Available online at www.sciencedirect.com
ScienceDirect
Journal of the European Ceramic Society 34 (2014) 3841–3848
www.elsevier.com/locate/jeurceramsoc

Fabrication of textured alumina by magnetic alignment *via* gelcasting based on low-toxic system

Agnieszka Szudarska[a,*], Tadeusz Mizerski[a], Yoshio Sakka[b], Tohru S. Suzuki[b], Mikolaj Szafran[a]

[a] *Warsaw University of Technology, Noakowskiego 3, 00-664 Warsaw, Poland*
[b] *National Institute for Materials Science, Tsukuba, Ibaraki, Japan*

Received 16 January 2014; received in revised form 29 April 2014; accepted 10 May 2014
Available online 28 May 2014

Abstract

The controlled development of texture in ceramics allows to improve some electrical, piezoelectric and mechanical properties of advanced ceramics materials by tailoring the microstructure. A highly textured microstructure of undoped dense alumina was attained by applying a novel combination of gelcasting techniques with magnetic alignment followed by sintering. A newly synthesized low-toxic acrylic monomer based on galactose was introduced to the gelcasting procedure. Thermal analysis of the gelcast has been performed to examine decomposition process of the new binder and match appropriate sintering rate. The effectiveness of the proposed procedure has been related to rheological properties of the suspension by clarifying the influence of the powder and the dispersant content. It has been also related to the idle time of polymerization by clarifying the influence of the initiator content. The new method is compared to slip casting in high magnetic field that has been used widely so far.

Keywords: Alumina; Gelcasting; Magnetic field; Texture control

1. Introduction

The fabrication of textured ceramics have been carefully studied recently, due to a growing necessity of manufacturing advanced ceramics materials that have to meet definite requirements concerning their properties. A controlled development of crystalline texture in ceramics makes it feasible to improve some electrical, piezoelectric and mechanical properties of those materials.[1,2]

There are few methods of producing textured ceramics elaborated so far, including template grain grow (TGG)[3–5] or hot forging.[6,7] Magnetic alignment is one of the most recent and well researched method. The idea is to make the ceramic particles in a suspension aligned during the colloidal processing due to the influence of a high magnetic field. The strong texture can be developed during subsequent sintering of the shaped elements.[8] The shaping itself is conducted usually by slipcasting method[9–12] or electrophoretic deposition (EPD).[13,14] This way of fabricating textured ceramics is becoming popular because it does not require large templates or high uniaxial pressure. The main principle of the process is that a crystal with anisotropic magnetic susceptibility will rotate to an angle minimizing the system energy when it is placed in a magnetic field.[15,16]

There are definite requirements for obtaining textured ceramics by slip casting in a high magnetic field, that has been reported.[17] The particle of the ceramic powder should be a single crystal and the crystal structure should be non-cubic, so then an anisotropic magnetic susceptibility should appear. The particles should also be well dispersed in the ceramic slurry of the viscosity low enough to enable the rotation of the particles with a low energy, while magnetic energy should be higher than the thermal motion energy. When the particles of ceramic powder are spherical, the grain growth is also essential to get a highly oriented structure.

α-Alumina, commonly used in ceramics processing, belongs to the rhombohedral crystalline system. It exhibits anisotropic susceptibility, but this anisotropy had been more or less ignored due to its very low value.[8] The development of superconducting magnet has extended the potential applications of strong

* Corresponding author at: Department of Chemical Technology, Faculty of Chemistry, Warsaw University of Technology, 3 Noakowskiego Street, 00-664 Warsaw, Poland. Tel.: +48 22 234 5586; fax: +48 22 6282741.
E-mail address: aszudarska@ch.pw.edu.pl (A. Szudarska).

http://dx.doi.org/10.1016/j.jeurceramsoc.2014.05.017

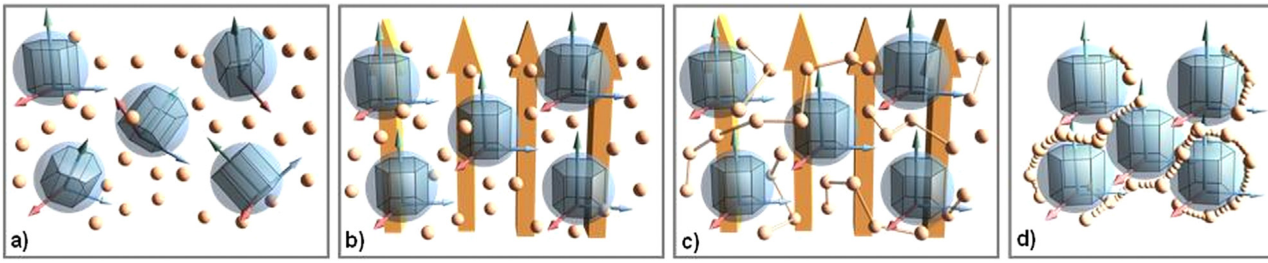

Fig. 1. The idea of magnetic field alignment method combined with gelcasting. (a) Randomly oriented particles dispersed together with monomers molecules, (b) influence of high magnetic field makes the particles oriented, (c) polymerization reaction during the exposure to magnetic field, and (d) oriented particles locked by the polymer macromolecular network.

magnetic fields, where the energy of crystal anisotropy becomes comparable to or larger than the energy of thermal motion.

The key factor in this case is the proper dispersion of solid particles in the ceramic suspension used for the casting.[15–18] The strong interaction between the particles which are agglomerated hinders them from rotating. That is the reason why colloidal processing is implemented here, as it allows to control the dispersion and the consolidation of the suspension during powder processing.[15,16]

One of the motivations for the present research was to introduce gelcasting instead of slip casting as the main colloidal processing method for shaping the elements, because of several advantages and economical benefits. The idea of gelcasting process is to consolidate the suspension of ceramic powder and monomer particles by controlled *in situ* polymerization after pouring to a not-porous mould,[19] whereas the consolidation in slipcasting process occurs due to the absorption of the solvent by a porous plaster mould. Gelcasting is then faster than slip casting, comparing the time of solidification.[20] The use of gelcasting instead of slipcasting for consolidation of suspensions might allow to reduce the exposure time to high magnetic field, especially when the particles after the rotation are locked by the macromolecular network of the polymer created *in situ*. The idea of producing textured ceramics *via* magnetic alignment combined with gelcasting technique is schematically shown in Fig. 1. Moreover, elements obtained by gelcasting present high mechanical strength in the green state, so that low cost green machining could be applied.[21] Green bodies are characterized also by high relative densities due to high solid contents of the slurries thus cold isostatic pressing is not needed before the sintering. Moreover it allows to obtain large and more complex-shape elements, so it opens a variety of different application possibilities.

Few successful attempts have been made to combine magnetic alignment with gelcasting technique based on acrylamide, for fabrication of textured iron titanate, as a prototypic anisotropic ceramics,[22] ferroelectric ceramics[23] or ZnO-based ceramics.[24] However, there was no further investigation of the problem has been carried out and alumina powder has not yet been applied for this combination of the methods.

The gelcasting technique is becoming more and more popular for manufacturing advanced ceramic materials, however it still needs some further developments. One of the main disadvantage is the necessity to use toxic acrylic monomers, such as acrylamide, which is probably carcinogenous substance. It also contains nitrogen atom in the molecule, so during the sintering process nitrogen oxides are released to the atmosphere, what is harmful for the environment. The monomer plays a pivotal role in the shaping process, because after the controlled reaction of polymerization, it has to ensure a good mechanical strength of the green element. That is the reason why there has been a lot of research done focused on replacing toxic processing agents, but other commercial monomers, such as 2-hydroxyethyl acrylate or 2-hydroxyethyl methacrylate have been found less effective than acrylamide.[25,26] Following the 'green chemistry' trend most recently some entirely new low-toxic acrylic monomers, based on polyhydroxy compounds, have been synthesized at the Warsaw University of Technology.[27–30] One of them, the newest, is galactose monoacrylate (6-O-acryloyl-D-galactose), which was successfully used in the gelcasting process,[31] so it has been applied also in the present work.

2. Experimental details

2.1. Materials

Ceramic powder used in the research was a high purity (>99.9%) spherical α-Al_2O_3 TM-DAR of mean particle size 0.15 μm (*Taimei Chemicals Ltd.*, Japan). Ammonium polycarboxylate A6114 (*TOAGOSEI, Aron Dispersant Series*, Japan) was used as a dispersant, in the form of a 40 wt.% aqueous solution. N,N,N′,N′-tetramethylethylene-diamine, TEMED (*Fluka*) used us a 10 wt.% aqueous solution played the role of activator (catalyst) and ammonium persulfate, APS (*Aldrich*) in the form of 1 wt.% aqueous solution was the initiator of polymerization. The monomer applied in the process was newly synthesized 6-O-acryloyl-D-galactose (6-Akr-Gal) in 30 wt.% aqueous solution. The synthesis was based on the former studies and was carried out in three stages.[31] The initial substrate was D-(+)-galactose (>98%, *Sigma–Aldrich*) which had its four from five hydroxyl groups in the molecule blocked by reaction with acetone in acidic medium in the first stage of the synthesis. Then one remaining hydroxyl group was estrificated by acryloyl chloride in the presence of an amine. After the acryloyl group had been inserted into the molecule, acidic hydrolysis could be carried out as the final step of synthesis to unlock the remaining hydroxyl groups. Such procedure ensures that only one acryloyl group is introduced into the molecule and there are no other