chemistry and structure

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Combined analysis of textured alumina ceramics by neutron diffraction

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The orientation distributions of α-Al_2O_3 textured ceramics are determined from neutron diffraction spectra. A curved position-sensitive detector coupled to a tilt angle (χ) scan allowed treatment of the whole diffraction pattern in a Rietveld-WIMV-Popa algorithm. Four textured alumina ceramics were prepared by slip-casting under a high magnetic field and sintered at 800°C, 1300°C, 1400°C and 1600°C. With increasing sintering temperature, the texture strength is enhanced and the c-axis distribution is sharper.
The effectiveness of this approach for determining crystallite size is also evident. As a global trend, the calculated crystallite size and observed grain size are similar and increase with increasing sintering temperature. We see here the power of Rietveld texture analysis to provide a basis for the correlation of texture, microstructural parameters and anisotropic properties.

Texture analysis is being increasingly recognised as an important tool in the characterisation of many polycrystalline materials in order to understand their anisotropic properties. The development of texture in α-Al_2O_3 ceramics is required to improve thermal conductivity, optical, and mechanical properties [1, 2]. The particular type of texture often found in α-Al_2O_3 textured materials is an axially symmetric fiber texture (*i.e.*, random in-plane distribution of crystallite a and b axes). This type of texture can be best characterised by the c-axis orientation distribution, but in the case of slightly oriented materials, this is difficult to measure using X-rays because of the low structure factors of all the (00*l*) reflections. To overcome this problem, the solution is to calculate the orientation distribution of the crystallites from several other (*hkl*) distributions, *i.e.*, to acquire as many pole figures as possible for the non-parallel (*hkl*) crystallographic planes. In this paper, we used an approach that combines classical Rietveld, analysis with texture and crystallite size analysis, as implemented in the MAUD software [3]. We illustrate the advantages to analyse jointly structure, texture and microstructure of α-Al_2O_3 ceramics. The mechanism of texture development during the sintering treatment is discussed.

The materials were processed using the slip casting method [4]. A high magnetic field of 10T, parallel to the slip casting direction, was applied to a suspension of fine α-Al_2O_3 spherical particles (∅~150 nm) at room temperature. Due to the anisotropic magnetic susceptibility of the hexagonal structure, a high magnetic field applied during the slip casting is highly effective in rotating the particles and generating a (001) preferred orientation parallel to the magnetic field. After slip casting, four samples were prepared by sintering at 800°C, 1300°C, 1400°C and 1600°C. The specimens were measured using D1B. Diffracted neutrons are collected by a curved position sensitive detector composed of 400 cells spread over 80° (resolution 0.2°) in 2θ. An Eulerian cradle allows χ angle rotation. Scans were performed from χ = 0 to 90° (step 5°) using a fixed incidence angle ω of 35.6° ({006} Bragg position).

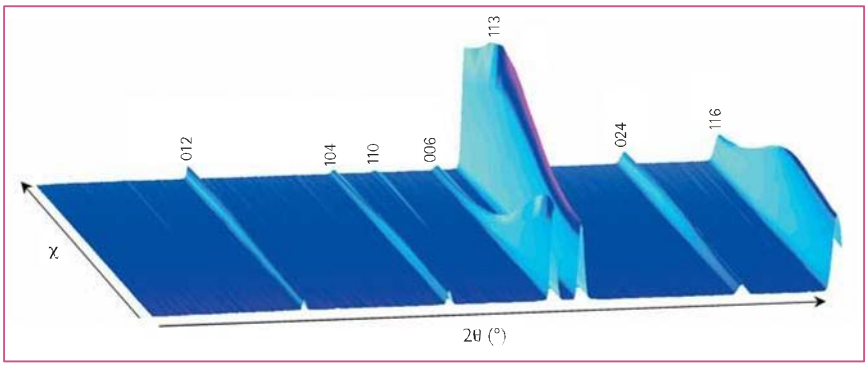

Figure 1: Neutron diffraction patterns for a 0° to 90° χ-scan (sample sintered at 1300°C).

Figure 1 shows a typical neutron diffraction pattern obtained for our samples using a 0 to 90° χ-scan (sample sintered at 1300°C). This graph highlights without ambiguity the (00l) texture of the α-alumina specimen. In particular, we can clearly observe the intensity decrease of the (006)

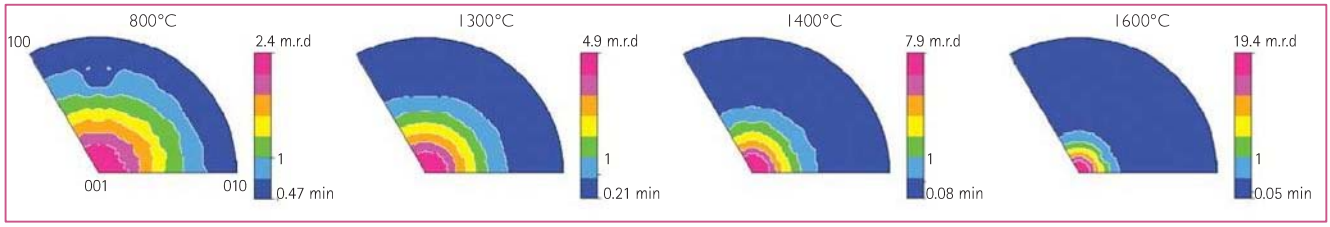

Figure 2: Inverse pole figures calculated for the z fibre direction (parallel to the magnetic field) for samples sintered, respectively, at 800°C, 1300°C, 1400°C and 1600°C. Major (001) component. Linear density scale, equal area projection.

line when χ increases and the appearance of the (110) line when χ tends to 90°. The refinement reliability is established by R_p, R_{p1}, R_w and R_B factors equal to 4.6%, 4.1%, 6.5% and 4.8%, respectively. The corresponding fit/measured visual agreement can be checked elsewhere [5].

The orientation distribution (OD) is represented by inverse pole figures (figure 2) calculated for the z fiber direction (parallel to the magnetic field). This figure illustrates two interesting points. First, it shows that the only component present in the four samples is the (00l) crystallographic planes perpendicular to the slip casting direction. Secondly, the degree of orientation is improved for the higher sintering temperatures: the maximum of the distribution density is increased and the c-axis distribution gets sharper. Based on the peak deconvolution (Popa formalism), the calculated crystallite sizes have been found to be equal to 137 nm for the sample sintered at 800°C and larger than 500 nm for samples sintered at 1300°C, 1400°C and 1600°C (crystallite sizes larger than 500 nm are not measurable under our resolution conditions). These sizes are well correlated to the scanning electron microscopy (SEM) analyses where we determined the grain sizes by the linear intercept method on the surface parallel to the magnetic field. The measurement was performed for two scanning directions, i.e. parallel and perpendicular to the magnetic field. The values, reported in Table 1, showed the spherical shape of the grains in a sample sintered at 800°C, where the grain size and the respective aspect ratio increase with increasing sintering time. Table 1 shows a resume of the results. The correlation of the calculated (001) orientation distribution, accessed by the Texture Index values, crystallite sizes, and microstructural observations, clearly proves that the growth of the initial small particles starts at temperatures above 800°C and that enhanced anisotropic grain growth by increasing sintering temperature is an important factor for texture development [5]. This mechanism is consistent with previous conclusions reported in different papers on the texturation of α-Al_2O_3 ceramics [6,7].

In conclusion, the Rietveld texture methodology combined with neutron diffraction is promising for generalising the characterisation of the polycrystalline materials and extending the refinement to the determination of the structure, μstrain [8], and phase proportions.

Specimens (sintering temperature)	ODF (001) inverse pole figure Min	Max	Texture Index (F2)	Refined crystallite size (nm)	SEM Calculated grain size(nm) $d_{//}$	$d_{\perp}$	Aspect Ratio ($d_{\perp}/d_{//}$)
800°C	0.47	2.4	1.24	137 (13)	~150	~150	1
1300°C	0.21	4.9	2.13	> 500 nm	1100	1170	1.063
1400°C	0.08	7.9	3.16	> 500 nm	2610	2970	1.138
1600°C	0.05	19.4	7.78	> 500 nm	7300	8800	1.205

Table 1: Minima and maxima of (001) inverse pole figures and texture index. Refined crystallite size (Popa formalism) and calculated grain size by line intercept method (SEM analysis), parallel ($d_{//}$) and perpendicular ($d_{\perp}$) to the magnetic field direction.

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