MECHANO-CHEMICALFABRICATIONOF NON-FIRING CERAMIC

Apiluck Eiad-ua[1*], Jiti Nukaew[1], Takashi Shirai[2] and Masayoshi Fuji[2]

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Abstract

Novelroute for non-firing ceramic was successfully fabricated in this paper. This study is particularly taking advantage of mechanochemically assisted chemical solidification for improving mechanical strength of ceramic green body. This method allows the surface activation of powder and control of the properties of powder not only by varying mechano-chemical treatment but also by managing the chemical solidification by alkaline binding. After drying at 25°C for 3 days, the ceramic green body (non-firing ceramic) was obtained. The results revealed that mechano-chemical treatment has a certain effect on the total bonding (Potassium ananate, $KAlH_4$) and mechanical strength of non-firing ceramic. An increase in total number bond and high strength was obtained with milling time of mechano-chemical process.

Keywords: Non-firing ceramic, novel route, mechano-chemical, solidification

Introduction

Nowadays, with background of lively activity there is a large demand in ceramic industry. Firing ceramics is one of the most common techniques in fabrication porous or dense materials (Rahanan, 2003). However, this method has disadvantages in terms of its high cost and its potential to be harmful to the environment because the coal which is burnt as fuel emits CO_2, SO_x and NO_x, ash dust during the burning process. CO_2 is inevitably created by burning fuels such as oil, natural gas, and diesel during burning process. Carbon dioxide emissions, therefore, are the most important cause of global warming. The emissions of CO_2 have been dramatically increased within the last 50 years and are still increasing each year (Meinshausen *et al.*, 2009). Ceramic industry today requires low cost ceramic fabrication, while at the same time demanding excellent performance. Recently, Low Temperature Co-fired Ceramics (LTCCs) (Jantunen *et al.*, 2000) cement (Eroglu *et al.*, 2007) and geopolymer (Fletcher *et al.*, 2005) have been intensively

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[1] *College of Nanotechnology, King Mongkut's Institute of Technology Ladkrabang, 1 Soi Chalongkrung 1, Ladkrabang, Bangkok 10520, Thailand. Tel. +66-2329-8000 ext 3132; Fax: +66-2329-8265; E-mail: keapiluc@kmitl.ac.th*

[2] *Ceramics Research Laboratory, Nagoya Institute of Technology, 3-101-1, Honmachi, Tajimi, Gifu 507-0033, Japan.*

* *Corresponding author*

studied for these applications. However, this is the high cost and remaining carbon dioxide emissions of process. Non-firing ceramic by mechanochemically assisted chemical solidification has been proposed. Mechano-chemical treatment is a novel method involving the mechanical activation (Delogu *et al.*, 2003), a process for introduction of additional energy to the system applies worldwide. During the treatment, various structural changes of the material are taking place. These structural changes of the material cause changes of its properties (chemical, electrical, thermal, mechanical, etc) as well as improvement of its reactivity. The most popular devices, in which mechano-chemical processes can be conducted, are vibratory, planetary and attritor ball mills. They differ in their capacities, efficiencies of milling, and additional arrangements such as cooling. In all these devices, the ground material is periodically thrown into zones of ball collisions. Energy transfer to the powdered particles takes place by shearing and/or impact action of the balls (Deloguand Cocco, 2003; Delogu *et al.*, 2003). Mechano-chemical treatment by planetary ball mill is now recognized as a powerful tool for the synthesis of materials, such as amorphous alloys (Delogu and Cocco, 2003), nanocrystalline metals and alloys (Tsuzuki and McCormick, 2000), and ceramic materials (Stojanovic, 2003). This method is very simple and economical for the production of powders of very small size. The use of fine starting materials and mechano-chemical treatment by efficient activation are the key steps in this process. Therefore, this study examined whether mechano-chemical treatment of starting materials using ball milling activation can reduce the sintering temperature of solid state reacted (Balema, 2000; Hsing and Yen, 2003). Solidification phenomena are the heart of most of the products of manufacturing process. From manufacturing process like foundry casting, solidification phenomena play a dominant role in process as well as product optimization. Besides these, the new advances in the understanding of the process have led to the development of several new technologies (Leeand van Deventer, 2007). Solidification involves the mixing of raw material with binder to cast the ceramic specimen. Alkaline activation of aluminosilicate using alkaline hydroxide and/or silicate solutions can be used to synthesize inorganic geopolymeric binders, or alkaline activated cements, displaying excellent physical and chemical properties (Perera *et al.*, 2007). On the other hand, it is well known that alkaline solution is a good binder between ceramic particles, which is responsible for solidification to form a strong green body at room temperature (Eiad-ua *et al.,* 2004; Eiad-ua *et al.*, 2010).

Alumina iswidely known because it is an important industrial raw material. Alumina surface has a variety of applications in ceramic industry. However, the fabrication route to create the surface of alumina results in surface defects in the crystal lattice. In this work, we investigated the surface activation of alumina on mechanochemicallyassisted chemical solidification to provideceramic green body (non-firing ceramic) with improved mechanical strength. The physical and chemical properties are determined with XRD, FTIR, Raman, ICP, TEM and mechanical strength.

Experimental Procedure

Sample Preparation

The raw materials were α-Al_2O_3 (AA-04) of 0.5 μm mean particle size from Sumitomo Chemical Co., Ltd, and KOH pellets (JIS K 8574). Planetary ball mill was used for mechano-chemical treatment of starting material. The mill was rotated at 300 rpm and different milling timesfm 0 to 60 min. After being mixedwith 3 mol dm^{-3} of KOH (alkali treatment) at 2000 rpm for 5 min with an electric mixer, the slurry was poured to Teflon mold and kept at room temperature until solidification. After demolding, the specimens were dried at 25°C, 50% RH for 3 days in the steam oven to obtain ceramic green body.

Analytical Techniques

X-ray diffraction (Model RINT 1000, Rigaku, Cu Kα, 40kV) was used for the qualitative determination of the composition and crystalline phases present in samples over the scan range 20-70° for wide angle XRD. The IR diffuse reflectance spectra of the prepared products were measured in the range of 400-4000 cm^{-1} with the Fourier transform IR spectrometer (Jasco, FTIR-6200). Inductively coupled plasma (ICP) is an analytical technique used for the detection of trace metals using Plasma Spectrometer (SPS-7800). Surface activation of alumina was observed in the micrographs obtained using TEM (JEOL-2100F) operating at 299kV, after depositing the powder dispersed on carbon coated grids. The mechanical property was evaluated by three points testing (Compression strength) using universal testing machine (Shimadzu AGS-G). All tests were performed at room temperature. The test results were recorded using an average value of three measurements.

Results and Discussion

Key concept of non-firing ceramic is shown in Figure 1, providing ceramic green body with an improved mechanical strength, wherein the ceramic green body is fabricated by activating ceramic powder through mechano-chemical treatment and solidifying the activated powder through alkali treatment. Activated ceramic powder having mechanochemically activated surfaces is obtained by milling ceramic powder using planetary ball mill (mechano-chemical process), and ceramic green body is obtained by adding alkali water solution containing alkaline metal hydroxide to the powder (alkali treatment process).

Figure 2 shows the X-ray diffraction patterns for 0, 15, 30, and 60 min at 300 rpm (Raw, 300-15, 300-30, and 300-60), respectively of the activated alumina in Figure 2(a), and ceramic green body in Figure 2(b). From Figure 2(a), it can be seen that no significant change in the XRD patterns after milling were observed, while the intensity peaks slightly decreased and broadened with milling time. This can indicate that amorphous phase formed with shearing force during mechano-chemical treatment. On the other hand, we have detected the new bonding of Potassium Alanate ($KAlH_4$) in range 2θ = 28-32° shown in Figure 2(b). It can be confirmed that Potassium Alanate can be obtained from Al_2O_3 and KOH via alkali treatment.

Figure 3 shows the FTIR spectra from 4000-1000 cm^{-1} of the activated alumina in Figure 3(a), and ceramic green body in Figure 3(b). The adsorption bands were sharp and shift with prolong milling time. The adsorption bands from 3600-3200 cm^{-1} correspond to OH from water. As Figure 3(b) show the several infrared active Al-H vibrations between 2000-1000 cm^{-1} (Ares *et al.*, 2009). The spectra region of 1700-1450 cm^{-1} were combination bands of Al-H and absorbance peaks also sharp in ceramic

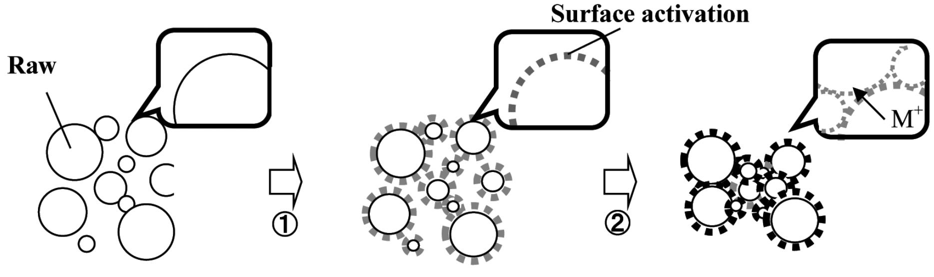
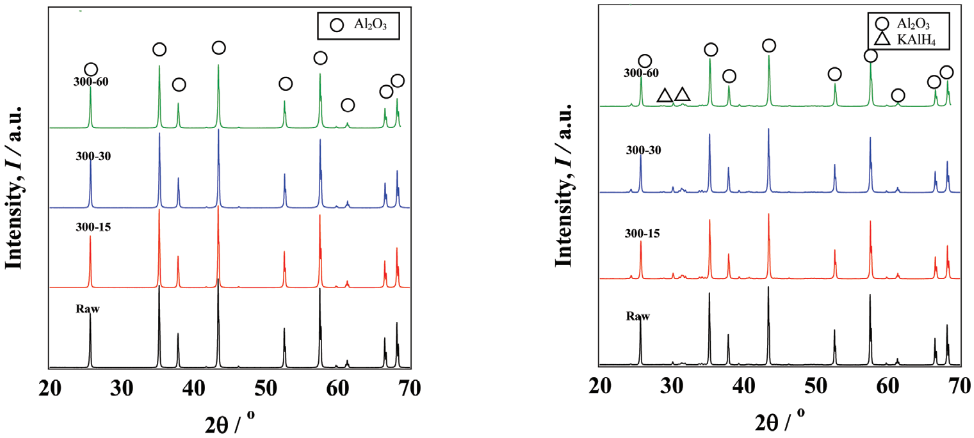


Figure 1. Novel fabrication route for non-firing ceramics by mechanochemically assisted chemical solidification

green body. It is the possibility of formation of hydrogen bonds between AlH_x^- and K^+ could be assigned to the formation of $KAlH_4$ or an intermediate compounds.

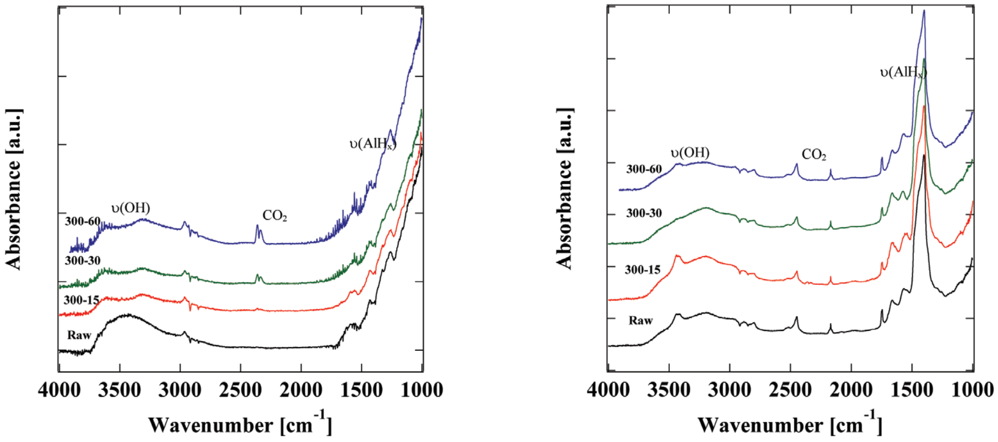
Figure 4 shows Raman spectra of powder of activated alumina in Figure 4(a), and ceramic green body in Figure 4(b). Figure 4(a) show all bands of corundum compound in mehanochemically activated powder (Assih *et al.*, 1998). On the other hand, the increasing and sharpening of all bands in Figure 4(b) were remarkable. This is because of effect of OH group from H_2O during alkaline activation (Schoonover *et al.*, 2003). The new band at 1080 cm^{-1} related to aluminate band (Agnew and Schoonover, 1996; Schoonover *et al.*, 2003).

Figure 5 shows the elution behavior of activated and unactivated alumina powders at different mechano-chemical treatments. The Al^{3+} increased with increasing milling time. This is unstable phenomenon on the surface of



(a) (b)

Figure 2. X-ray diffraction patterns of powder at different conditions; Activated powder, (b) Ceramic green body



(a) (b)

Figure 3. FTIR spectra of powder at different conditions; (a) Activated powder, (b) Ceramic green body

alumina with shearing force during mechanochemical treatment. The dissolving alumina surface with mechano-chemical treatment will therefore be enhanced in enriched in Al, which is consistent with a surface reaction-controlled mechanism (Lee and van Deventer, 2007).

TEM measurements can be applied for estimating the composition and microstructure of samples. A high resolution obtained in this type of microscopy allows single grains of the samples to be observed. The particle morphology of the raw, activated and ceramic green body samples were shown by TEM images in Figure 6(a), 6(b), and 6(c), respectively. As shown in Figure 6(a), the alumina powder consists of regularly shaped plate-like particles. The presence of amorphous layer in the activated powder surface was confirmed in Figure 6(b). Mechanical milling induced amorphization and temperature rise may promote interaction between ball and powder. On the other hand, it can be seen in Figure 6(c) that certain lattice planes are continuous across the particle boundaries, which indicates the presence of bonding or solid bridge as a Potassium Alanate. The single oxide powder of Al_2O_3 consists of solid bridge or bonding with a reasonably narrow size distribution (Eiad-ua *et al.*, 2004; Eiad-ua *et al.*, 2010).

Results of compression strength of ceramic green body were shown in Figure 7. The strength increased with milling time and the presence of Potassium Alanate. Maximal strength was reached, when activated for 30 min. Then, this strength decreased slightly in prolonged milling time. These early high strength characters are noteworthy in view of practical applications.

Conclusions

Non-firing ceramic has been fabricated by mechanochemically activated chemical

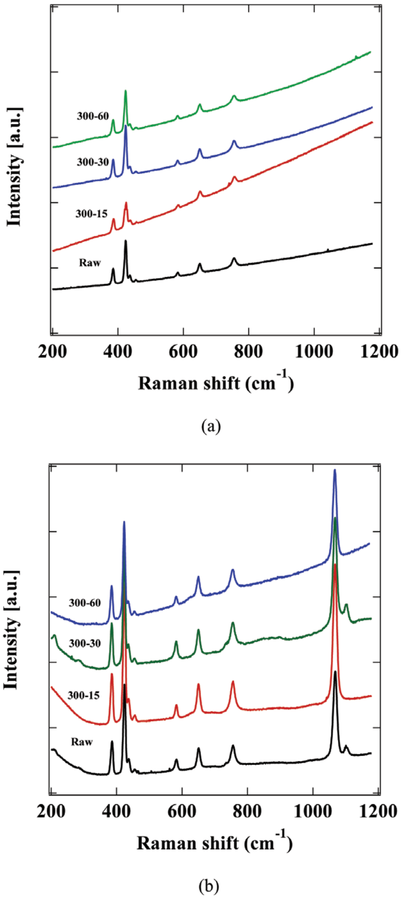


Figure 4. Raman spectra of powder at different conditions; (a) Activated powder, (b) Ceramic green body

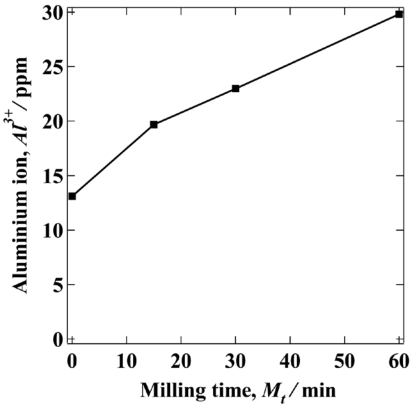


Figure 5. Elution behavior of mechanochemically activated alumina

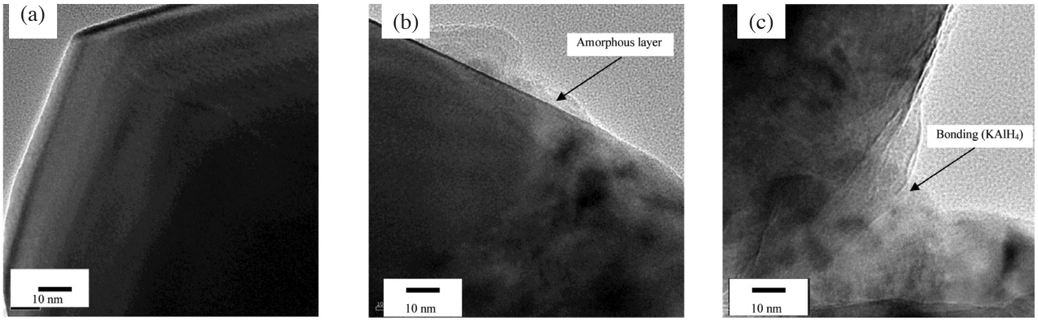


Figure 6. Transmission electron micrographs of powder at different conditions; (a) Raw, (b) 300-60, (c) Ceramic green body

solidification process. The intrinsic advantage of mechanno-chemical treatment lies in its ability to affect a solid-state reaction through mechano-chemical activation, instead of by the calcinations and/or sintering at high temperature. Mechano-chemical treatment increased the number of particle contacts, enhanced dissolving of alumina surface to form new bonding, improved the mechanical strength, and enhanced microstructure development. This new route is applicable to the fabrication of other functional materials with its simple process, low cost, and friendly environment.

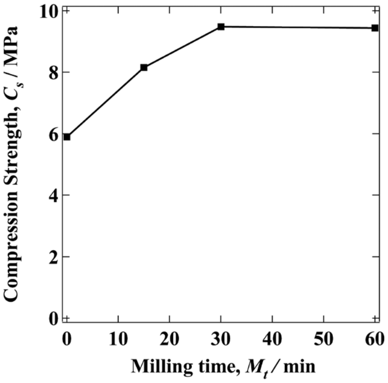


Figure 7. Compression strength of ceramic green body

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