*

Sample	Density (g/cm³)	Average Grain Size (μm) (Std. Dev)	E (GPa)	G (GPa)	K (GPa)
40%HF-1-hr	3.180±0.002	3.57±1.30	416±8	177±4	215±4
40%HF-4-hr	3.180±0.002	3.02±0.61	417±8	177±4	215±4
40%HF-24-hr	3.180±0.001	2.80±0.89	417±8	177±4	215±4
50%HF-1-hr	3.210±0.001	2.89±0.99	423±9	180±4	219±4
50%HF-4-hr	3.210±0.001	2.71±0.82	424±9	180±4	220±4
50%HF-24-hr	3.200±0.001	2.63±0.79	417±8	177±4	216±4
HF&HNO₃- 1-hr	3.170±0.001	6.68±1.39	418±8	178±4	216±4
HF&HNO₃- 4-hr	3.140±0.002	1.75±0.26	412±8	175±4	213±4
HF&HNO₃- 24-hr	3.090±0.002	1.57±0.44	407±8	173±4	208±4

Coarse SiC (Saint Gobain)

Coarse SiC samples' microstructural images can be seen in Figure 3. It showed slight differences in grain size, however the grain morphology appeared to be similar between samples. All samples showed equiaxed grain shape, and with increasing the oxygen content, the average grain size of samples increased. Sample produced with freshly etched powder (day 0) had 1.68 μm average grain size, while the higher oxygen content sample (day 9) had 3.34 μm average grain size. It showed that the oxygen content of the starting powder affects the grain size of the materials. This result was also supported with the literature, when Vassen et. al. mentioned that oxygen content of silicon carbide caused grain growth and inhibited the densification [29].
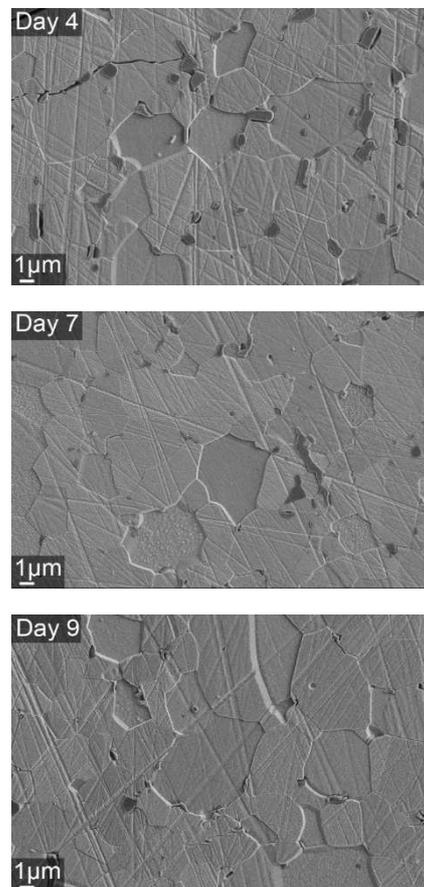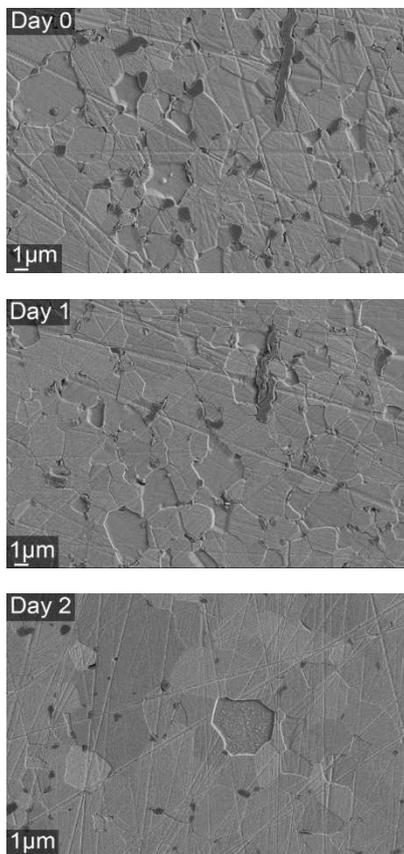

Figure 3. *Microstructure images of aged coarse SiC for different days.*

Density, grain size and elastic properties of dense coarse silicon carbide results can be seen in Table 4. The density of the sample changed from 3.17 g/cm³ to 3.14 g/cm³, the Young's modulus decreased from 417 GPa to 407 GPa, the shear modulus changed from 177 GPa to 173 GPa, and the bulk modulus dropped from 217 GPa to 210 GPa while aging the powder. It was clear that increasing the oxygen content had a negative effect on the density and elastic properties of samples. Increasing the oxygen content decreased the density and elastic properties values.

***Table 4.** Density, grain size and elastic properties of dense coarse SiC for different days.*

Sample	Density (g/cm^3)	Average Grain Size (μm) (Std. Dev)	E (GPa)	G (GPa)	K (GPa)
Day 0	3.170±0.001	1.68±0.46	417±8	177±4	217±4
Day 1	3.160±0.002	1.85±0.33	415±8	176±4	215±4
Day 2	3.160±0.001	2.69±0.41	413±8	175±4	214±4
Day 4	3.160±0.002	2.73±0.81	412±8	175±4	213±4
Day 7	3.160±0.003	3.07±1.16	411±8	174±4	213±4
Day 9	3.140±0.002	3.34±0.93	407±8	173±4	210±4

4. Conclusion

In this research, a fine particle size SiC (H. C. Starck UF 25 Silicon Carbide), coarser particle size SiC (Saint Gobain Silicon Carbide) were densified using SPS with boron carbide and carbon additives. The oxygen content of the starting powders was modified by acid etching and adding carbon. It showed that when the concentration of hydrofluoric acid increased, the efficiency of acid etching also increased to remove the oxide layer. Moreover, with increasing the etching time, the oxygen content of the etched powder decreased. Since the 1 hour etching process was more effective in the time period, the silicon carbide powder etched for 1 hour with 50%HF was selected.

SEM images of fine SiC (H. C. Stark silicon carbide) showed that they had mainly elongated grains. In addition to this, by increasing the etching time, the grain size decreased. It means that the high oxygen content of powders caused grain coarsening. Ultrasound analysis results showed that the high oxygen content also affected the elastic properties and density. Sample with the high oxygen content had a lower Young's modulus, shear modulus and bulk modulus, and they could not reach full density due to the oxide layer.

The coarse SiC (Saint Gobain) powder was etched only for 1 hour with 50% HF, and the powders were sintered at different times to see the effect of oxygen content of powder. SEM images of coarse silicon carbide showed that they had equiaxed grain shape, however the grain sizes changed by day. Sample made with freshly etched powder (day 0) had smaller grain sizes while higher oxygen content powder (day 9) showed bigger grain sizes. Sample made with the lowest oxygen content also had higher density and mechanical properties (Young's modulus, shear modulus, and bulk modulus) than higher oxygen content powders. It was clear that increasing the oxygen content decreased the density and elastic properties.

Acknowledgements

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