

## ENHANCING THE DENSITY OF SILICON CARBIDE WITH THE ADDITION OF NITRATE-BASED ADDITIVES

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### Abstract

Dense monolithic silicon carbide (SiC) was successfully sintered by hot pressing at 1750 °C for 1 h under an applied pressure of 20 MPa with the addition of a nitrate-based additive. With the addition of MgO-Y<sub>2</sub>O<sub>3</sub> and Al<sub>2</sub>O<sub>3</sub>-Y<sub>2</sub>O<sub>3</sub> in nitrate form, a relative density of more than 98% was achieved, while in the oxide form it was 85.0 and 96.0%, respectively. In fact, MgO-Y<sub>2</sub>O<sub>3</sub> showed poor densification due to the eutectic temperature of 2110 °C, however, the addition of the nitrate form of MgO-Y<sub>2</sub>O<sub>3</sub> greatly enhanced the densification. The sintering mechanism in the nitrate-based additive is liquid phase sintering, identified by the presence of an oxide phase, i.e., Y<sub>2</sub>O<sub>3</sub> in the SiC with the addition of Al<sub>2</sub>O<sub>3</sub>-Y<sub>2</sub>O<sub>3</sub> in nitrate form. Moreover, the addition of nitrate form suppressed the grain growth of SiC, which was believed to be due to the adequate rearrangement stage during sintering.

**Keywords:** Silicon carbide; Nitrate; Oxide; Sintering additive; Density

### 1. Introduction

Structural application at high temperature needs material that can withstand harsh conditions, such as oxidation and corrosion, and maintain the mechanical properties. Indeed, advanced ceramics are suitable for those conditions, and one of the advanced ceramics that attracts enormous attention is silicon carbide (SiC) [1]. The utilization of SiC is quite broad, from the abrasive material like sandpaper to the sophisticated application in the fusion reactor [2–5]. Thanks to the highly covalent bonding of SiC which gives excellent resistance in the chemical and thermal, as well as outstanding mechanical properties [5,6]. However, like other covalent bonding materials, the sintering of SiC is practically challenging due to the low diffusivity of Si and C. Although the densification of SiC is also possible without sintering additives [7–10], the sintering condition of those SiC is in harsh conditions. For instance, a sintering temperature > 1800 °C is needed to obtain near the theoretical density of SiC [7,8,10]. Therefore, the sintering additive plays an important role in enhancing the

density of SiC at a relatively lower temperature and mild pressure.

Generally, the oxide additives, such as Al<sub>2</sub>O<sub>3</sub> and/or Y<sub>2</sub>O<sub>3</sub>, are well-known sintering additives for SiC. The sintering mechanism with the addition of those additives is liquid phase sintering. The additive forms a liquid phase during sintering due to the reaction between the additives or the additive itself melting during sintering. The liquid phase acts as a path for the SiC particle diffusion, enhancing the densification. The other additives that are effective for the sintering of SiC are Y<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>-Y<sub>2</sub>O<sub>3</sub>-MgO, Al<sub>2</sub>O<sub>3</sub>-Y<sub>2</sub>O<sub>3</sub>-CaO, Sc<sub>2</sub>O<sub>3</sub>, Ho<sub>2</sub>O<sub>3</sub>, Yb<sub>2</sub>O<sub>3</sub>, and Al<sub>2</sub>O<sub>3</sub>-RE<sub>2</sub>O<sub>3</sub> [11–21]. Certainly, one of the key factors that is important in the liquid phase sintering is the homogeneous distribution of the liquid phase, which corresponds to the particle size of the additives [22]. The effort to increase the density of SiC by improving the homogeneous distribution of the sintering additive is reported by Lidén et al. (1995), which found that the sintering additive in the form of sols is better than the powder form for enhancing the density of SiC [23]. Another report showed that the use of sintering

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additives obtained by the coprecipitation method [19] improved the density and suppressed the grain growth. For instance, the average grain size of oxide and oxide from coprecipitation was 540 and 360 nm [24], respectively. The simple way to obtain the fine particle size of the additive is mechanical milling. The relative density of SiC with the addition of  $\text{Sc}_2\text{O}_3$  (average particle size  $> 2 \mu\text{m}$ ) was 85.4%, which can be enhanced by the addition of milled  $\text{Sc}_2\text{O}_3$  (average particle size = 118 nm) to 93.5% [16], showing the effectiveness of fine particle size to enhance the density of SiC. Instead of decreasing the particle size of additives, the use of another form of additives is interesting to investigate. Huang et al. [25] reported the first nitrate additive to enhance the densification of SiC. They used  $\text{MgO-Al}_2\text{O}_3\text{-Y}_2\text{O}_3$  as a sintering additive in the nitrate form and revealed the relative density of 98.1% with the addition of nitrate compared to 97.7% in the oxide form. Further study was reported by Noviyanto et al. [26], which used rare earth nitrate as a sintering additive. They found that Sc, Yb, Em, Er, and Ho were effective as a sintering additive for SiC, which showed a near theoretical density of SiC after sintering at 1750 °C for 1 h.

Based on those perspectives, the present study examined the effect of nitrate additive to enhance the densification of SiC. The additives used in this study were  $\text{MgO-Y}_2\text{O}_3$ , both in oxide and nitrate form.  $\text{MgO-Y}_2\text{O}_3$  is a well-known sintering additive for  $\text{Si}_3\text{N}_4$  [27–30]; however, it is rarely used in SiC. We also investigated the  $\text{Al}_2\text{O}_3\text{-Y}_2\text{O}_3$  additive in oxide and nitrate form for comparison purposes.

## 2. Experimental

Commercial SiC powder (NanoAmor Inc., USA) with an average particle size of 52 nm was used in this study. The sintering additives used in this study were  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ,  $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ , and  $\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ . SiC powder was mixed with sintering additives using ball milling for 24 h and ethanol as a medium. After mixing by ball milling, the mixed powder was dried in a rotary evaporator at 70 °C and 80 rpm to maintain the homogeneity of the mixed powder. The ratio between SiC and sintering additives was 95:5 in weight. A binary system of additives was used, and the composition of additives was calculated based on the oxide form. According to the eutectic point, the weight ratio of  $\text{Al}_2\text{O}_3\text{:Y}_2\text{O}_3$  was 80:20, and  $\text{MgO:Y}_2\text{O}_3$  was 55:45. Prior to sintering, the SiC powder containing nitrate additives was calcined at 500 °C for 2 h in a nitrogen atmosphere to decompose the nitrate. In addition to the nitrate-based additives, the oxide-based sintering additive was also prepared for comparison. Sintering was performed in a hot-pressing furnace at 1750 °C for 2 hours under an applied pressure of 20 MPa in an argon atmosphere.

The morphology of  $\beta\text{-SiC}$  particle was characterized using high-resolution transmission electron microscope (HR-TEM, Tecnai G2 F20 S-twin, FEI, USA). The density of sintered SiC was estimated using the Archimedes principle. After sintering, the morphology of the sample was observed using a scanning electron microscope (SEM, S-4800, Hitachi, Japan). Finally, the phase of sintered SiC was identified using X-Ray Diffraction (XRD, X'Pert, PANalytical, Netherlands).

## 3. Results and discussion

Table 1 shows the density of sintered SiC with the addition of nitrate and oxide-based sintering additives. The density of sintered SiC with the addition of nitrate-based sintering additives enhanced remarkably compared to the oxide-based additives, showing the densities of sintered SiC  $\geq 3.20 \text{ g/cm}^3$ , corresponding to the  $> 98\%$  of relative density, as shown in Table 1. In the oxide form of additive, the density of sintered SiC with the addition of  $\text{Al}_2\text{O}_3\text{-Y}_2\text{O}_3$  (AY(O)) and  $\text{MgO-Y}_2\text{O}_3$  (MY(O)) was 3.11 and 2.78  $\text{g/cm}^3$ , respectively. The eutectic temperature of  $\text{Al}_2\text{O}_3\text{-Y}_2\text{O}_3$  and  $\text{MgO-Y}_2\text{O}_3$  was 1820 and 2110 °C, respectively. Therefore, the poor densification in the MY(O) can be explained by the liquid phase of eutectic composition that did not form yet at sintering temperature 1750 °C. Instead of the formation of the eutectic phase, the liquid phase might be formed due to the reaction between additive and  $\text{SiO}_2$ . Since the particle size of SiC in this study is very fine, the formation of  $\text{SiO}_2$  on the surface of SiC is inevitable [31]. Figure 1 presents the thin layer of  $\text{SiO}_2$  on the surface of  $\beta\text{-SiC}$  particle characterized by HR-TEM.

The eutectic temperature of  $\text{MgO-SiO}_2$ ,  $\text{Al}_2\text{O}_3\text{-SiO}_2$ , and  $\text{Y}_2\text{O}_3\text{-SiO}_2$  is 1548, 1587, and 1660 °C, respectively, which is lower than the sintering temperature at present study. However, although MgO

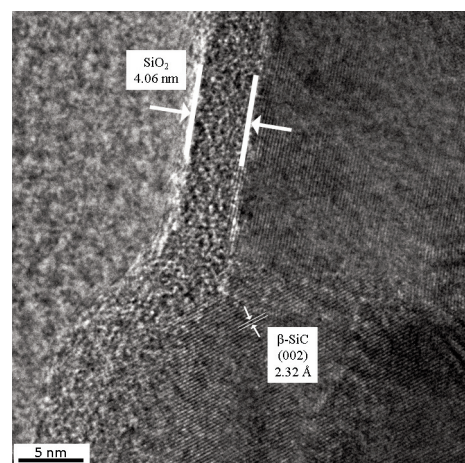


Figure 1. HRTEM image of  $\beta\text{-SiC}$  particle showing the thin layer of  $\text{SiO}_2$  on the surface



**Table 1.** Chemical composition of magnesite

Sintering additives	Sample name	Theoretical density (g/cm <sup>3</sup> )	Density (g/cm <sup>3</sup> )	Relative density (%)
Al <sub>2</sub> O <sub>3</sub> -Y <sub>2</sub> O <sub>3</sub> nitrate	AY(N)	3.26	3.21	98.5
MgO-Y <sub>2</sub> O <sub>3</sub> nitrate	MY(N)	3.27	3.21	98.2
Al <sub>2</sub> O <sub>3</sub> -Y <sub>2</sub> O <sub>3</sub>	AY(O)	3.26	3.13	96.0
MgO-Y <sub>2</sub> O <sub>3</sub>	MY(O)	3.27	2.78	85.0

has the lowest eutectic temperature with SiO<sub>2</sub> among the other additives, the sintered density of SiC with MY(O) is the lowest in this study. Therefore, the densification of SiC in the oxide-based additive is more likely due to the effectiveness of each additive. For instance, AY(O) densification is more likely considerably influenced by Al<sub>2</sub>O<sub>3</sub>. The effectiveness of Al<sub>2</sub>O<sub>3</sub> as solely additive to enhance the density of SiC was also reported by Noviyanto and Yoon (2012) [32], showing the relative density of SiC was 99% by the addition of 5 wt.% of Al<sub>2</sub>O<sub>3</sub>. On the other hand, the addition of 5 wt.% MgO or Y<sub>2</sub>O<sub>3</sub> only increased the relative density up to 90%, which is lower compared to Al<sub>2</sub>O<sub>3</sub>. The presence of Y<sub>2</sub>O<sub>3</sub>, both in AY(O) and MY(O), did not influence densification significantly, which was indicated by poor densification in the MY(O). Therefore, it can be concluded that Al<sub>2</sub>O<sub>3</sub> is more effective in enhancing the density of SiC in this study compared to MgO and Y<sub>2</sub>O<sub>3</sub>. On the other hand, sintered SiC with the addition of nitrate-based additive has a relative density higher than 98%. As shown in Table 1, AY(N) and MY(N) have a similar density of 3.21 g/cm<sup>3</sup>. In the oxide form of additive, Al<sub>2</sub>O<sub>3</sub> plays an important role in enhancing the density. However, it seems all additives in nitrate form

contribute equally to enhance the density, showing almost similar relative density.

Figure 2 shows the SEM images of sintered SiC with various nitrate and oxide additives. Dense microstructures were observed in the SiC with the addition of AY(N) (Figure 2 (a)) and MY(N) (Figure 2 (c)). These results are in agreement with Table 1, whereas the SiC with the addition of nitrate-based additive showed relative density > 98%. It should be pointed out that excessive grain growth occurred during the sintering of SiC with the addition of AY(O) compared to AY(N), as shown in Figures 2 (a) and (b), respectively. For the sintered SiC with the addition of MY(O), as shown in Figure 2 (d), the densification did not occur completely, showing the existence of both very fine particle SiC and partial densification. As shown in the red dash line Figure 2 (d), the localized densification for the sintered SiC with MY(O) is the evidence of the inhomogeneous distribution of the sintering additive. Moreover, the very fine particle size in the MY(O) is another fact that the SiC particle diffusion did not occur even though at sintering temperature 1750 °C. As stated in the introduction, the self-diffusivity of SiC particles is very tough due to the nature covalent bonding of SiC, which demands the sintering additives to promote densification. The poor densification of SiC with the addition of MY(O) indicates that the stages of densification stop in the arrangement process, which is indicated by the fine particle's existence, as shown in Figure 2 (d). Moreover, it confirms the Ostwald ripening mechanism, whereby the large grain consumes the small grain. Therefore, the nitrate-based additive enhances the densification of SiC compared to an oxide-based additive.

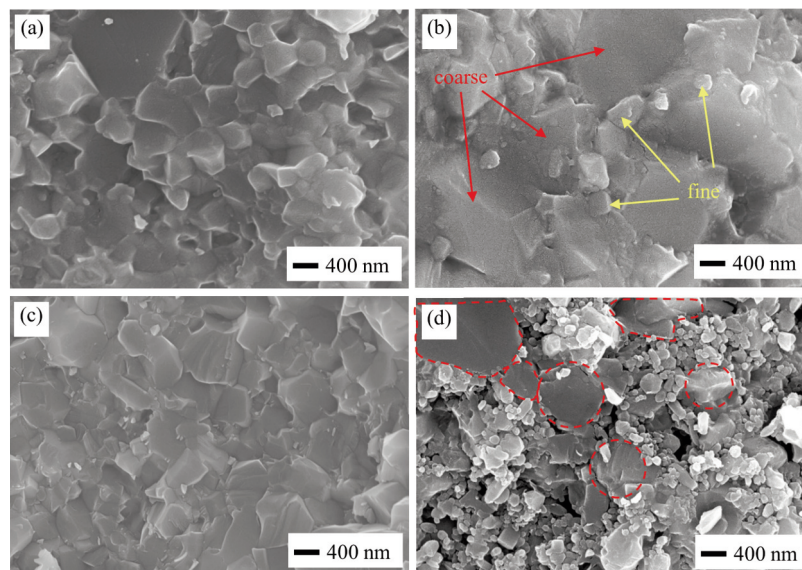
**Figure 2.** SEM images of sintered SiC with the addition of: (a) AY(N), (b) AY(O), (c) MY(N), and (d) MY(O)

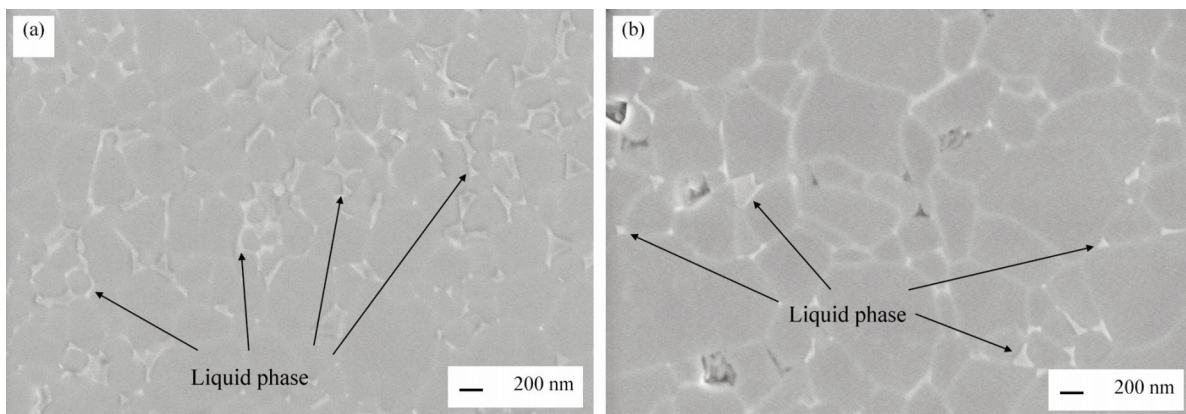
Figure 3 shows the SEM images of the etched surface of sintered SiC with the addition of AY(N) and AY(O). No open-pore was observed in the sintered SiC with the addition of AY(N), as shown in Figure 3 (a), implying the high density of this sample. In contrast, the open pores are observed in the SiC with AY(O) addition, as shown in Figure 3 (b). It should be noted that grain growth can be suppressed by adding AY(N) compared to AY(O). The average grain size of sintered SiC with AY(N) and AY(O) was 442 and 609 nm, respectively. The liquid phase is located in the triple junction in the SiC with the addition of AY(O), as shown in Figure 3 (b), while the addition of AY(N) revealed some of the liquid phase covering the SiC grains. It suggests that grain growth can be suppressed more likely due to grain covering in the SiC with the addition of AY(N), while notable grain growth is observed with the grain that liquid phase located in the triple junction, as shown in Figure 3 (a).

There are three stages of liquid phase sintering: rearrangement, solution-precipitation, and solid-state [22]. Since significant grain growth occurs in the second stage, i.e., solution-precipitation, the degree of grain growth differs between AY(N) and AY(O). It seems the first stage, i.e., rearrangement, is running well for SiC with the addition of AY(N) to obtain adequate density, thus minimizing the grain growth in the second stage. Furthermore, the distribution of additive enhances with the nitrate-based additive, thus increasing the density of SiC [26]. For instance, the relative density of SiC with the addition of Sc-oxide and nitrate was 85.4 and 99.3% [26], respectively. Therefore, that report is in agreement with these results, showing the effectiveness of nitrate additive to enhance the density of SiC.

Figure 4 shows the schematic illustration of the sintering mechanism with the addition of oxide- and nitrate-based additives. As shown in Figure 4, the most important step is dissolving the nitrate additive in the ethanol, which ionizes into a metal ion. The

ionic radius of  $\text{Al}^{3+}$ ,  $\text{Y}^{3+}$  and  $\text{Mg}^{2+}$  was 0.39, 0.9, 0.72 Å, respectively. Indeed, the mixing of SiC particles with ion metals in the solvent leads to the homogeneous distribution of the additives. After removing ethanol, the ion metals become nitrate additive and distribute evenly in the SiC particle, while the poor distribution of additive occurs in the oxide additives. The nitrate additive transforms into metal after calcination [26]. Furthermore, the nitrate additive transforms into oxide due to the reaction between the metal and  $\text{SiO}_2$  on the SiC particle. This result is confirmed in XRD patterns (Figure 5), whereas the  $\text{Y}_2\text{O}_3$  phase was detected in the SiC with the addition of AY(N). Since the nitrate-based additive distributes evenly in the SiC particle, the rearrangement process proceeds smoothly to reach adequate density. The fact about this phenomenon happens in the SiC with the addition of MY(N), while SiC with MY(O) showed a poor rearrangement process identified by the local densification. Sufficient density in the first stage leads to lessening grain growth, showing the grain size of SiC with the addition AY(N) finer compared to AY(O).

Figure 5 shows XRD patterns of sintered SiC with the addition of oxide and nitrate-based additives. No carbide or silicide phases were observed in the sintered SiC, which means the sintering additives used in this study categorize as effective sintering additives [33]. The sintering additives were also not detected for sintered SiC with the addition MY(O), MY(N), and AY(O), as shown in Figures 5 (a) – (c), except for the AY(N) (Figure 5 (d)).  $\text{Y}_2\text{O}_3$  phase was identified in the SiC with the addition of AY(N). The occurrence of this oxide is a confirmation of the two steps reaction, as explained previously—furthermore, the amount of sintering additive is relatively low to detect by XRD after sintering. The phase transformation from  $\beta$ -SiC to  $\alpha$ -SiC appeared in all SiC samples after sintering, which generally occurs at high-temperature sintering.



**Figure 3.** SEM images of etched SiC with the addition of: (a) AY(N) and (b) AY(O), showing the presence of white phase (liquid phase)

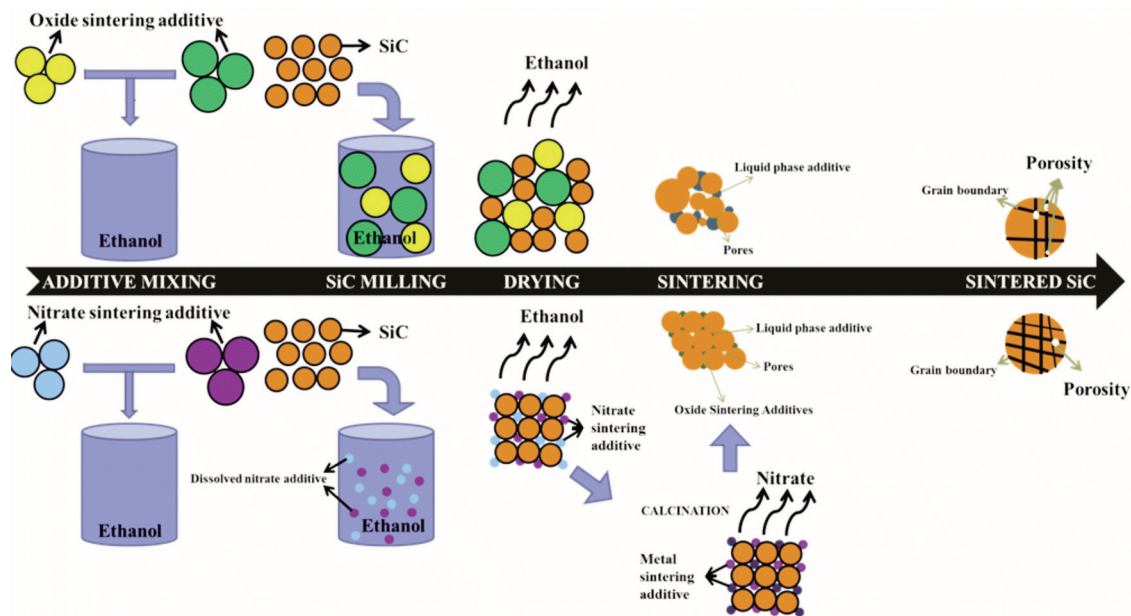


Figure 4. Schematic illustration of SiC densification with the addition of oxide- and nitrate-based additives

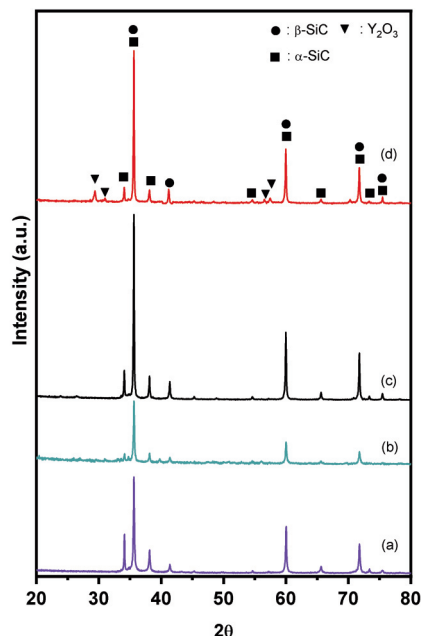


Figure 5. XRD patterns of sintered SiC with the addition of: (a) MY(O), (b) MY(N), (c) AY(O), and (d) AY(N)

#### 4. Conclusion

Dense monolith SiC was successfully fabricated by hot pressing technique with the addition of the nitrate-based additive. Obviously, the density of SiC enhanced with the addition of nitrate-based additive compared to oxide-based additive. The density difference is quite broad, particularly for the MgO-Y<sub>2</sub>O<sub>3</sub>, showing the relative density of 85.0 and 98.2%

for oxide and nitrate-based additive, respectively. It is suggested that the formation of a liquid phase additive is adequate to enhance the rearrangement stage in the SiC with the addition of a nitrate-based additive. Moreover, the nitrate-based additive distributes homogeneously in the SiC particle due to fine ionic size, which dissolves in the ethanol during mixing. To the best of the author knowledge, the use of MgO-Y<sub>2</sub>O<sub>3</sub> additive is rarely reported in the sintering of SiC, therefore the MgO-Y<sub>2</sub>O<sub>3</sub> in nitrate form is an alternative for SiC sintering additive.

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#### Author contributions

MD: Formal Analysis and writing-original draft. DA: discussed the results, writing-review and editing. AN: conceived and design the research, collects data, writing-review and editing, funding acquisition, supervision. AHY: discussed the results, writing-review and editing, supervision. NTR: supervision, writing-review, and editing. All authors have read and agreed to the published version of the manuscript.

#### Data availability

The data of this study are available on request from the corresponding author (AN).



### Declaration of competing interest

The authors declare no conflict of interest.

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## POVEĆANJE GUSTINE SILICIJUM KARBIDA DODATKOM ADITIVA NA BAZI NITRATA

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### Apstrakt

Gusti monolitni silicijum karbid (SiC) je uspešno sinterovan toplim presovanjem na 1750 °C tokom 1 h pod pritiskom od 20 MPa uz dodatak aditiva na bazi nitrata. Dodatkom MgO-Y<sub>2</sub>O<sub>3</sub> i Al<sub>2</sub>O<sub>3</sub>-Y<sub>2</sub>O<sub>3</sub> u nitratnom obliku postignuta je relativna gustina veća od 98%, dok je u oksidnom obliku iznosila 85,0 i 96,0%, pojedinačno. U stvari, MgO-Y<sub>2</sub>O<sub>3</sub> je pokazao slabo zgušnjavanje zbog eutektičke temperature od 2110 °C, međutim, dodavanje nitratnog oblika MgO-Y<sub>2</sub>O<sub>3</sub> u velikoj meri je povećalo zgušnjavanje. Mehanizam sinterovanja u aditivu na bazi nitrata je sinterovanje tečne faze, što se identifikuje po prisustvu oksidne faze, odnosno Y<sub>2</sub>O<sub>3</sub> u SiC sa dodatkom Al<sub>2</sub>O<sub>3</sub>-Y<sub>2</sub>O<sub>3</sub> u nitratnom obliku. Štaviše, dodatak nitratnog oblika potisnuo je rast zrna SiC, za koji se verovalo da je posledica odgovarajuće faze preuređenja tokom sinterovanja.

**Ključne reči:** Silikon karbid; Nitrat; Oksid; Aditiv za sinterovanje; Gustina

