The effect of temperature on phases, density and fracture toughness K_{1C} of SiAlON ceramics

Tran Van Cuong[1*], Ninh Duc Ha[1], Dang Quoc Khanh[2], Nguyen Van Canh[1], Le Thanh Bac[1], Nguyen Tai Truong[1], Trieu Khuong[1], Ngo Minh Tien[1]

[1]Institute of Materials, Biology and Environment, Academy of Military Science and Technology, 17 Hoang Sam, Nghia Do, Hanoi, Vietnam;
[2]HaNoi University of Science and Technology, 1 Dai Co Viet, Hai Ba Trung, Hanoi, Vietnam.
[*]Corresponding author: trancuong.hhvl@gmail.com
Received 15 Aug. 2025; Revised 1 Oct. 2025; Accepted 16 Oct. 2025; Published 18 Nov. 2025.
DOI: https://doi.org/10.54939/1859-1043.j.mst.IMBE.2025.15-21

ABSTRACT

In this study, the effects of temperature on phase formation, density, and fracture toughness (fracture toughness, K1C) of SiAlON ceramics were investigated. The synthesis conditions were optimized at 1650 °C for 4 hours in N_2 environment at 1.0 MPa pressure. The temperature affected the phase composition, density, and fracture toughness, K1C of SiAlON ceramics. The optimal synthesis of SiAlON ceramics showed results: the high density of 3.21 g/cm^3; the low water absorption of 0.20%; the porosity of 0.61% and the fracture toughness, K1C of 5.80 MPa.m$^{1/2}$.

Keywords: Al_2O_3/Y_2O_3 ratio; Temperature; SiAlON ceramics.

1. INTRODUCTION

SiAlON was synthesized from the main components of silicon (Si), aluminum (Al), oxygen (O), and nitrogen (N), developed from Si_3N_4 (Silicon Nitride) material by partially replacing Si^{4+} ions with Al^{3+} and N^{3-} ions with O^{2-} in the crystal lattice. This replacement process is balanced by the presence of cations such as Y^{3+}, La^{3+}, Mg^{2+}, etc. This substitution imparts to SiAlON properties such as high mechanical strength at high temperatures, high thermal shock resistance, high fracture toughness K_{1C}, high wear resistance and corrosion resistance [1-5].

The sintering process of SiAlON ceramics faced challenges due to the strong covalent bond nature of Si_3N_4. Therefore, a liquid phase needs to be formed during the sintering process to promote ion diffusion. Liquid phase sintering aid, such as the mixture of Al_2O_3 and Y_2O_3 was often used to form a liquid phase at the sintering temperature (1700 - 1800 °C), stabilizing the crystal structure, controlling the desired phase [6-9].

The presence of a mixture of Al_2O_3 and Y_2O_3 reduces the sintering temperature and shortens the firing time. Al_2O_3 could react with N_2 under a high Al_2O_3/Y_2O_3 ratio to form AlN, an important component that enhances the mechanical properties and thermal stability of SiAlON. Y_2O_3 could react with Al_2O_3 to form $Y_4Al_2O_9$ (Yttrium Aluminum Monoclinic, YAM) and $YAlO_3$ (Yttrium Aluminum Perovskite, YAP) phases. These phases improve and oxidation resistance of SiAlON. If the content of Y_2O_3 was high, the excessive Y_2O_3 could form yttrium silicate, which reduced fracture toughness and stability of SiAlON [10-15].

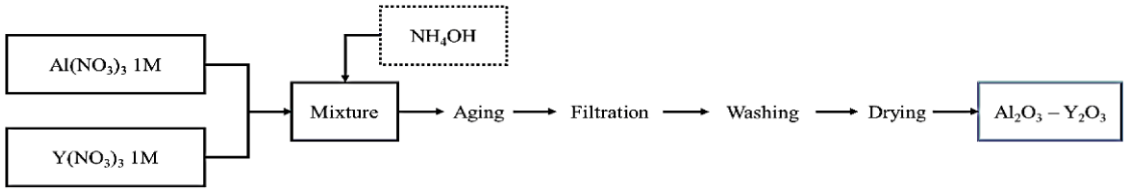
In this study, we synthesized SiAlON with different Al_2O_3/Y_2O_3 ratios to investigate the influence of temperature on phase formation, density, porosity, Water absorption and fracture toughness of SiAlON ceramics.

2. EXPERIMENTAL

2.1. Synthesis process of Al_2O_3 -Y_2O_3 mixture powders

The Al_2O_3 and Y_2O_3 powders were synthesized via a coprecipitation technique. Equimolar (1 M) aqueous solutions of $Al(NO_3)_3 \cdot 9H_2O$ and $Y(NO_3)_3 \cdot 6H_2O$ were prepared and thoroughly mixed.

Aqueous ammonia was added dropwise as the precipitating agent until the solution pH stabilized between 8.1 and 8.3. The resulting suspension was aged under mild agitation at 80 °C for 30 minutes to allow complete precipitation and maturation of the hydroxide phases. The precipitates were then collected via vacuum filtration, washed repeatedly with ethanol and deionized water to remove residual ions, and dried at 100 °C. The dried powders were manually ground using an agate mortar and subsequently calcined at an appropriate Al_2O_3/Y_2O_3 ratio to convert the hydroxides into their respective oxide forms. Al_2O_3-Y_2O_3 was synthesized by the co-precipitation method as shown in scheme 1.



Scheme 1. *The synthesis process of Al_2O_3 - Y_2O_3 mixture.*

The obtained Al_2O_3 and Y_2O_3 powders were weighed according to predetermined molar ratios (table 1) and dispersed in isopropanol. The Al_2O_3/Y_2O_3 ratio was determined by analyzing the Al_2O_3 and Y_2O_3 contents. Al_2O_3 is determined according to TCVN 11659:2016 (ISO 12315:2010) standard. Y_2O_3 is determined by ICP-MS method. The resulting suspension was subjected to planetary ball milling for 30 minutes to ensure fine and homogeneous mixing. The mixture was then dried at ~100 °C to remove the solvent.

Table 1. *Initial composition (%) of the mixture converted to Al_2O_3 and Y_2O_3 oxides.*

No / **Composition**	1	2	3	4	5	6	7	8
Al_2O_3, %	0.0	18.8	25.4	29.2	37.9	43.8	62.9	100.0
Y_2O_3, %	100.0	81.2	74.6	70.8	62.1	56.2	37.1	0.0

2.2. Synthesis of SiAlON ceramics

The synthesis of SiAlON ceramics was carried out by pressure-assisted sintering under N_2. First, all the precursors (Si_3N_4, Al_2O_3 - Y_2O_3 mixture) were wet-mixed in the planetary ball milling machine with the grinding speed of 400 rpm, ball-to-powder ratio of 2:1, water-to-solid mass ratio of 1:1 for 6 hours. Based on the molar ratio of Al:Y in the precursor, we labelled the corresponding samples as 0A10Y, 2A8Y, 2.5A7.5Y, 3A7Y, 3.8A6.2Y, 4A6Y, 6A4Y and 10A0Y. The weight percentages of each component in the precursor mixture are shown in table 2.

Table 2. *Symbols and compositions of raw materials for the fabrication of SiAlON ceramics.*

No.	**Sample**	**Composition,** wt%			**Ratio** Al_2O_3*100/ (Al_2O_3+Y_2O_3), %
		Si_3N_4	Al_2O_3	Y_2O_3	
1	0A10Y	90.00	0.00	10.00	0.00
2	2A8Y	90.00	1.88	8.12	18.70
3	2.5A7.5Y	90.00	2.54	7.46	25.37
4	3A7Y	90.00	2.92	7.08	29.08
5	3.8A6.2Y	90.00	3.79	6.21	37.89
6	4A6Y	90.00	4.38	5.62	43.82
7	6A4Y	90.00	6.29	3.71	62.96
8	10A0Y	90.00	10.00	0.00	100.00

After mixing step, the obtained mixtures were shaped to pellet using a cylindrical mold with a diameter of Ø20 x 20 (mm), pressed by a hydraulic press (Germany) with a cylinder shaft diameter

of 115mm with a pressure of 7000 kgf/cm^2. Then the samples were dried at 70 ºC for 4 hours and 110 ºC for 4 hours to completely remove moisture.

The samples were sintered in a furnace at: 1450 ºC, 1500 ºC, 1550 ºC, 1600 ºC and 1650 ºC for 4 hours in N_2 atmosphere while the pressure was maintained at 1.0MPa.

2.3. Materials characterization

XRD analysis (Bruker, Germany) of the solid was performed at 2-theta from 10º to 80º. SEM images were captured using HITACHI S-4800, Japan, with a voltage of 5 kV. The differential thermal analysis (DTA) (Linseis thermal analyser, Germany) was measured in the Al_2O_3/Y_2O_3 ratio temperature range from 30 °C to 1650 ºC with a ramping rate of 5 ºC/min, under N_2 atmosphere with a gas flow rate of 83 mL/min. Physical parameters, including bulk density, porosity, and water absorption, were measured by Archimedes' method. fracture toughness, K1C was determined by JIS R1607-2010 Standard (E = 300GPa) and Liihara formula:

$$K_c = 0.067\left(\frac{E}{H_v}\right)^{0.4} H_v a^{0.5}\left(\frac{c'}{a}\right)^{-1.5}, \qquad c'/a > 3.0$$

Where:

E is the elastic modulus, Gpa;
Hv is the hardness Vickers, Gpa;
P is the compressive pressure, N.
c' is 1/2 the crack length, mm;
a is 1/2 the size of the indenter, mm;

3. RESULTS AND DISCUSSION

3.1. Effect of temperature on phase formation in the synthesis of SiAlON ceramics

XRD is a powerful technique to determine the structure of powder samples. In this study, we investigated the influence of sintering temperature on the synthesis of SiAlON by XRD analysis of sintered samples. Figure 1 shows the XRD patterns of 3A7Y samples sintered range from 1450 ºC to 1650 ºC.

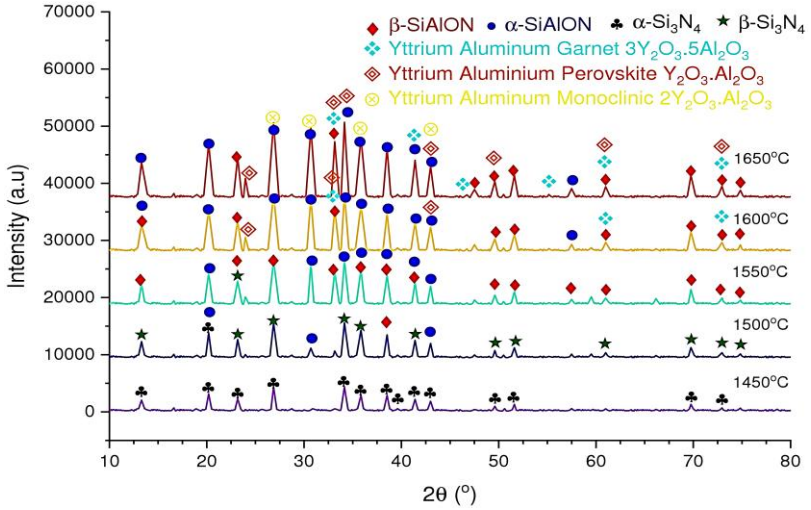


Figure 1. *XRD patterns of 3A7Y sample when sintered at temperatures: 1450 ºC, 1500 ºC, 1550 ºC, 1600 ºC, 1650 ºC.*

The XRD patterns results from figure 1 show that when the temperature increases from 1450 ºC to 1500 ºC, the 3A7Y sample has a transformation from α-Si_3N_4 to β-Si_3N_4 phase.

At 1500 ºC, the α-SiAlON crystal phase appears at the peaks 2Θ of 20.2º, 30.7º and 43.0º. The β-SiAlON phase also appears at the peaks 2Θ = 38.5º and the Aluminum Nitride AlN phase also

appears at the peaks 2Θ of 33.2° and 38.5°. When the temperature was increased to 1550 °C, the peaks of the α-SiAlON, β-SiAlON and Aluminum Nitride AlN phases increased completely, while the α-Si_3N_4 phase disappeared completely, and the β-Si_3N_4 phase remains only at the peak 2Θ = 23.2°. When the temperature was increased to 1600 °C, the peaks of the β-SiAlON phase decrease due to the transformation into the α-SiAlON phase, the peaks of the β-Si_3N_4 and Aluminum Nitride AlN phases disappeared completely. Yttrium Aluminum Perovskite $Y_2O_3.Al_2O_3$ phase, appears at the peak 2Θ of 24.0°; 33.2° and 43.0°. The Yttrium Aluminum Garnet $3Y_2O_3.Al_2O_3$ phase appears at the peak 2Θ of 33.2°; 61.0° and 72.9°.

When the temperature was increased to 1650 °C, the α-SiAlON phase developed completely, the β-SiAlON phase maintained a stable structure, the Yttrium Aluminum Perovskite $Y_2O_3.Al_2O_3$ phase appeared with new peaks at 2Θ of 34.1°; 49.6°; 61.0° and 72.9°. The Yttrium Aluminum Monoclinic $2Y_2O_3.Al_2O_3$ phase appeared at the peaks 2Θ of 26.9°; 30.7°; 35.8° and 43.0°. The Yttrium Aluminum Garnet $3Y_2O_3.Al_2O_3$ phase appeared at the peaks 2Θ of 41.4°; 46.7° and 55.2°.

After sintering at 1650 °C, the obtained powder showed several characteristic peaks of both α and β-SiAlON: at 2Θ of 13.3°, 20.2°, 26.9°, 30.7°, 34.41°, 35.8°, 38.5°, 41.4°, 43.0°, 57.5° and at 2Θ of 23.2°, 33.2°, 47.5°, 49.6°, 51.6°, 61.0°, 69.8°, 72.9°, 74.8°, respectively. The phase composition (%wt) involved 26% of α-SiAlON, 55% of β-SiAlON, 5% of $3Y_2O_3.Al_2O_3$, 3% of $Y_2O_3.Al_2O_3$, 1% of $2Y_2O_3.Al_2O_3$ and 11% of the glass component.

3.2. Effect of temperature on the bulk density of SiAlON ceramics

SiAlON ceramics samples were prepared from Si_3N_4 powder with the composition ratios, preparation and sintering process presented above. The research results for SiAlON ceramics when fired at various Al_2O_3/Y_2O_3 ratios and temperature for 21 hours in N_2 environment (sintering is 4 hours) at 1.0MPa pressure are shown in figure 2.

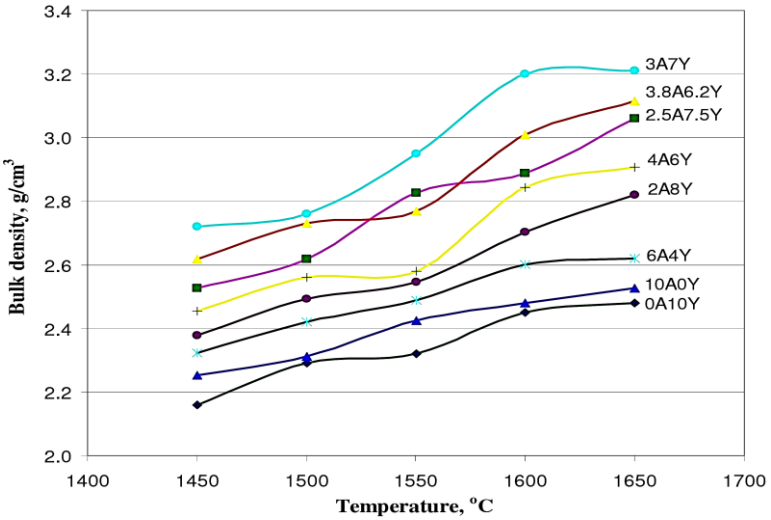


Figure 2. *Effect of temperature on bulk density of SiAlON ceramics.*

From figure 2, it can be seen that the bulk density of 2A8Y sample was increased from 2.38 g/cm^3 to 2.82 g/cm^3; The bulk density of 2.5A7.5Y sample was increased from 2.53 g/cm^3 to 3.06 g/cm^3; The bulk density of 3.8A6.2Y sample was increased from 2.62 g/cm^3 to 3.12 g/cm^3; The bulk density of 4A6Y sample was increased from 2.45 g/cm^3 to 2.91 g/cm^3; The bulk density of 6A4Y sample was increased from 2.32 g/cm^3 to 2.62 g/cm^3; The bulk density of 10A0Y sample was increased from 2.25 g/cm^3 to 2.53 g/cm^3. The bulk density of 0A10Y sample was increased the least, from 2.16 g/cm^3 to 2.48 g/cm^3 and the bulk density of 3A7Y sample was increased the most, from 2.72 g/cm^3 to 3.21 g/cm^3 when the temperature was increased from 1450 °C to 1650 °C. The bulk density of 3A7Y sample was the largest, while the 0A10Y sample was the smallest. The

bulk density of the samples did not increase much when the temperature was increased from 1600 ºC to 1650 ºC, proving that the appropriate calcination temperature from 1600 ºC to 1650 ºC and the suitable sample was 3A7Y.

When the temperature was increased so the liquified Al_2O_3/Y_2O_3 mixture could reduce the surface energy, promote the agglomeration, help the β-SiAlON particles to grow in rod shape, thus filling the voids. In addition, the increase in Al_2O_3 content promoted the formation of AlN and SiAlON crystals, which have denser crystal structures; Thus, the density was increased.

3.3. Effect of temperature on the porosity of SiAlON ceramics

From table 3, the effect of temperature on the porosity of the samples after sintering is shown in figure 3.

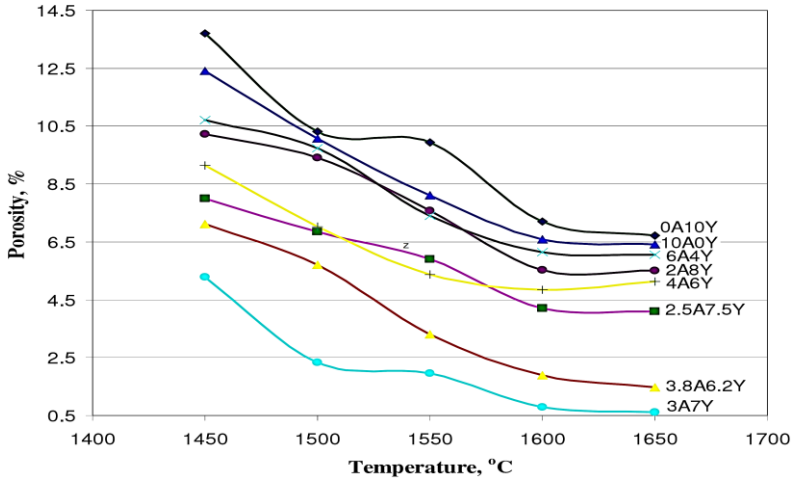


Figure 3. *Effect of temperature on porosity of SiAlON ceramics.*

Also from figure 3, it can be seen that the porosity of 2A8Y sample was decreased from 4.42% to 2.08%; The porosity of 2.5A7.5Y sample was decreased from 3.33% to 1.44%; The porosity of 3.8A6.2Y sample was decreased from 2.86% to 0.48%; The porosity of 4A6Y sample was decreased from 3.86% to 1.91%; The porosity of 6A4Y sample was decreased from 4.70% to 2.31%; The porosity of 10A0Y sample was decreased from 5.51% to 2.54%. The porosity of sample 0A10Y was decreased the most from 6.34% to 2.71% and the porosity of 3A7Y sample was decreased the least from 1.94% to 0.20% when the temperature was increased from 1450 ºC to 1650 ºC. The porosity of sample 3A7Y was the smallest, while that of sample 0A10Y was the largest. The samples had a small decrease in porosity when the temperature was increased from 1600 ºC to 1650 ºC. This is consistent with the law of change in the bulk density of the sample.

In contrast, the temperature showed a reversed influence on water absorption and porosity of the sample. These observations could be explained in a similar way that the development of denser β-SiAlON facilitated a tight structure, reducing porosity.

3.4. Effect of temperature on fracture toughness, K_{1C} of SiAlON ceramics

The fracture toughness, K_{1C}, of the samples was examined as shown in figure 4.

From figure 4, it can be seen that the fracture toughness K_{1C} of 3A7Y sample has the largest (5.80 $MPa.m^{1/2}$), the fracture toughness K_{1C} of 0A10Y sample is the smallest. As the density increased, the fracture toughness K_{1C} also increased. This is consistent with the law of change in bulk density, porosity and water absorption of the samples.

High density and uniform fine microstructure (many rod-shaped β-SiAlON particles) enhance the crack bridging mechanism (crack bridging, crack deflection), thereby improving the fracture toughness K_{1C}.

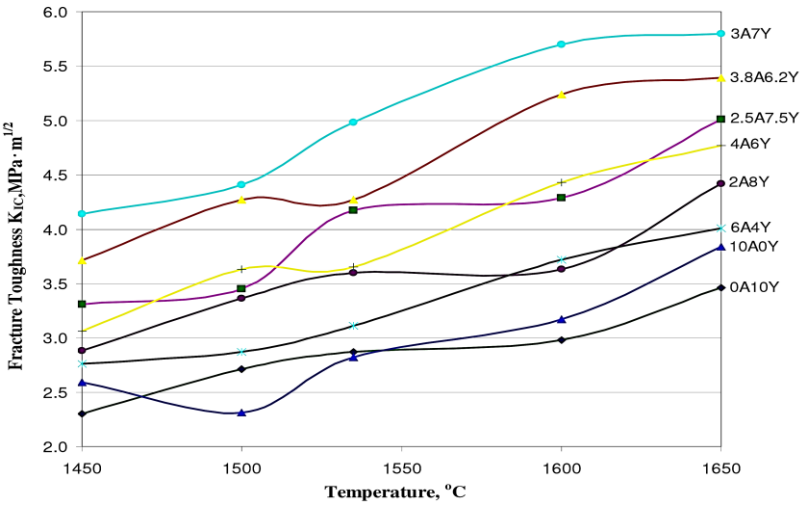


Figure 4. *Effect of temperature on fracture toughness, K1C of SiAlON ceramics.*

4. CONCLUSIONS

The SiAlON ceramics were successfully synthesized from Si_3N_4 powders with the aid of Al_2O_3/Y_2O_3 mixture, confirmed by XRD. The influence of sintering temperature on the properties of the sample was examined.

When the temperature was increased, the liquified Al_2O_3/Y_2O_3 mixture could reduce the surface energy, help the β-SiAlON particles to fill the voids, thus, the density was increased.

By changing the Al_2O_3/Y_2O_3 ratio and sintering temperature, a high density of 3.21 g/cm^3, low water absorption of 0.20%, low porosity of 0.61% and exceptional fracture toughness K_{1C} of 18.00 GPa were achieved at the Al_2O_3/Y_2O_3 ratio of 3:7, under 4 hours of sintering at 1650 ºC in N_2 atmosphere.

REFERENCES

[1]. Y. K. Kshetri et al., *"Electronic structure, thermodynamic stability and high-Al_2O_3/Y_2O_3 ratio sensing properties of Er-α-SiAlON ceramics"*, Sci. Rep., 10, pp. 1–13, (2020), doi: 10.1038/s41598-020-61105-z.

[2]. K. A. Kim, A. S. Lysenkov, M. G. Frolova, and Y. F. Kargin, *"Effect of calcium aluminates content on the formation of Ca-α-SiAlON ceramics obtained by hot-pressing"*, Ceramics International, 50, pp. 47886–47891, (2024), doi: 10.1016/j.ceramint.2024.09.134.

A. A. M. El-Amir et al., *"SiAlON from synthesis to applications: an overview"*, Journal of the Asian Ceramic Societies, 9, pp. 1390–1418, (2021), doi: 10.1080/21870764.2021.1987613.

[3]. J. Zhou et al., *"The effects of in-situ SiAlON on the properties and fracture behavior of alumina-based castables: Based on microcrack toughening mechanism"*, Ceramics International, 51, pp. 4549–4559, (2024), doi: 10.1016/j.ceramint.2024.11.429.

[4]. M. Estili, R.-J. Xie, K. Takahashi, S. Funahashi, T. S. Suzuki, and N. Hirosaki, *"Robust and orange-yellow-emitting Sr-rich polytypoid α-SiAlON ($Sr_3Si_{24}Al_6N_{40}:Eu^{2+}$) phosphor for white LEDs"*, Science and Technology of Advanced Materials, 25, pp. –, (2024), doi: 10.1080/14686996.2024.2396276.

[5]. Y. Zhang et al., *"The synthesis of single-phase β-SiAlON porous ceramics using self-propagating high-Al_2O_3/Y_2O_3 ratio processing"*, Ceramics International, 48, pp. 4371–4375, (2022), doi: 10.1016/j.ceramint.2021.10.188.

[6]. M. Z. Falak et al., *"Spark plasma sintering of SiAlON ceramics synthesized via various cations charge stabilizers and their effect on thermal and mechanical characteristics"*, Crystals, 11, (2021), doi: 10.3390/cryst11111378.

[7]. S. Zhang et al., *"Thermal conductivity of Ca-α-SiAlON ceramics with varying m and n values"*, Journal of the American Ceramic Society, 106, pp. 5642–5647, (2023).

[8]. B. Chaudhary et al., *"Up- and down-conversion photoluminescence in Nd-doped SiAlON ceramics"*, Ceramics International, (2025), doi: 10.1016/j.ceramint.2025.01.349.

[9]. Q. Liu, Z. Yin, F. Guo, and J. Yuan, *"Effects of binary sintering additives (SmF_3–Sm_2O_3) and sintering Al_2O_3/Y_2O_3 ratio on β-SiAlON ceramic tool materials"*, Ceramics International, 50, pp. 51456–51464, (2024), doi: 10.1016/j.ceramint.2024.10.062.
[10]. X. Tian et al., *"Fabrication and oxidation behavior of β-SiAlON powders in presence of trace Y_2O_3"*, Ceramics International, 48, pp. 32464–32469, (2022), doi: 10.1016/j.ceramint.2022.07.192.
[11]. D. Bruce et al., *"A critical assessment of the Archimedes density method for thin-wall specimens in laser powder bed fusion: Measurement capability, process sensitivity and property correlation"*, Journal of Manufacturing Processes, 79, pp. 185–192, (2022), doi: 10.1016/j.jmapro.2022.04.059.
[12]. D. Liu et al., *"Densification, microstructure and properties of α/β-SiAlON ceramic reinforced by SiC whiskers"*, Ceramics International, 50, pp. 42755–42765, (2024), doi: 10.1016/j.ceramint.2024.08.121.
[13]. X. Li et al., *"Preparing β-SiAlON ceramic foam filters with high oxidation resistance"*, Ceramics International, 49, pp. 34510–34519, (2023), doi: 10.1016/j.ceramint.2023.08.075.
[14]. Z. Tu et al., *"Effect of Si/Al ratio on in-situ synthesis of Al_2O_3–β-SiAlON composite ceramics for solar thermal storage by aluminothermic and silicothermic nitridation"*, Ceramics International, 49, pp. 22970–22978, (2023), doi: 10.1016/j.ceramint.2023.04.122.

TÓM TẮT

Ảnh hưởng của nhiệt độ đến thành phần pha, mật độ và độ bền đứt gãy K_{1C} của gốm tiên tiến SiAlON

Trong nghiên cứu này, ảnh hưởng của nhiệt độ đến sự hình thành pha, mật độ và độ bền đứt gãy (hệ số K_{1C}) của gốm tiên tiến SiAlON đã được khảo sát. Điều kiện tổng hợp được tối ưu hóa ở 1650 °C trong 4 giờ trong môi trường N_2 ở áp suất 1,0 MPa. Nhiệt độ có ảnh hưởng đến thành phần pha, mật độ và hệ số K_{1C} của gốm tiên tiến SiAlON. Kết quả tổng hợp tối ưu của gốm tiên tiến SiAlON cho thấy: mật độ cao 3,21 g/cm^3; độ hút nước thấp 0,20%; độ xốp 0,61% và hệ số K_{1C} 5,80 MPa.m$^{1/2}$.

Từ khoá: Tỷ lệ Al_2O_3/Y_2O_3; Nhiệt độ; Gốm sứ tiên tiến SiAlON.