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Effects of process on microstructure and properties of translucent Dy-α-SiAlON sintered at lower temperatures

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Hot pressing (HP) at higher sintering temperature has been a traditional and prevalent technique for the fabrication of α-SiAlON. In order to prepare translucent SiAlON more easily, LiF was used as a non-oxide sintering additive to lower the sintering temperature to ≤ 1650°C. As a result, all of the samples possessed a good hardness and fracture toughness. At the same time, the lower temperature sintered samples showed a higher optical transmittance in the range of 2.5–5.5 μm wavelength (0.5 mm in thickness). The maximum infrared transmission reached 68% at a wavelength of 3.3 μm. The present work shows that the sintering process has a strong effect on microstructure and property of α-SiAlON. To be exact, a lower sintering temperature and longer holding time can produce some fully-developed microstrcture, which is beneficial for the optical transmittance.

Key-words : SiAlON, Hot-pressing, Densification, Optical transmittance

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1. Introduction

Hot-pressing (HP) of α-SiAlON has been a traditional and prevalent technique for the fabrication since the first work on it.[1),2)] Using this method, large numbers of SiAlON ceramics have been successfully fabricated.[3)–6)] As one difficult issue, the full densification of the green body could only be achieved at very high sintering temperatures (usually > 1700°C, mostly 1800°C), which should not be ignored. Therefore, it would be of great signification for sintering SiAlON at lower temperatures by HP.

The reason for the difficult sintering of SiAlON materials is caused by the covalent nature of Si–N bonding. Besides more extra energy supplied by high pressure and high temperature with HP, adding oxide sintering additive, especially rare earth oxides Ln_2O_3 (Ln = Nd, Sm, Gd, Dy, Y, Er and Yb),[7),8)] has been taken as another effective way to promote α-SiAlON densification in the past years. But, the sintering temperature still maintained very high for the Si_3N_4-solide solution system materials. Therefore, seeking new densification additives to achieve lower sintering temperatures (< 1700°C) should be a meaningful effort on SiAlON preparation.

In our previous work,[9)] LiF as an non-oxide additive has been used to facilitate the densification of SiAlON ceramics. As a result, α-SiAlON starting compositions have been successfully densified at lower temperatures (≤ 1650°C) by hot-pressing. A translucent sample was obtained at a certain low temperature of 1600°C, with a max transmittance of 63% (0.5 mm in thickness).

In the present work, different manufacturing processes have been further applied to study the relationship between the microstructure and the optical transmittance. To be exact, the lower sintering temperature and longer holding time can produce some fully developed grains, which is beneficial for increasing optical transmittance.

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2. Experimental

According to the formula of $M_{m/v}Si_{12-(m+n)}Al_{m+n}O_nN_{16-n}$, the α-SiAlON ceramic reported here has a nominal composition $Dy_{0.66}Si_{9.0}Al_{3.0}O_{1.0}N_{15.0}$ (M = Dy, $m = 2n = 2.0$, $v = 3$). High-purity powders of α-Si_3N_4 (SN–E10, Ube, Japan), AlN (Grade A, Starck, Germany), Dy_2O_3 (Yaolong Chemical Plant, Shanghai, China), and 0.1 mass% LiF (Sinopharm Chemical Reagent Co., Ltd., Beijing, China) were mixed in ethanol, milled using Si_3N_4 balls, dried at 80°C, sieved, and then a charge of 6.0 g each was loaded directly into a graphite die. The samples were hot pressed at 1600–1650°C for 60–90 min under a flowing nitrogen atmosphere of 1 atm. The input and output facets of the sintered samples were polished for mechanical and optical measurements.

Bulk density of specimens was measured by Archimedes principle. The Vickers hardness as well as indentation fracture toughness were determined at room temperature using a Vickers diamond indenter at a 10 kg load for 10 s. Crystalline phases were determined by X-ray diffraction(XRD, D/max 2550V, Rigaku) using Cu Kα radiation. Microstructure observation was performed under a transmission electron microscope (TEM; JEOL, Tokyo, Japan; JEM–2010/200CX/2100F). Optical transmissions of the translucent sample in wavelength of 2.5–6.5 μm were measured by FTIR (EQUINOX55, Bruker, Billerica, MA).

3. Results and discussion

3.1 Bulk density, transmittance and mechanical property of the sintered samples

Table 1 shows the effects of sintering temperature and soaking time on the densification of the mixed powders and the properties of the bulks as well. The bulk density values of the sintered sam-

Table 1. Bulk Density, Main Phase, Transmittance and Mechanical Property of the Sintered Samples

Sample	Temperature (°C)	Soaking time (min)	Bulk density ($g \cdot cm^{-3}$)	Main phase*	Transmittance** (%)	Mechanical property	
						HV10 (GPa)	K_{1C} ($MPa \cdot m^{1/2}$)
16060	1600	60	3.62	α'	63	20.0	3.8
16260	1620	60	3.64	α'	57	19.8	4.7
16560	1650	60	3.66	α'	61	19.7	5.0
16090	1600	90	3.63	α'	68	20.0	3.5
16290	1620	90	3.64	α'	67	19.8	4.8
16590	1650	90	3.65	α'	65	19.7	5.2

*α' = Dy-α-SiAlON phase; **0.5 mm thickness.

ples are 3.62–3.66 $g \cdot cm^{-3}$, showing a better densification result. All samples possess the excellent mechanical properties, in which the hardness values are > 19.0 GPa and the fracture toughness are about 3.8–5.2 $MPa \cdot m^{1/2}$. The optical transmittance of most samples is higher than 60%. All the results mentioned above are consistent with those publications previously reported, using oxides additive, but at higher sintering temperatures > 1700°C.

For the samples sintered at varied temperatures but for the same soaking time (60 or 90 min), the bulk density value of the sample was improved when the sintering temperature increased from 1600°C to 1650°C. At the same time, the increasing sintering temperature had an influence on mechanical properties, where the hardness value showed a decline while the fracture toughness value was improved greatly.

It is clearly that, the optical transmittance of the sample sintered for 90 min are all better than those sintered for 60 min. Therefore, a longer soaking time results in a higher optical transmittance. But for the same soaking time, the lower sintering temperature leads to a better transmittance, which can be confirmed by the samples sintered at 1600°C.

3.2 Relationship between the microstructure and optical transmittance

Figure 1 shows the optical transmittance curves in the range of 2.5–5.5 μm of the samples manufactured by different process. The cutoff wavelength of the samples is located at about 5.0 μm. It is also noted that one absorb peak appears at 2.8 μm. The electron transformation from rear earth ion Dy^{3+} might be a possible

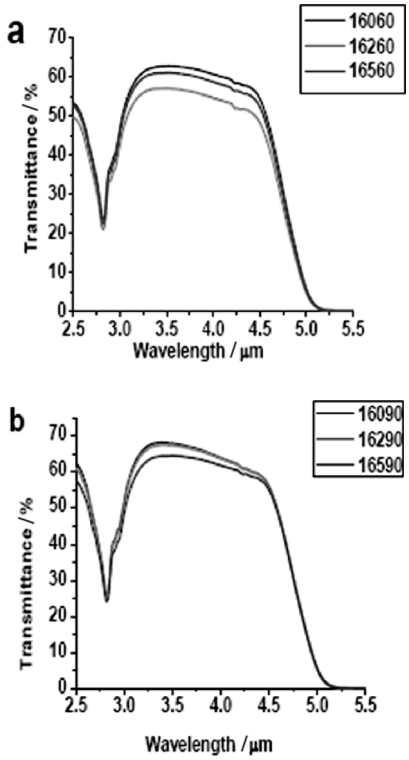

Fig. 1. Optical transmittance curves of Dy-α-SiAlON with 0.1 mass% LiF addition (0.5 mm in thickness) in the range of 2.5–5.5 μm (a) soaking for 60 min and (b) soaking for 90 min.

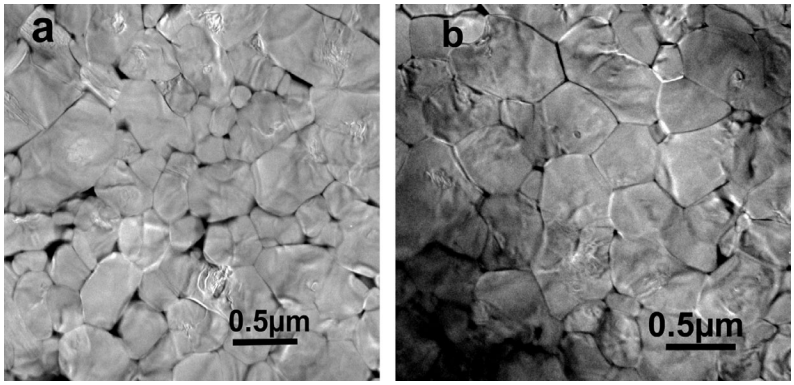

Fig. 2. TEM morphologies of samples (a) sample 16260, (b) sample 16290.

explanation for the absorptions, but the mechanism has not been very clear yet.

It is useful to find out the reason for the difference of the transmittance of the samples. So, TEM observation on the sample 16260 and 16290 in detail were carried out (seen in **Fig. 2**). No Pores can be observed obviously in both samples. And it is clear that Dy-α-SiAlON phase has been formed in both samples (see Table 1). Most of grain size is less than 1.0 μm. The grain morphology in sample 16290 is more uniform than those in sample 16260. Moreover, the crystal boundary in sample 16290 seems neater than that in sample 16260. It shows that a longer soaking time can promote the grain development, resulting in more uniform morphology with less crystal boundary. It is just the reason why the transmittance of the samples sintered for 90 min are better than those sintered for 60 min.

4. Summary

The Translucent α-SiAlON samples can be manufactured by the controllable temperatures and soaking time condition. Almost all the samples possess a maximum transmittance over 60%. At a same sintering temperature, a longer soaking time produces a better transmittance. For the same soaking time, the lower sintering temperature results in a better transmittance. The sample 16090 sintered at 1600°C for 90 min has the highest transmittance of 68%. The microstructure has a significant influence on the transmittance, where the fully developed grains with uniform grain size, morphology and less crystal boundary lead to a better transmittance.

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