Translucent $\alpha$-SiAlON Ceramics Produced by Gas Pressure Sintering (GPS) Method

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Abstract

In the present work, translucent Dy$^{3+}$-doped $\alpha$-SiAlON ceramics were successfully fabricated by gas pressure sintering (GPS) method and their microstructure were tailored by controlling sintering parameters to improve transmittance of products. The transmittance of the sintered Dy-$\alpha$-SiAlON ceramics were inspected in 2000–7000 cm$^{-1}$ wave number region by using FTIR. The microstructure and phase characterization of the samples were also carried out by using SEM and XRD techniques, respectively. Results showed that the main parameters determining the optical features of translucent SiAlON ceramics are grain size/grain size distribution in the sintered product, phase variety therein, the amount and distribution of grain boundary phases within the structure and density.

1. Introduction

SiAlON is a kind of ceramic alloy which is produced by combination of four elements (silicon (Si), aluminium (Al), oxygen (O) and nitrogen (N)). As a result of substitution for Si by Al with corresponding atomic replacement of N by O, SiAlON has superior properties than Si$_3$N$_4$ such as high strength (even at high temperatures), good thermal shock resistance and exceptional resistance to corrosion. Due to this reason SiAlON ceramics have been widely used for extremely harsh structural applications. In recent years, serious attention were taken to examine SiAlON ceramics optical properties as well. The main aim of these researchs to achieve better optical transmission without losing superior mechanical properties of SiAlIONs. Many reasearchers worked with different $\alpha$-SiAlON compositions and dopands such as alkali earth cations (Ca$^{2+}$ and Mg$^{2+}$) or rare earths (Lu$^{3+}$, Nd$^{3+}$, Sm$^{3+}$, Tb$^{3+}$) to improve optical properties of $\alpha$-SiAlIONs [1-4]. Additionally, in many researches, the transparency of the SiAlIONS has been tried to improve by using different sintering techniques such as spark plasma sintering (SPS), hot isostatic pressing (HIP) and hot press (HP) [5-7]. However, there is no report about production of translucent SiAlONs by the gas pressure sintering (GPS) method.

The aim of this study is to fabricate translucent Dy-$\alpha$-SiAlON ceramics by using GPS technique. Additionaly to investigate effects of GPS parameters on optical properties of products.

2. Experimental Procedure

High-purity powders of Si$_3$N$_4$ (UBE-10, containing 1.6 wt.% oxygen), AlN (Tokuyama, containing 1 wt.% oxygen), Al$_2$O$_3$ (99.99 wt.%, Sumitomo AES IIC) and Dy$_2$O$_3$ (99.99%, HC Starck) were used to prepare Dy$_{0.66}$Si$_{9.0}$Al$_{3.0}$O$_{1.0}$N$_{15.0}$ composition according to general formula of $\alpha$-SiAlION ($\text{Re}^{2+}$Si$_{12-m-n}$Al$_{m+n}$O$_n$N$_{16-n}$). The weighed starting powders were milled by planetary ball milling (Pulverisette 6 Fritsch, Germany) by using Si$_3$N$_4$ balls and a Si$_3$N$_4$ jar in order to obtain a homogeneous mixture. Ethanol used as solvent for milling process. After milling, the powder slurry was dried at 55°C and sieved (250 μm). The dry powders were pressed into tablet-shaped compacts by Uniaxial Pressing Equipment with 2 MPa pressure. Pellets of each specimen were cold isostatic pressed to achieve maximum green density. Shaped samples were sintered by GPS method using FCT GmbH-Germany branded a gas pressure sintering furnace. Sintering conditions of samples shown in Table 1. Also the entire process is performed in N$_2$ atmosphere. Infrared transmissions in wave number of 1000–7000 cm$^{-1}$ were measured by FT-IR (Bruker Tensor27). The densities of the sintered samples were measured by the Archimedes method in distilled water. The phase compositions were analyzed using the X-ray diffraction method (Rigaku Rint 2200) with Cu Kα radiation ($\lambda = 1.540$ Å) from 20° to 60° and a scanning speed of 1/min. The microstructure of the final products were characterized by scanning electron microscopy (Zeiss Supra 50 V).
### 3. Results and Discussion

The bulk densities and the porosity amount of the sintered specimens are listed in Table 2. According to density measurements, GDy2 specimens have a higher density value and lower porosity content than GDy1 specimen. GDy2 sample could densified to over 99% of its theoretical density however GDy1 sample includes high amount of porosity which is around %16. This difference is obviously related to their sintering conditions. GDy2 specimen sintered at higher temperature and pressure than GDy1 which can be seen from Table 1.

#### Table 1. Sintering conditions of sintered samples

<table>
<thead>
<tr>
<th>Sample Code</th>
<th>Sintering Temperature (°C)</th>
<th>Holding Time (h)</th>
<th>Pressure (bar)</th>
</tr>
</thead>
<tbody>
<tr>
<td>GDy1</td>
<td>1850</td>
<td>2</td>
<td>22</td>
</tr>
<tr>
<td>GDy2</td>
<td>1900</td>
<td>2</td>
<td>70</td>
</tr>
</tbody>
</table>

#### Table 2. Bulk density and porosity values of samples

<table>
<thead>
<tr>
<th>Sample Code</th>
<th>Density (g/cm³)</th>
<th>Porosity (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>GDy1</td>
<td>2.89</td>
<td>16.38</td>
</tr>
<tr>
<td>GDy2</td>
<td>3.23</td>
<td>1.01</td>
</tr>
</tbody>
</table>

SEM micrographs of the GPS-ed GDy1 and GDy2 samples are shown in Figure 1 and 2 respectively. SEM micrographs confirm the density measurement results. GDy1 specimen includes high amount of porosity which are shown as darker areas on micrograph and one of them marked as point 1 (Figure 1). On the other hand, any amount of porosity hardly seen from GDy2 samples micrograph. The samples consist of equiaxed and elongated α-SiAlON grains. The average grain size of equiaxed α-SiAlON grains very small, on the contrary average elongated α-SiAlON grains size approximately 30 μm. Some dark grains were observed in the GDy2 specimen. This secondary crystalline phase might be AlN-polytype. AlN-polytypes are commonly seen in the α-SiAlON structure after sintering. Due to its low amount of existence, AlN-polytypes couldn’t detect by XRD analysis.

The XRD analyses of the sintered samples were carried out to check the phase composition of the samples. X-ray diffraction pattern of the GPSed GDy1 and GDy2 samples are shown in Figure 3. The results show that α-SiAlON is
the major phase in both of the samples. No other crystalline phase or amorphous-glassy phase were not observed.

![X-ray diffraction pattern](image)

**Figure 3.** X-ray diffraction pattern of the samples, red pattern $GDy1$ and blue pattern $GDy2$.

The infrared transmittance of the specimens, ranging between 2000 and 7000 cm$^{-1}$, are shown in **Figure 4**.

![Infrared transmission](image)

**Figure 4.** Infrared transmission of SPS-ed specimens (a) $GDy1$-red (b) $GDy2$-blue (0.3 mm in thickness)

Maximum transmission values and phase contents of the specimens are shown in **Table 3** also. $GDy2$ samples highest transmission value is 21.489% at around ~2100 wavenumber for 3 mm sample thickness. However, $GDy1$ sample didn’t shown any transmission in range of 2000-7000 cm$^{-1}$. It is known that the microstructural inhomogeneity such as porosity, grain boundary, anisotropy of grains cause the internal light scattering. Due to this reason material behave like optical opaque. Because of high amount of porosity that $GDy1$ sample includes, $GDy1$ sample became optical opaque in contrast with $GDy2$ sample is translucent.

**Table 3.** Transmission values and phase contents

<table>
<thead>
<tr>
<th>Sample Code</th>
<th>Maximum Transmission (%)</th>
<th>Phase Content</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\alpha$-SiAlON</td>
<td>AIN-polytype</td>
</tr>
<tr>
<td>$GDy1$</td>
<td>-</td>
<td>vs</td>
</tr>
<tr>
<td>$GDy2$</td>
<td>21.489</td>
<td>vs</td>
</tr>
</tbody>
</table>


4. Conclusion

Dy-doped $\alpha$-SiAlON ceramics produced by gas pressure sintering method at different sintering conditions (temperature and pressure). $GDy2$ sample could fully densified at 1900°C and 70 bar pressure. However, $GDy1$ sample (which has same composition with $GDy2$) could not fully densified at 1850°C and 22 bar pressure. The results show that $GDy1$ specimen is optically opaque however $GDy2$ specimen is translucent. The main reason of the difference between samples optical properties is density distinctness.

As a conclusion, it is possible to produce translucent $\alpha$-SiAlON ceramics by gas pressure sintering technique. However, microstructure tailoring is must to obtain highly translucent $\alpha$-SiAlON ceramics.

References
[7] Qian Liu, Wei He, Hongmei Zhong, Kun Zhang, Linhua Gui. Transmittance improvement of Dy-$\alpha$-SiAlON in infrared range by post hot-isostatic-pressing. SiAlONs and Non-Oxide Ceramics, 2012; 32:1377-1381