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Magnetic field alignment in highly concentrated suspensions for gelcasting process

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Abstract

The aim of the current paper is to present the development of the method used for fabricating highly textured α-alumina elements. The experimental procedure described is based on a combination of two techniques: gelcasting is used in tandem with high magnetic field exposure. In the element shaping process, a new, low-toxic and environmentally friendly gelating system was implemented that is based on *in situ* polymerisation of an acryloyl derivative of galactose. Here, care was taken to achieve a highly textured structure of elements gelcast from the ceramic slurries of high solid contents (45–50 vol%), which have not been processed successfully for magnetic field alignment before. A secondary aim of the study was to investigate the combined effect of magnetic field exposure duration and the idle time of polymerisation on the effectiveness of the alignment. In the course of the experiment, high degrees of crystalline orientation of the gelcast samples were obtained ranging from 0.92 to 0.96, developed after the subsequent sintering at 1600 °C (the parameters were calculated on the basis of XRD pattern of the surface perpendicular to the magnetic field direction). The microstructures of the obtained elements are described in detail at the end of the paper, in a stereological analysis.

Keywords: A. Shaping; A. Suspensions; D. Alumina; Texturisation

1. Introduction

Recently a lot of studies have been conducted to develop a convenient way of fabricating textured ceramics which is a way of improving certain properties of the materials, including mechanical, electrical or piezoelectric ones, depending on the ceramic material used for texturing [1]. The advantage of the textured material over the same one of a random orientation is the possibility to tailor the properties, which differs with the direction of the interaction, due to their anisotropy. However, textured ceramics has not been produced for a large industry scale yet, due to insufficient method of manufacturing, which still need some development. A lot of research on the field of producing textured ceramics specifically refers to magnetic alignment which has become a very promising alternative for methods such as templated grain growth [2,3], reactive templated grain growth [4] or hot forging [5,6]. The main idea behind the process is to utilise high magnetic field to cause the rotation of the suspended ceramic particles. The basic requirement here is that single crystals of a given substance exhibit anisotropic magnetic susceptibility. Texture develops if the shaping process is followed by sintering at high temperatures, which results in large grain growth of the particles [7,8].

There are a lot of ceramic materials which are studied aiming at producing textured structure, mostly piezoelectric and ferroelectic, which has a high anisotropic magnetic susceptibility and so implementing magnetic field even of a low level is very effective for texturisation. α-Alumina, which was chosen for current research as a reference powder, widely used for industry applications, exhibits very low value of anisotropic susceptibility. As superconducting magnets have been developed, the potential

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application of strong magnetic fields becomes more and more available. Due to the influence of strong magnetic field, the energy of alumina crystal anisotropy becomes comparable or larger than the energy of thermal motion, so it can be textured by means of described method. It has been reported that textured alumina elements exhibit much higher bending strength than those of random orientation [9] and also excellent wear resistance [10]. This can be taken as an advantage for the needs of cutting tools industry [10]. It was also found that the optimum level of magnetic field to ensure proper alignment is 10–12 T [9].

Magnetic field alignment is typically accompanied with a shaping technique based on colloidal processing, such as slip casting [11,12] or electrophoretic deposition [13,14]. In this case, one of the major requirements for proper alignment is a good dispersion of the ceramic particles in the suspension of the lowest viscosity possible [15,16], since agglomeration and strong interactions between particles are a serious hindrance for rotation. One could make a point that the lower the concentration of ceramic powder in the slurry, the higher the probability of effective alignment. It needs to be remembered, though, that the greater the amount of solid content in the suspension, the more homogeneous products can be shaped from it. The industrial slip casting often applies ceramic slurries of high solid contents (> 50 vol%) and low viscosities at the same time. However, it has been reported [9], that the slurries of alumina concentration above 40 vol% cannot be textured effectively, due to strong interactions between particles. It has been stated that, for slip casting of submicron alumina powder, the maximum amount of solid loading in the slurry that assures unhampered rotation of the particles is 40 vol% (71.7 wt%).

Gelcasting, a method applied in the current study instead of slip casting, is based on a direct casting of highly concentrated suspensions (40–55 vol%) into nonporous moulds. An important advantage of this technique is that the obtained green products can exhibit high mechanical strength [17]. This is because the polymerisation of an organic monomer takes place *in situ* in the ceramic suspension, which results in creating a strong macromolecular network holding the ceramic particles together. Such a procedure lowers the cost of shaping, as there is no need to machine the sintered products by expensive diamond tools [18]. This advantage has caused constant development of the technique over the recent years [19,20], especially in the fields of new gelating systems or the monomers applied [21,22].

The idea behind utilising gelcasting with magnetic field exposure (henceforth, MFE) is that the creation of polymeric network can occur right after the rotation of the particles, blocking them from rotating back to another angle when the magnetic field is removed [23,24]. This has certain advantages over using slip casting, such as increasing the homogeneity of the obtained products, especially if complex shape is required for application. Preliminary research into the issue has already yielded promising results: a highly textured structure of sintered α-alumina was obtained from a slurry of 45 vol% (75.7 wt%) Al_2O_3 (TM-DAR, *Taimei Chemicals Ltd, Japan*) in the presence of magnetic field 12 T for 6 h [25]. This effect could be obtained, thanks to using a new self-synthesised environmentally-friendly compound-acryloyl-D-galactose – as a gelcasting agent [26]. The present research develops this idea, aiming at obtaining highly textured alumina elements gelcast from the suspension of the highest possible solid content – 50 vol% (79.2 wt%). The authors intention was to achieve the maximum level of grain orientation, as the higher level of texturisation, the better effect of properties improvement is observed. The secondary purpose of the study was to explore how factors such as viscosity of the slurries, the idle time of *in situ* polymerisation, and the duration of magnetic field exposure (MFE) affect the effectiveness of the alignment.

2. Experimental details

2.1. Materials

Spherical α-Al_2O_3 TM-DAR of average particle size 0.15 μm and high purity (> 99.9%) (*Taimei Chemicals Ltd.*, Japan) was used as the model ceramic powder. The monomer, 6-O-acryloyl-D-galactose, (Acr-Gal), which polymerised *in situ* after casting, was synthesized from galactose in three stages [26]. Ammonium polycarobxylate A6114 (*TOAGOSEI, Aron Dispersant Series*, Japan), applied as 40 wt% aqueous solution was chosen as a dispersant; N,N,N′,N′-tetramethylethylenediamine, TEMED (*Fluka*) in 10 wt% aqueous solution was an activator (catalyst) and ammonium persulfate, APS (*Aldrich*) in 1.0 wt% aqueous solution was the initiator of the polymerisation.

2.2. Experimental procedure

All the operations were conducted at room temperature. Firstly, the ceramic slurries were composed. The solvent (distilled water) was poured to the plastic containers and the solution of the monomer was added. Then the solution of the dispersant was dropped slowly, followed by addition of the activator. The amounts of the components are listed in Table 1. Finally, alumina powder was added to the premix (45 and 50 vol%) and the suspensions were mixed by a magnetic stirrer for 90 min followed by 10 min mixing and degassing in THINKY ARE-250 planetary mixer in order to attain good homogeneity. Just after adding the specified amount of the initiator (0.3–0.7 wt% based on monomer content) and additional 3 min mixing by the magnetic stirrer, the slurries were poured to circular plastic moulds (20 mm diameter, 10 mm height) stuck to a plastic surface and placed in a superconducting magnet (Japan Supercoductor Technology, Inc. JMTD-12T100). A strong magnetic field of 12 T was applied to cause the rotation of the particles for 15 min to 6 h. Providing that the magnetic field level was uniform in the volume of the samples (in the disc of 40 mm in diameter, the distribution of magnetic field is within 1.3%), the influence of the thickness of the samples to align each particle is negligible in the region of thickness of at least 40 mm. The diameter of the samples depends on the bore size of the magnet (100 mm in diameter used in this experiment) and the uniformity depends on the distribution of the magnetic field. The thickness of the sample is also an important factor for the gelcasting process itself, as more difficulties with proper drying occur with enlarging the gelcast product. Hence, small diameter of the samples was chosen to limit the number of factors influencing