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Thermal Shock Resistance of Liquid Phase Sintered SiC

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Abstract. The aim of this study was to evaluate the effect of Y_2O_3:Al_2O_3 additives and sintering temperature on thermal shock resistance of silicon carbide sintered via liquid phase. Silicon carbide samples containing 10 mol% Y_2O_3:Al_2O_3 (1:3 and 1:4) were prepared, compacted and sintered at 1750, 1850 and 1950 °C in a graphite resistive furnace. Thermal shock resistance was evaluated after each thermal cycle performed at 600, 750 and 900 °C followed by abrupt cooling in water. Samples with two Y_2O_3:Al_2O_3 proportions did not show major differences when sintered at the same temperature, though, rising the sintering temperature improves Y_2O_3:Al_2O_3 modified-SiC thermal shock resistance.

Introduction

Silicon based ceramics, such as silicon carbide and nitride, exhibit high mechanical resistance at high temperatures as well as low thermal expansion coefficient and high thermal conductivity, which leads to a good thermal shock resistance [1,2], although degradation of materials resistance under sudden temperature gradient represents an essential factor for the determination of the material operating life. Silicon carbide behavior with regard to thermal shock resistance is a theme of great interest due to applications where reliability about rapid temperature variations is needed. Among these applications, it is possible to highlight gas turbines, aircraft propulsion systems, refractories and cutting tools [3].

Rapid temperature variations induce thermal tensions that cannot be relieved by plastic deformation and lead to formation and propagation of crack. Rapid cooling of a specimen is more likely to inflict such thermal shock than heating, because the induced surface stresses are tensile. Crack formation and propagation from surface flaws are more probable when an imposed stress is tensile [4,5].

In order to evaluate material damages caused by thermal shock, several parameters may be adopted: number of thermal cycles required for the beginning of cracking, mass variation after determined the number of cycles, porosity variation with respect to the number of cycles, flexural strength before and after thermal shock, crack propagation by an indentation test, and also monitoring of properties like Young's modulus and permeability [6,7,8].

In this paper, it was studied how the proportion of Y_2O_3:Al_2O_3 additives and different sintering parameters may influence silicon carbide's thermal shock resistance. In this case, both number of cycles and temperature were pre-determined and the thermal shock resistance was evaluated by means of the modulus of elasticity and rupture.

Experimental

Samples were prepared with 90 mol% SiC (BF17, H.C. Starck with 90% β-SiC) and 10 mol% Y_2O_3:Al_2O_3. The mixture of additives was prepared by fixing yttria (99% HC Starck, Grade C) to alumina (99.8% CT 3000 SG Almatis) molar proportion at 1:3 and 1:4, and sample designation as FYAG and FY4, respectively.

The previous reaction between Y_2O_3 and Al_2O_3 was conducted by means of oxides mixture for 2 h; powder compaction in the shape of 60 x 12 x 5 mm bars; thermal treatment at 1100 °C for 1 h; grinding of the bars; drying at 110 °C; powder deagglomeration and sieving. After additives previous reaction, mixtures containing SiC were prepared in an Atrittor mill for 4 h at 300 rpm.

Ceramic suspensions were dried in rotary evaporator and, after deagglomeration, compacted as 60 x 6 x 3 mm bars for uniaxial and isostatic pressing. All samples were placed over protection powder bed in a graphite resistive furnace (Astro 1000, FP20) and sintered with a heating rate of 15 °C/min at 1750, 1850 and 1950 °C for 1 h under argon atmosphere. Density of sintered samples was obtained based on Archimedes principle and microstructural characterization was performed with a Philips XL 30 scanning electron microscope.

For thermal cycle test, samples were heated in a tubular furnace for 30 min and then, rapidly immersed in water at room temperature. Cycles were performed at three temperatures: 600, 750 and 900 °C. Then, modulus of elasticity was determined by a non-destructive natural frequency vibration method [9] after each cycle and the modulus of rupture was determined after 1, 25 and 50 cycles.

Results and Discussion

By taking into account the associated standard deviation values (Table 1), it is possible to verify that changing Y_2O_3:Al_2O_3 molar proportion from 1:3 to 1:4 is not sufficient to exert a significant modification over the values of density and modulus of elasticity.

Table 1: Samples density and modulus of elasticity after sintering.

Sample	Relative Density (%)			Modulus of Elasticity (GPa)		
	1750 °C	1850 °C	1950 °C	1750 °C	1850 °C	1950 °C
FYAG	81.5±2.1	95.3±1.0	94.9±0.6	209.4±14.8	327.4±9.5	323.5±11.7
FY4	82.8±1.8	95.2±1.0	95.2±0.5	227.9±12.7	325.3±6.9	320.9±9.3

Samples that were sintered at 1750 °C showed lower values of density and elastic modulus, which is very consistent, since pre-existing defects, such as micro-cracks and pores, are responsible for the reduction in density and modulus values [5,10]. As the sintering temperature reaches 1850 ºC, a significant improvement on both properties is achieved, nevertheless, no noteworthy differences were observed between samples sintered at 1850 and 1950 ºC. Modulus value was determined after each cycle of thermal shock at 600 °C, 750 °C and 950 °C by adopting 1, 25 and 50 cycles for each sample. The obtained result was expressed by relative loss of modulus after each cycle (E/E_0).

Regardless of the initial modulus of elasticity and sintering temperature, for thermal shock at 600 °C (Fig. 1 - 3), an initial reduction of the modulus was observed after the first cycle, followed by stabilization which remained until 25 and 50 cycles. The modulus stabilization feature is given by equation [11]:

$$E(N) = E_0 - A(1 - \exp(-\alpha N)) \qquad (1)$$

Where E(N) is the Young's modulus as a function of the number of thermal shock cycles (N) in a given temperature difference (ΔT), E_0 is the Young's modulus free of micro-cracks, and A is a saturation constant that is defined as E_0-E_{sa}t, wherein E_{sat} is a stationary state of the modulus carried out after a determined number of cycles and α is a velocity constant that measures the rate with which thermal fatigue reaches saturation. This stabilization occurs due to formation of short cracks, which are interpreted as fissures which length is comparable to grains size or other microstructural characteristics.

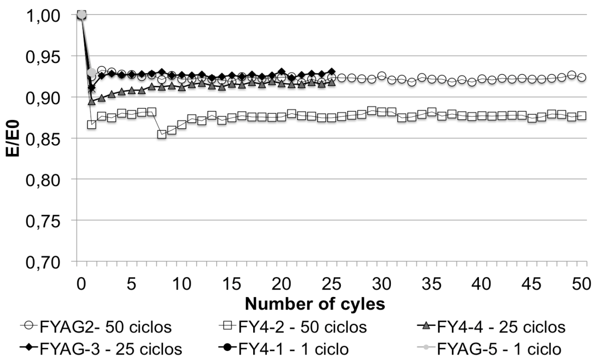


Fig. 1: Modulus of elasticity after thermal cycles (600 °C) for samples sintered at 1750 °C.

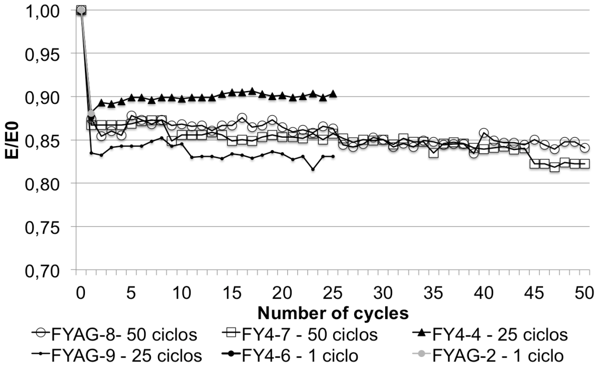


Fig. 2: Modulus of elasticity after thermal cycles (600 °C) for samples sintered at 1850 °C.

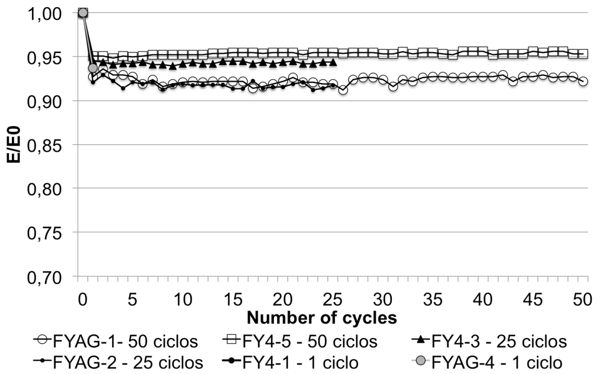


Fig. 3: Modulus of elasticity after thermal cycles (600 °C) for samples sintered at 1950 °C.

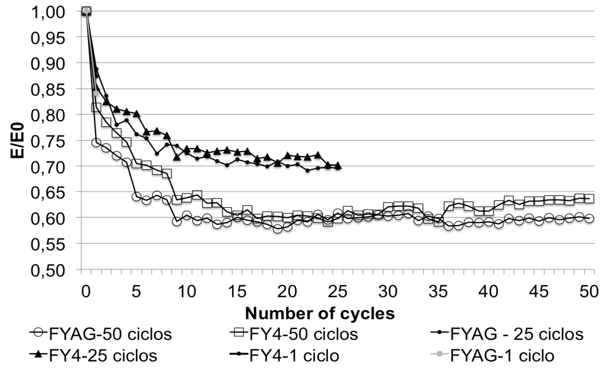


Fig. 4: Modulus of elasticity after thermal cycles (750 °C) for samples sintered at 1750 °C.

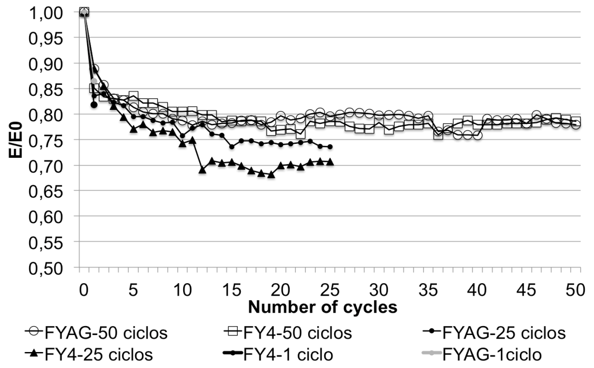


Fig. 5: Modulus of elasticity after thermal cycles (750 °C) for samples sintered at 1850 °C.

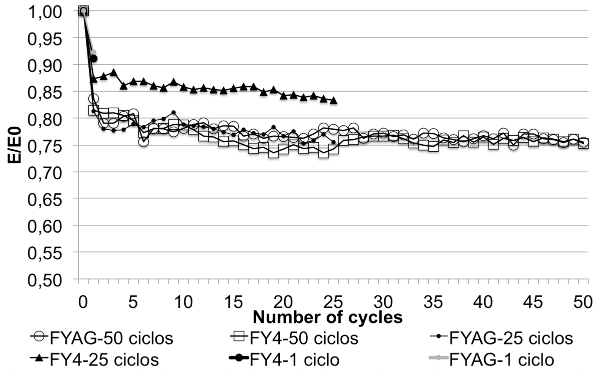


Fig. 6: Modulus of elasticity after thermal cycles (750 °C) for samples sintered at 1950 °C.

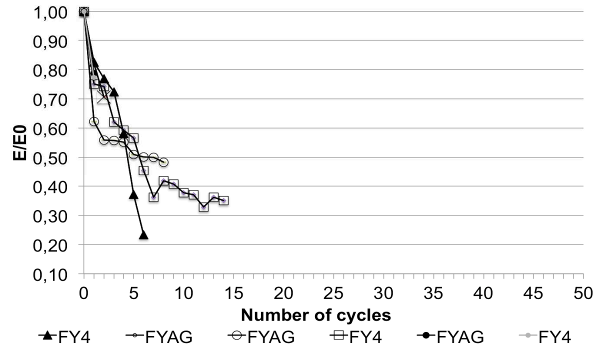


Fig. 7: Modulus of elasticity after thermal cycles (900 °C) for samples sintered at 1750 °C.

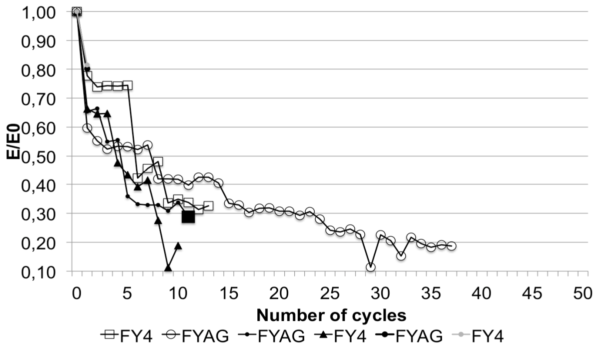
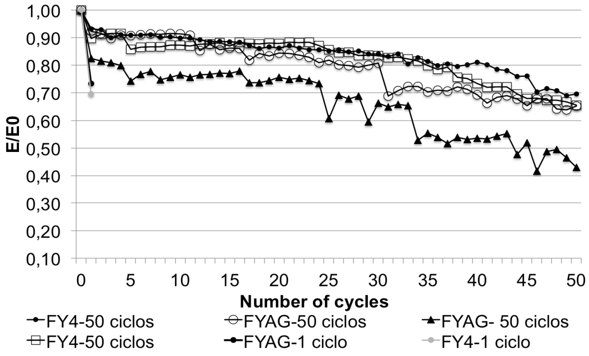


Fig. 8: Modulus of elasticity after thermal cycles (900 °C) for samples sintered at 1850 °C.



(C)

Fig. 9: Modulus of elasticity after thermal cycles (900 °C) for samples sintered at 1950 °C.

The stabilization behavior, represented by Eq. 1, may occur for a less severe thermal shock, because more severe cases generate crack coalescence and even more significant fractures [11]. Thus, the temperature of 600 °C (Fig. 1 - 3) does not constitute the worst thermal shock condition. Results of thermal cycles conducted at 750 °C (Fig. 4 - 6) also show a stabilization behavior, nevertheless, this behavior occurs after a greater number of cracks, indicating the formation of more micro-cracks compared to cycles at 600 °C.

The lowest loss of modulus after 50 thermal cycles comes at 600 ºC for all tested samples, independently of the sintering temperature, for E/E_0 remains above 0.80 whereas samples tested at 750 ºC, even showing a stationary behavior, achieved values of E/E_0 below 0.80. Some of these values reached around 0.60, e.g. samples sintered at 1750 °C.

Samples FYAG and FY4 sintered at 1750 °C showed the worst performance at all temperatures of thermal cycle. Those tested at 900 °C presented the shortest operating life in comparison to the other samples. The results are directly related to the initial characteristics since these are the ones with the lowest density.

The results of cycles at 900 °C (Fig. 7 - 9) show a distinct behavior in relation to the other temperatures, because stabilization does not occur with regard to samples sintered at 1750 °C and 1850 °C, indicating a significant reduction of the modulus of elasticity with the rise of thermal cycles. This led to specimens rupture which compromised measurement of the modulus frequency and made it difficult to monitor samples at 25 and 50 cycles. These results evidence that 900 °C is a condition wherein severe thermal fatigue takes place.

When this thermal fatigue phenomenon occurs, stabilization behavior does not happen due to the unstable growth of cracks as a result of the rise in cycle temperature. In this case, the microstructural interactions associated with short crack fatigue behavior are exceeded by the long crack behavior [2,11] It is possible to observe the presence of cracks in samples that did not withstand thermal shock cycles at 900 °C (Fig. 10).

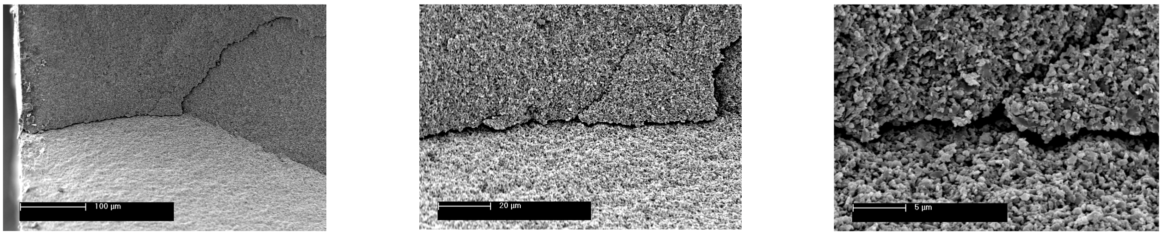
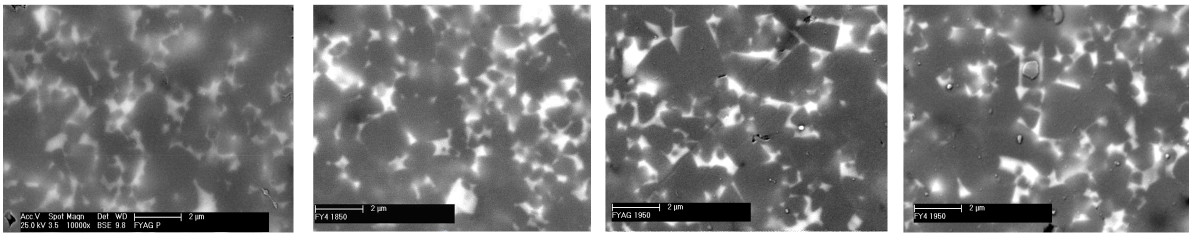


Fig. 10: Results of SEM examination of FY4 sample sintered at 1750 °C and fractured at 900 °C.

Samples sintered at 1950 °C are the only ones that withstand to 50 cycles of thermal shock at 900 °C, whereas samples sintered at 1850 °C show a major loss on the modulus of elasticity after the first thermal shock cycle. As these samples present a similar behavior with respect to density and the initial modulus, this difference might be related to microstructure, since samples sintered at 1950 ºC have a larger grain size (Fig. 11), determined by studying grain size distribution with Quantikov software.



(A) FYAG 1850 °C (B) FY4 1850 °C (C) FYAG 1950 °C (D) FY4 1950 °C

Fig. 11: Results of SEM examination of sintered samples.

According to Kalantar and Fantozzi [12], a microstructure with elongated grains and higher aspect ratio may generate some toughness mechanisms caused by crack deflection or bridging. In these mechanisms, a great amount of energy is consumed for crack propagation, reducing the way it travels and, consequently, the damage induced by thermal shock. Therefore, this leads to an enhanced resistance to samples sintered at 1950 °C.

The modulus of rupture was determined before and after thermal shock cycles (1, 25 and 50 cycles), as shown in Table 2. By considering the relevant number of samples that fractured over the cycles performed at 900 °C, the modulus was determined only for 1 cycle in samples sintered at 1750 °C and 1850 °C; 1 and 50 cycles in samples sintered at 1950 °C.

Table 2: Modulus of rupture after 1, 25 and 50 cycles of thermal shock at 600, 750 and 900 °C.

Sample	Temperature (°C)	Modulus of rupture (MPa)								
		Before cycles	Cycles at 600 °C			Cycles at 750 °C			Cycles at 900 °C	
			1	25	50	1	25	50	1	50
FYAG	1750	134.2	34.6	34.1	26.2	32.5	25.3	26.6	33.2	**
FY4		145.0	41.2	33.8	30.9	44.5	24.6	26.4	38.1	**
FYAG	1850	275.5	44.0	30.4	31.4	45.7	26.7	20.9	43.6	**
FY4		283.2	47.5	46.1	50.6	49.3	27.7	26.7	37.0	**
FYAG	1950	263.7	79.3	69.0	69.8	69.0	27.5	23.8	70.4	20.1
FY4		267.7	75.3	78.3	70.7	71.2	29.7	21.5	84.6	24.3

For the cycles of thermal shock at 600 °C, the modulus of rupture presents a behavior similar to the modulus of elasticity, once a more pronounced loss occurs with one cycle of thermal shock. For the other cycles, there is no major reduction on the modulus regardless of the sintering temperature. Further, it is possible to observe that samples sintered at 1750 °C and 1850 °C show greater resistance to decay in comparison to samples sintered at 1950 °C with one cycle.

Samples submitted to thermal shock at 750 °C show a decline on the modulus of rupture at the first cycle, and this also occurs between cycles 1 and 25. When analyzing the damage caused by thermal shock only by means of the modulus of rupture, it is difficult to observe in which cycle stabilization begins, as it also occurs with the modulus of elasticity.

For the cycles of thermal shock at 900 °C, a reduction on modulus was observed between cycles 1 and 50 in relation to samples sintered at 1950 °C. This behavior is similar to the modulus of elasticity, indicating that 900 ºC induces severe thermal fatigue. According to the results, it is possible to verify that rising the temperature of thermal shock leads to an unstable growth of cracks and material failure, in accordance with literature [7].

Independently of the thermal shock cycle temperature, the major decline on modulus of rupture occurs at the first cycle with a reduction over 70%, which is higher when compared to the behavior of the modulus of elasticity and evidences that the onset of the first micro-cracks substantially influences the modulus of rupture.

Conclusions

Samples containing Y_2O_3:Al_2O_3, with molar proportion fixed at 1:3 and 1:4, do not show major differences in relation to thermal shock, when sintered at the same temperature.

Temperatures of 600 and 750 °C do not represent the worst thermal shock conditions for SiC samples modified with Y_2O_3:Al_2O_3, though a significant initial decline on both modulus of elasticity and rupture was observed at 750 °C independently of the sintering temperature. No stabilization behavior was observed after cycles performed at 900 °C, indicating that this might be the worst condition of thermal shock wherein an unstable growth of cracks occur.

Independently of cycle temperature, samples FYAG and FY4 showed the best performance against thermal shock when sintered at 1950 °C. This samples presented greater grain sizes, which might have led to toughness mechanisms, thus reducing the damage caused by thermal shock. Finally, the temperature value of 1950 °C is the most appropriate to be used on sintering of SiC modified with Y_2O_3:Al_2O_3.

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