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Mechanochemical activation of aluminum powder and synthesis of alumina based ceramic composites

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Abstract

The microstructure and chemical reactivity of aluminum powder ball milled in ethanol medium have been investigated in this study. It has been shown that the milling environment plays an important role on the chemical behavior of the milled aluminum powder. Aluminum particles ball milled in ethanol could physically bond a significant amount of hydroxyl (OH) and alkyl (CH) functional groups, the presence of which drastically changes the oxidation behavior of the milled powder. Such mechanochemically activated (MA) aluminum powder obtained by ball milling in ethanol was found to oxidize at temperatures less than half of that for oxidation of original powder. This is thought to be because of refinement and modification of particle surface of MA aluminum powder. Moreover, even at the reduced temperature of oxidation, the MA aluminum powder oxidizes mainly in the form of high temperature phases such as γ-Al_2O_3 and α-Al_2O_3 (corundum). Utilizing the specific chemical behavior of MA aluminum powder, we demonstrated that it is possible to prepare alumina–SiC ceramic composites directly from the mixture of MA aluminum and SiC compounds at significantly lower temperature. Some important parameters of the synthesized ceramics have been investigated.

Keywords: A. Milling; A. Powders: chemical preparation; B. Composites; D. Al_2O_3

1. Introduction

Ball milling has been a powerful tool employed by many research groups for preparation of nanostructured materials [1–3]. Generally, the production of nanomaterials via mechanical processing techniques occurs by two different mechanisms: (i) by refinement of coarser grained structures, and (ii) as a product of mechanochemical reactions [2]. A large variety of aluminum-based nanocomposites have been synthesized by mechanically induced displacement reactions using aluminum (Al) powder as a reducing metal [2,3]. Mechanochemical displacement reactions may progress in two different ways: (i) gradually—by producing nanostructured powders, and (ii) by combustion [2–4]. In the latter case, the heat liberated during combustion melts the reaction mixture and destroys the nanostructures. Inert additives could be added to avoid combustion and control the fine microstructure of the products [2]. Several organic solvents (ethanol, acetone and toluene) have been used as inert additives in vibratory ball milling to avoid combustion of highly exothermal mixtures of reactants containing Al powder [4,5]. However, the presence of different solvents exerts different influences on the microstructure and post-processing behavior of Al containing mixtures. Here, a series of experiments were carried out to investigate the changes in microstructure and chemical reactivity of Al powder on ball milling in different organic media. Specifically, we focused on studying the oxidation behavior of Al powder during ball milling in ethanol, and describe a new approach of preparing alumina based ceramic composites.

2. Material and methods

The starting materials used were Al powder (99.5%) with particle sizes 10–30 μm, obtained from AlfaÆsar, and ethanol (Anhydrous, 99.8%, AcroSeal) obtained from Fisher Scientific. SiC powder (99.5% metals basis) of particle sizes 3–5 μm, obtained from AlfaÆsar was used to prepare green mixture of

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alumina–SiC ceramic composite. Mechanical processing and dispersion of Al powder were carried out by using two types of ball mills: (i) vibratory ball mill with 4 mm amplitude and 25 Hz frequency, and (ii) planetary ball mill (Pulverisette 5, Fritsch, Oberstein, Germany) at 300 revolutions per minute (rpm). The vibratory mill had two stainless steel cylindrical containers of 100 ml volume, each of which was filled in with 30 steel balls of 10 mm diameter. The weight of starting Al powder was 3 g dispersed in 30 ml of ethanol. The planetary mill used for preparing green mixture for ceramic composites had 2 stainless steel bowls of 250 ml capacity, each of which was filled with 10 stainless steel balls of 20 mm diameter. The weight of starting Al powder was 12 g dispersed in 120 ml of ethanol. Based on the results of thermal analysis, we made a rough estimate and compared the effective processing time for both type of mills with the above described parameters. We found that in order to reproduce mechanochemically activated aluminum powder with the similar oxidation behavior, the time of processing in case of planetary milling was 5 times more in comparison with the vibratory milling technique.

XRD measurements were performed on a Philips X'Pert diffractometer in θ–θ geometry with Cu-Kα radiation. The XRD peaks were identified using the JCPDS database. The presented XRD patterns contain both the Kα_1 and Kα_2 contributions. For line width analysis, the Kα_2 sub-peaks were removed numerically.

Differential thermal analysis (DTA) and thermo-gravimetric analysis (TG) were carried out in temperature interval from 200 °C to 1000 °C by using a “Q-1000” high temperature differential thermal analyzer. All analyses were performed in air at a heating rate of 10 °C/min, and the sample weight was 30 mg.

Iron contamination of the samples was investigated using an energy dispersive X-ray (EDX) analyzer of JEOL EM-2010F transmission electron microscope (TEM).

The IR spectra of Al powder dispersed in ethanol were measured on a Thermo-Nicolet OMNIC FTIR (Fourier Transform Infrared) spectrometer (4000–400 cm^{-1}, 2 cm^{-1} resolution). Solid-state IR spectra were recorded using the KBr disc technique. Before the mechanochemical activation of Al powder in ethanol, Al powder was dried in a vacuum chamber for 2 h at about 50 °C in order to remove free or absorbed ethanol.

Bulk discs of Al–SiC composites of 20 mm diameter were consolidated from the green mixture of 50 mol% of activated Al and 50 mol% of SiC powder at 40 MPa pressure by ordinary uniaxial pressing in graphitic die and punch. In order to investigate the dependence of the product density and hardness from consolidation pressure, some of the samples were compressed using cold isostatic pressure (CIP) of up to 400 MPa. The raw mixture of ceramic composite has been prepared by the following way. Al powder was milled in ethanol medium in the planetary ball mill bowl for 21 h; then the mill was stopped and SiC powder was added into the bowl (mol ratio: 50% Al: 50% SiC). After adding SiC powder into the bowl the milling was continued for additional 1 h, then stopped and Al–SiC green mixture was unloaded and dried in vacuum at 50 °C for 1 hour. The dried green mixture was consolidated under 40 MPa pressure in the form of discs with 20 mm diameter by using the graphite die and punch. The consolidated samples were sintered under ambient pressure in air at temperature interval from 800 °C up to 1200 °C for 1 h in order to produce the final product. The samples were heated to the desired temperatures at 5 °C/min rate, held for the prescribed times, and then the furnace was cooled down. During sintering, specific oxidation of MA aluminum resulted in transformation of the entire aluminum content of the green mixture into alumina, thus forming final Al_2O_3–SiC composite. The green density and final density of the sintered bodies were estimated by the Archimedes method in kerosene and water, respectively. The relative density of alumina ceramic was based on 3.98 g/cm^3, and the relative densities of alumina–SiC composites were calculated according to the weight percentage of SiC in the composite, assuming that d=3.21 g/cm^3 for α-SiC. The Vickers hardness of the formed composites was measured by a testing machine (MVK-E), Akashi Co., Japan, under load of 98.1 N applied to the samples for 15 s. The average hardness value was determined from 10 indentation measurements.

Electrical resistivities of the bulk ceramic samples were evaluated by an Insulation Tester Model 307 (BK Precision) at 1000 V DC.

3. Results and discussion

The results of XRD analysis of the starting Al powder and the powder after 60 min vibratory ball milling in ethanol are shown in Fig. 1. It is evident from both the figures that only main characteristic peaks of Al are seen. However, broadening and shift of XRD spectra of the milled Al powder (line 2 in Fig. 1) indicate that the microstructure of the powder has been changed significantly after ball milling. Table 1 presents a comparison of the peak positions and full width of half maximum (FWHM) of (111) XRD peak of initial and MA in ethanol Al powders.

By using peak positions and FWHM, a Williamson Hall graph was plotted for initial Al powder as well as for Al powder ball milled in ethanol for 60 min. The average domain sizes and lattice strain have been calculated and are listed in Table 2.

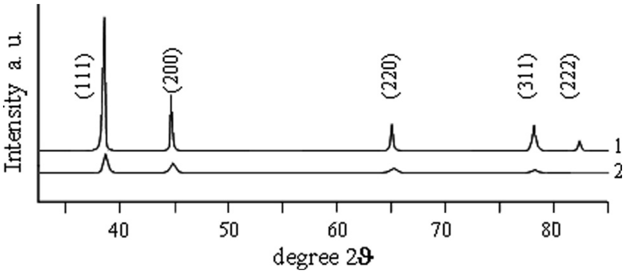

Fig. 1. XRD patterns of initial Al powder and Al powder ball milled for 60 min in ethanol medium: (1) initial Al powder and (2) Al powder ball milled for 60 min in ethanol.

Table 1
Peak position and FWHM of (111) XRD peak for initial Al powders and after 60 min ball milling in ethanol.

Sample	*hkl*	Peak position 2θ (deg)	FWHM (deg)	Int. breadth (deg)
Initial Al	111	38.3254	0.1213	0.1465
MA in ethanol Al	111	38.4369	0.2759	0.3403

Table 2
The average strain and domain sizes of initial and 60 min ball milled in ethanol Al powders.

Sample	Average strain	Domain average size (nm)
Initial Al	6.54×10^{-4}	10^3
MA in ethanol Al	5.07×10^{-2}	10^2

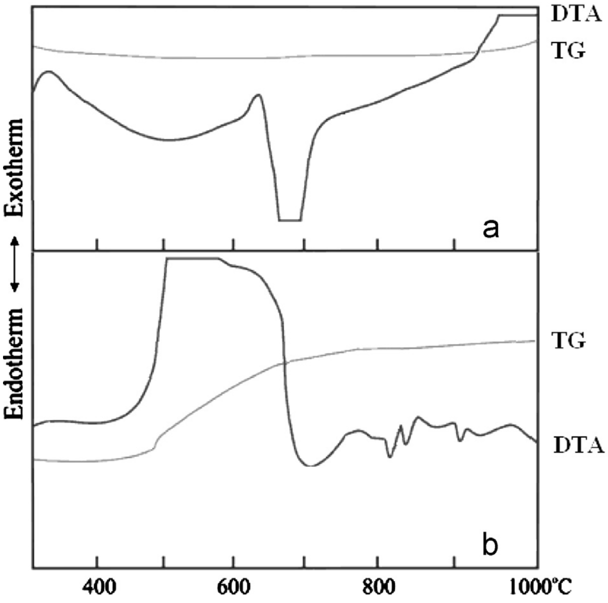

Fig. 2. DTA and TG traces of initial Al powder and Al powder ball milled for 60 min in ethanol: (a) initial Al powder and(b) Al powder ball milled for 60 min in ethanol.

As evident from Table 2, both domain size and strain of aluminum lattice have been significantly changed due to mechanical processing in ethanol medium.

Fig. 2 shows the results of differential thermal analysis (DTA) and thermo-gravimetric analysis (TG) for initial and activated Al powders. As evident from Fig. 2, the DTA trace of initial Al powder (Fig. 2a) does not indicate any oxidation process before reaching the melting point of Al at about $T_m = 680$ °C. Endothermic peak centered at 680 °C describes the process of Al melting. The absence of oxidation is also supported by the absence of any change (Fig. 2a) in sample weight until 900–950 °C. The oxidation of initial Al powder starts well above 900 °C. Contrary to the initial Al powder, the milled powder shows a completely different oxidation behavior. Fig. 2b clearly shows that the DTA trace of MA Al powder has well defined exothermic peak before reaching the melting point of aluminum. Both DTA and TG traces shown in Fig. 2b prove that oxidation of MA Al powder starts at about 450 °C and ends around 700 °C. Based on the results of thermal analyses we notice that chemical behavior of MA Al powder has changed drastically and it starts oxidizing at considerably lower temperature (450 °C) in comparison to the initial (900 °C) temperature. We assume that such a drastic change in chemical behavior of MA Al powder could be contributed by several factors, such as reduction in particle sizes, creation of nucleation centers for oxidation due to a possible iron contamination of the sample and because of modification of superficial layers of aluminum particles. Based on XRD analysis, we already observed that the particle sizes of 60 min ball milled Al powder decrease to about 1 order of magnitude from the initial particle size. Although XRD does not provide information about possible iron contamination or modification of particle's superficial layers, both these factors could play an important role in oxidation behavior of MA Al powder. It is known that iron compounds such as hematite (α-Fe_2O_3) or $Fe(NO_3)_3$ are commonly used as seeding agents in aluminum hydroxide gels in order to facilitate the formation of aluminum oxide and especially to transform it to α-Al_2O_3 at lower temperatures [6,7]. Surface modification of Al particles could significantly increase the reactivity of Al powder. In particular Deng et al. [8] showed that modified γ-Al_2O_3 Al particles could continuously react with water and generate hydrogen at room temperature under atmospheric pressure. EDX analysis was used to investigate iron contamination of MA aluminum powder and IR spectroscopy was used to investigate the structure of superficial layer of Al particles ball milled in ethanol medium.

EDX analysis of the samples reveals that 1 h activated aluminum powder sample may contain up to 0.8 wt% of iron, and the content of iron in 5 h ball milled sample could reach up to 1.2%. Since the diffusion coefficients of atoms in Fe_2O_3 are 10 orders of magnitude higher than those for atoms in Al_2O_3, the iron content, especially in the form of Fe_2O_3, can play an important catalytic effect on the oxidation behavior of alumina [6]. Moreover, Fe_2O_3 has been reported to substantially lower the formation temperature of α-Al_2O_3 [6,7]. Similar observation was also made in our study.

IR spectra of Al powder ball milled in ethanol for 1 h and 5 h are presented in Fig. 3. Since Al has no absorption in IR field, the IR spectrum of pure Al does not contain any characteristic peak. To be infrared active the vibration must result in a change of dipole moment during the vibration, which in our case could be valid only for heteronuclear diatomic molecule or group contained in ethanol [9]. IR spectrum of ethanol (CH_3CH_2OH) has a CH stretch, OH stretch, CO stretch and various bending vibrations. The OH stretch appears as a broad band at approximately 3300–3500 cm^{-1}, likewise the CH stretch appears at about 3000 cm^{-1} [10,11]. In Fig. 3 (line 1), we can see that the characteristic peaks of both OH and CH groups are centered at about 3664 cm^{-1} and 2995 cm^{-1}. These peaks appeared in IR spectrum of Al powder after 1 h milling in ethanol and drying

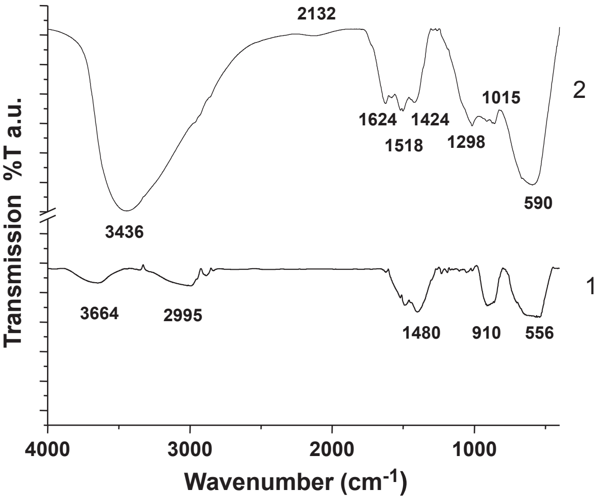

Fig. 3. IR spectrum of Al powder ball milled in ethanol medium. (1) spectrum of Al powder after 1 h of milling and (2) spectrum of Al powder after 5 h of milling.

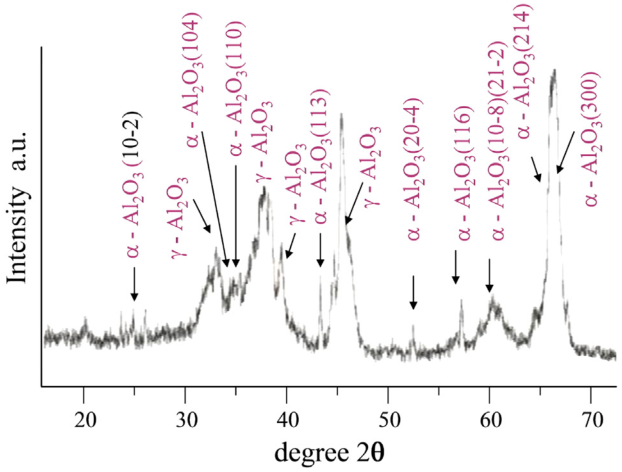

Fig. 4. XRD patterns of Al powder ball milled for 60 min in ethanol and annealed at 500 °C.

in vacuum. After ball milling, the samples have been dried in vacuum at 50 °C; however it still contains a significant amount of OH and CH functional groups. The spectrum of 1 h milled sample in the range 500–1550 cm^{-1} presents three wide patterns with maximum absorbance around 556, 910 and 1480 cm^{-1}. The broadness of the bands indicates the presence of mixture of different aluminum compounds along with some bonded ethanol. From the comparison, we can note that amorphous $AlO(OH)\cdot xH_2O$ is dominant in the mixture [12,13]. All 3 bands match the fundamental modes of vibration in a nonlinear AlO(OH) molecular structure [12]. The amount of the bonded groups was found to increase with increasing time of mechanical processing. From Fig. 3, it is clear that the amounts of bonded groups are significantly higher for the sample ball milled for 5 h (line 2) in comparison with the sample milled for 1 h (line 1). In the spectrum of 5 h milled sample, the bands at 556 cm^{-1} and 910 cm^{-1} overlapped and shifted to higher energy with maximum absorbance around 590 cm^{-1}, and shoulder at about 1298 cm^{-1}. The band at 1480 cm^{-1} also shifts to higher energy and splits to 3 bands with absorbance at 1424, 1518 and 1624 cm^{-1}. The spectrum of 5 h milled sample fits very well with the fundamental modes of vibration of boehmite, which has an amorphous surface structure existing in nanocrystals. It encapsulates the nanocrystals in an amorphous shell and has a thickness of about 2.2 nm [12]. The weak band at 2132 cm^{-1} on the spectrum is also a characteristic absorbance for such structure [12]. Based on the IR results, we can assume that ball milling transforms superficial layers of aluminum mainly to boehmite phase. The detail mechanism of modification of aluminum particles superficial layers to boehmite is beyond the scope of this article and will be the objective of our future investigations. However, ball milling induced ethanol decomposition and oxidation could be considered as a reasonable path for such modification. Since the water content in ethanol is only about 0.2%, the decomposition and oxidation seems the most probable process that may supply water into the system for supporting the continuous modification process. After decomposition, ethanol produces ethyl ether and water ($C_2H_5OH=(C_2H_5)_2O+H_2O$) [14] and it produces acetaldehyde and water ($2C_2H_5OH+O_2=2CH_3CHO+2H_2O$) on oxidizing [14]. Since water is one of the products of both reactions, it is reasonable to assume that the water needed for boehmite formation and continuous growth could be supplied by this mechanism.

A series XRD analysis had been performed to investigate the phase composition of MA Al powder after annealing the powder at 500 °C. XRD spectrum of 60 min ball milled and 30 min annealed in air at 500 °C powder is shown in Fig. 4. The XRD patterns show that all Al content has been transformed to α-Al_2O_3 and γ-Al_2O_3 during 30 min of annealing at 500 °C. It is also important to point out that part of activated Al has been oxidized directly into corundum form (α-Al_2O_3) despite the lower annealing temperature (500 °C). We suppose that boehmite-modified superficial layers of MA Al powder later play a role in low temperature formation of corundum via the $\gamma\text{-AlOOH}\rightarrow\gamma\text{-Al}_2\text{O}_3\rightarrow\alpha\text{-Al}_2\text{O}_3$ path [15]. The iron contamination of the sample plays the role of a seeding agent and provides an additional catalytic effect during oxidation at significantly lower temperatures [15]. Since our research shows that it is possible to oxidize and transform MA aluminum into corundum at such a low temperature, we explore this specific behavior of MA aluminum powder to prepare alumina based ceramic composites.

3.1. Composition, microstructure, hardness and density of the prepared Al–SiC composite

The unique property of mechanochemical activation and oxidization of the ball milled aluminum powder at relatively low temperature has been exploited to prepare Al_2O_3–SiC ceramic composites. Some preliminary results of XRD and SEM analysis along with density and hardness measurements of the prepared composites are listed below. Fig. 5 shows a series of XRD patterns describing the dynamics of changes occurring in phase composition of Al–SiC mixture ball milled in ethanol for 22 h and annealed up to 1200 °C. As we can see from the 1st graph of Fig. 5, the green mixture contains XRD characteristic peaks of only Al and SiC. The annealing of green mixture up to 800 °C (2nd graph in Fig. 5) changes the composition of the mixture drastically; thus instead of SiC and Al, we can see the mixture of SiC and Al_2O_3 on the graph, where the oxidized phase of aluminum represents the mixture of α-Al_2O_3 and γ-Al_2O_3. Further annealing of the mixture up to 1000 °C (Graph 3 in Fig. 5) transforms the entire alumina content of the composite to the corundum phase. It is important to note that the intensities of both SiC and corundum peaks decrease when the annealing temperature reaches 1200 °C (Graph 4 in Fig. 5) and a new diffraction peak appears in the graph at $2\theta = 33.174$. Based on XRD analysis, we assumed that the peak describes a new product phase—mullite. This assumption was based on the fact that the peak appeared well matched to the most intense peak (200) of mullite at $2\theta = 33.176°$ and the other characteristic peaks of mullite such as (210), (001) (201) and (331) were also are visible in the graph. This assumption is also supported by the research of Sakka et al. [16], where they synthesized SiC–mullite–Al_2O_3 nanocomposites by consolidating SiC and Al_2O_3 powders. Interestingly, Sakka et al. [16] first oxidized the surface of SiC particles and then synthesized mullite by reacting it with alumina.

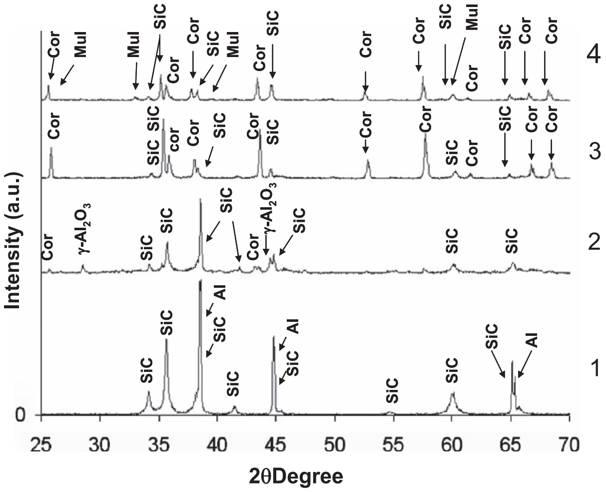

Fig. 5. XRD patterns of 22 h ball milled and consolidated at 40 MPa green mixture in an initial state and after 1 hour annealing at different temperatures: (1) green mixture; (2) green mixture annealed at 800 °C; (3) green mixture annealed at 1000 °C; and (4) green mixture annealed at 1200 °C.

Fig. 6a–c shows SEM images of the green mixture (Al–SiC), the mixture after annealing at 1000 °C (corundum–SiC) and annealing at 1200 °C (corundum–mullite–SiC). The green mixture (Fig. 6a) represents a uniform conglomerate of Al and SiC particles with particle size of about 1 μm and less, where hard SiC particles are covered by Al matrix. Annealing up to 1000 °C transforms the entire Al content of the mixture to the corundum phase as we saw from XRD analyses in Graph 3 of Fig. 5. However as we can see from Fig. 6b, the structure of the formed composite is still considerably porous with non-uniform surface. Contrary to this sample, the sample annealed at 1200 °C (Fig. 6c), shows much uniform surface structure with less porosity.

The value of hardness and density of some prepared composites are listed in Table 3. As seen from Table 3, both hardness and density of the composite increase as the temperature increases. The maximal density of the sample compacted by applying simple uniaxial pressure is about 71.5% of theoretical density (accepted as 3.55 g/cm^3 for Al_2O_3/SiC 50:50 composite) after 1200 °C annealing. However, the density of CIP compacted and annealed at 1200 °C sample is about 75.7% of the theoretical density. Hardness of the formed Al_2O_3/SiC composite after subsequent uniaxial pressing and annealing at 1200 °C reaches up to 11.50 GPa. The hardness of the same CIP compacted composite reaches up to 15 GPa after 1 h annealing at 1200 °C. By analyzing the results presented in Table 3 and the hardness data of mullite available from the book [17], we assume that the samples annealed at 1200 °C show a higher value of hardness due to mullite formation. Single crystal studies yield a microhardness of mullite near 15 GPa and the microhardness of polycrystalline mullite ceramics can vary considerably from about 10 to 14.5 GPa, which is in good agreement with our experimental data. A more detail analysis of the mechanisms leading to mechanical activation, as well as more effective conditions for preparing green mixture and alumina based composites will be the focus of our future research.

4. Conclusions

The process of chemical activation of Al powder induced by ball milling in ethanol medium was investigated and the

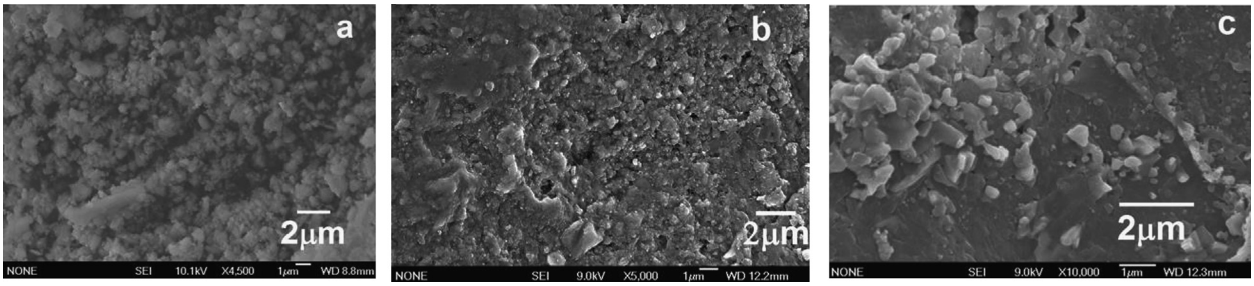

Fig. 6. SEM image of 22 h ball milled green mixture (non-compacted) and composites obtained from that mixture after consolidating at 40 MPa and annealing at different temperatures: (a) green mixture; (b) green mixture annealed at 1000 °C; and (c) green mixture annealed at 1200 °C.

Table 3
The density and hardness of the synthesized from ball milled Al–SiC green mixture composites after consolidating at 40 MPa and annealing at different temperatures.

Time of processing of Al–SiC mixture (h)	Compacting pressure (MPa)	Cold isostatic pressure (CIP) (MPa)	Annealing temperature (°C)	Density (g/cm^3)	Hardness HV (GPa)
22	40	–	800	1.98	2.4
22	40	–	900	2.2	2.8
22	40	–	1000	2.68	6.0
22	40	–	1100	2.32	7.3
22	40	–	1200	2.57	11.50
22	40	392	800	2.41	3.26
22	40	392	900	2.47	3.90
22	40	392	1000	2.5	9.20
22	40	392	1100	2.42	10.80
22	40	392	1200	2.72	14.80

specific chemical behavior of the activated powder was implemented for synthesis of alumina based ceramics. It was established that ball milling in ethanol medium significantly decreases oxidation temperature of Al powder (from 950 °C down to 450 °C and less) by modifying particles superficial layers, providing iron-based nucleation centers and by reducing particles sizes. Alumina–SiC ceramic composites were prepared directly from Al–SiC green mixture by using a specific oxidation behavior of Al powder activated in ethanol and by annealing the activated Al–SiC mixture in temperature range between 800 and 1200 °C. It was established that alumina in (Al_2O_3–SiC) composite prepared by this way appears mainly in corundum phase for 1000 °C annealing and in corundum/mullite mixture phase for 1200 °C annealing. The synthesized composite possesses high hardness (about 15 GPa) at relatively low sintering temperature of 1200 °C and exhibits excellent insulator property (electrical resistance $>$10 GOM at 1000 V DC). Due to low temperature of synthesis and good mechanical and insulator properties, we believe that the composites synthesized this way could find a variety of application in many production areas.

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References

[1] D.D. Radev, M. Marinov, V. Tumbalev, I. Radev, L. Konstantinov, Mechanically activated self-propagated high-temperature synthesis of nanometer-structured MgB_2, Physica C: Superconductivity 418 (2005) 53–58.

[2] L. Takacs, Self-sustaining reactions induced by ball milling, Progress in Materials Science 47 (2002) 355–414.

[3] L. Takacs, Metal–metal oxide systems for nanocomposite formation by reaction milling, Nanostructured Materials 2 (1993) 241–249.

[4] A.R. Torosyan, V.G. Martirosyan, S.S. Karakhanyan, Accumulation of mechanical energy and slow burning transition to detonation in the CuO–Al system, International Journal of Self-Propagating High-Temperature Synthesis 1 (1998) 87–90.

[5] A.R. Torosyan, V.S. Baghdasaryan, V.G. Martirosyan, S.G. Aloyan, H.G. Balayan, H.V. Navasardyan, S.A. Barseghyan, Diamond synthesis by mechanically induced detonation, Materials Science Fourm 386–388 (2002) 229–234.

[6] L. Baca, J. Lipka, I. Tóth, L. Pach, Study of crystallisation of Al_2O_3–Fe_2O_3 gels by mossbauer spectroscopy, Ceramics-Silikáty 45 (2001) 9–14.

[7] J.L. McArdle, G.L. Messing, Transformation and microstructure control in boehmite-derived alumina by ferric oxide seeding, Advanced Ceramic Materials 3.4 (1988) 387.

[8] Z. Deng, Y. Liu, Y. Tanaka, J. Ye, Y. Sakka, Modification of Al particle surfaces by γ-Al_2O_3 and its effect on the corrosion behavior of Al, Journal of the American Ceramic Society 88 (2005) 977–979.

[9] E.K. Plyler, Infrared spectra of methanol, ethanol, and n-propanol, Journal of Research of the National Bureau of Standards 48 (1952) 281–286.

[10] J. Datka, B. Gil, P. Baran, IR study of heterogeneity of OH groups in zeolite HY-splitting of OH and OD bands, Journal of Molecular Structure 645 (2003) 45–49.

[11] K.M. Murdoch, T.D. Ferris, J.C. Wright, T.C. Farrar, Infrared spectroscopy of ethanol clusters in ethanol–hexane binary solutions, Journal of Chemical Physics 116 (2002) 5717–5724.

[12] S. Ram, Infrared spectral study of molecular vibrations in amorphous, nanocrystalline and $AlO(OH)\cdot\alpha H_2O$ bulk crystals, Infrared Physics and Technology 42 (2001) 547–560.

[13] A.L. Delgado, L. Fillali, J.A. Jimenez, Synthesis of α-alumina from less common raw material, Journal of Sol–Gel Science and Technology 64 (2012) 162–169.

[14] Herbert E. Morris, Reactions of ethyl alcohol, Chemical Reviews 10 (1932) 465–506.

[15] Xuelian Du, Yanqin Wang, Xinghua Su, Jiangong Li, Influences of pH value on the microstructure and phase transformation of aluminum hydroxide, Powder Technology 192 (2009) 40–46.

[16] Y. Sakka, D.D. Bidinger, I.A. Aksay, Processing of silicon carbide–mullite–alumina nanocomposites, Journal of the American Ceramic Society 78 (1995) 479–486.

[17] H. Schneider, S. Komarneni, Mullite, Wiley-VCH, Germany, 2006.