

# The influence of $\text{Al}_2\text{O}_3$ +Al powders stirring time on the quality of alumina based ceramics

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Received: 01.09.2010. • Érkezett: 2010.09.01. <http://dx.doi.org/10.14382/epitoanyag-jsbcm.2010.21>

In present paper the influence of mixing time of two powders (alumina and aluminum) on the properties of alumina-based ceramics is studied. It is shown that upon stirring in an ultra low speed ball mill up to 150 hours the homogeneity of the mixture increases with a following decrease at longer stirring times. The optimal mixing time (around 140 hours) for the fabrication of dense (97%) and hard (21 GPa) alumina ceramics was found. The increase of microstructure homogeneity upon stirring was found as well.

Keywords: alumina, stirring, mixture, microstructure

## Introduction

Alumina has been used in production for several decades already, but in terms of advanced physicochemical properties this material is being of current importance for the research due to cheap raw materials and relatively simple synthesis technology. There is quite a number of works, dedicated to fabrication of pure alumina [1–3] as well as composite ceramics with the addition of magnesium, titanium [4], zirconium [5] oxides and so on. Among them the synthesis of  $\text{Al}_2\text{O}_3$ -based ceramics by dry powder technology appears to be one of the simplest and fastest methods. Moreover, the usage of alumina coupled with aluminum allows fabricating monolithic ceramics with high mechanical properties [6, 7].

It's well known that while using powder synthesis route one has to provide the homogeneous mixing of the starting powders in order to obtain advanced ceramics. The best mixtures of the components can be achieved at co-synthesis of the powders (coprecipitation, for instance). The main problem here is drying of the mixture. As it leads to a formation of hard particle aggregates, it becomes extremely complicated to fabricate dense ceramics [8]. According to this it's important to study the influence of the  $\text{Al}_2\text{O}_3$  and Al powders mixing conditions on the characteristics of the alumina ceramics.

## Experimental

In present study two types of powders were used: the commercial 99.8% pure alumina (Inframat Advanced Materials, USA) and passivated aluminum, obtained by the explosion of the wires method (Institute of Electrophysics, Ekaterinburg, Russia) [9]. According to SEM analysis alumina powder consisted of 150 nm particles, sealed to 40 mkm aggregates. These aggregates were destroyed by the ultrasonic treatment in isopropanol, leading to a formation of stable suspension in it. The aluminum powder consisted of 140 nm weakly aggregated particles having the «core-shell» structure (aluminum spheres were covered with 2–4 nm thick amorphous alumina layer).

According to the preliminary data [7] the mixture of the alumina powder with 15 wt.% of aluminum powder was picked as the initial one. For this purpose the suspensions of these

powders were mixed in isopropanol in ultralow speed mill with corundum balls for 260 hours. During stirring a number of sample probes were taken and analyzed.

The obtained mixtures were shaped into discs (10 mm in dia and 0.3–1 mm thick with 0.57–0.61 relative density) at uniaxial magnetic-pulsed press [10] at the pressures of 0.3–0.4 GPa in amplitude. After that the green bodies were sintered at 1550 °C for 30 minutes in air.

The density of the samples was measured by Archimede's method. SEM (Jeol Jem 2100) and AFM (Solver 47p) as well as X-ray (D8 Discover GADDS) were used to study the structure of the materials. The standard scanner (4800 dpi, color 8 bit) was used for estimating of the mixture homogeneity. The microhardness of the ceramics was studied by Vickers indentation method at 2 N load (Nanotest 600).

## Results and discussion

In *Figure 1*. the AFM images of the compacts, obtained from different mixtures are presented. The data are given at “mag-cos” (phase contrast) regime. Here one can clearly see large round-shaped blocks that are typical for the non-sintered polycrystalline solid materials. The blocks are separated by contrast boundaries. The variable brightness of the blocks surfaces (dark spots over the light surface), probably, reveals the heterogeneity of the material. The analysis of the AFM images showed that the size of the block monotonically changes upon mixing time (*Figure 2a*). At that the average block size, obtained at “height” (relief) regime increases, and at “mag-cos” regime – decreases. As “mag-cos” regime allows to point out the regions with different elastic characteristics, the observed change could be connected with the increase of the “discernibility” of the interphase boundaries upon stirring. This obviously indicates more uniform distribution of the mixture components.

The visual control of the color distribution showed that upon stirring the mixture becomes more homogeneous (the color peak width of the sample surface, *A*, is decreasing) (*Figure 2b*). At this stage the critical mixing time is revealed. In case of the mixing time more than 150 hours the heterogeneity of the

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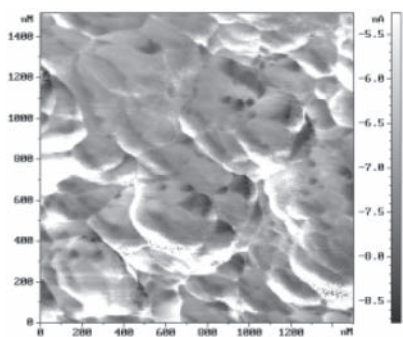
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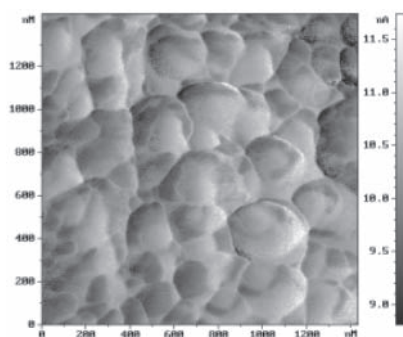
1991. His area of scientific interests is connected with sintering of oxide ceramics with ultrafine structure and investigation of its properties.

He'd graduated Urals State University at 1984. His PhD thesis was “Development and investigation of ceramics based on alumina zirconia and ceria nanopowders”. He is the author of over 50 scientific publications.

stock appears (the heterogeneity of the coloring increases on 20% with respect to the minimal value).



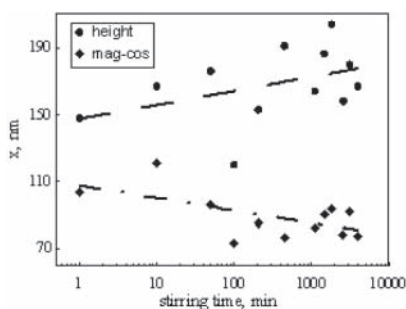
(a)



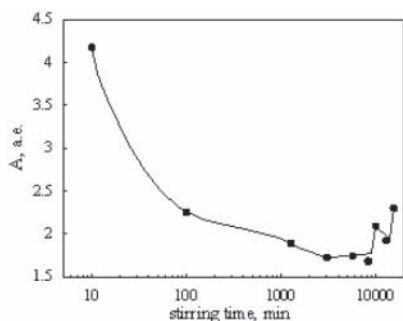
(b)

Fig. 1. AFM images of the surface of the compacts, obtained from  $\alpha$ - $\text{Al}_2\text{O}_3$  and Al mixtures after (a) 3 and (b) 43 hours of stirring

1. ábra Az  $\alpha$ - $\text{Al}_2\text{O}_3$ -Al porelegyből előállított tömör testek felületéről készített AFM felvételek (a) 3 óra keverési idő (b) 43 óra keverési idő



(c)

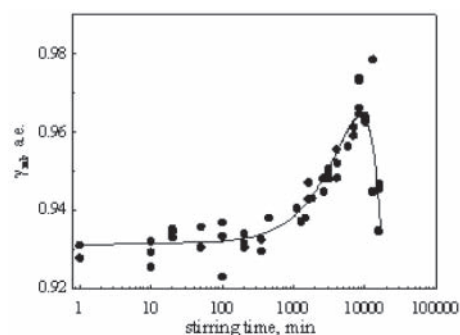


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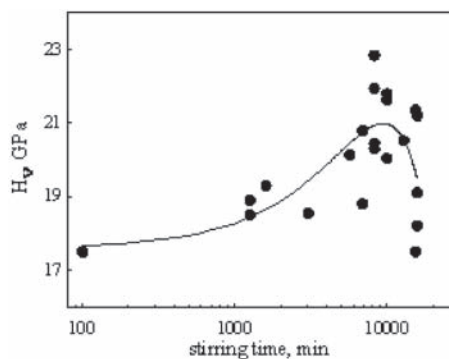
Fig. 2. The influence of stirring time on (a) average linear size,  $x$ , of AFM image and (b) color intensity peak width,  $A$ , of the compacts surface

2. ábra A keverési idő hatása (a) a tömör kerámia felületéről készített AFM felvétel átlagos lineáris ( $x$ ) méretére és (b) a színintenzitás csúcs szélességére ( $A$ )

The most effective method to estimate the quality of the obtained ultradispersed composite powder is to measure the density of the sample sintered from the specific powder. As seen at Figure 3a the density of the sintered ceramics is almost constant at the starting stage of the stirring (up to 20 hours). At longer mixing times the ceramics density significantly increases with the maximum at around 140 hours mixing time. Further mixing leads to the rapid decrease of the ceramics density. Such non-monotone behavior of the ceramics density, sintered from the composite powder, could be explained from the particle aggregation point of view. Probably, the destruction of the aggregates of the initial  $\alpha$ - $\text{Al}_2\text{O}_3$  powder leads to the improvement of the sintering process. Soft Al deforms and slows down the reaggregation of the alumina. At the same time the alumina particles prevent Al particles from sticking together. At the flex point the aggregation process starts to dominate. Perhaps, there are no surfaces free from of Al left and the particles, being “composite”, start conglomerating into new aggregates.



(a)



(b)

Fig. 3. The dependence of (a) relative density and (b) microhardness of the alumina ceramics from the stirring time

3. ábra Az alumínium-oxid kerámiák (a) relatív sűrűségének és (b) mikrokeménységének alakulása a keverési idő függvényében

The ceramic fracture images prove the observed dependence of the properties from the mixing time of the initial powders. It is clearly seen in Figure 4a that the structure consists of large 2  $\mu\text{m}$  grains together with extensive accumulation of small crystals that have faceting. It can be suggested that these crystals were formed due to homogeneous mixture of the powders where the metal component was in excess. The uniform structure is observed in the ceramics, which was sintered from the composite powder mixed for 133 hours (Figure 4b).

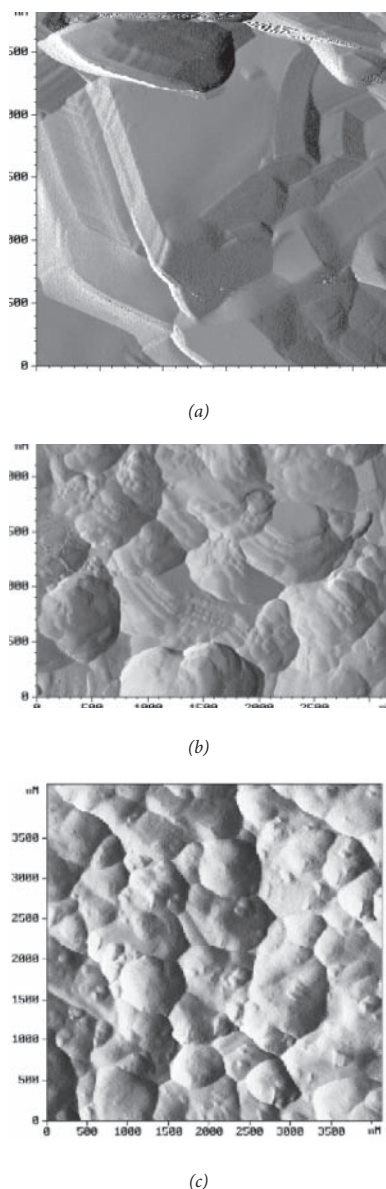


Fig. 4. AFM fracture images of the alumina ceramics, obtained from powder mixtures after (a) 6, (b) 133 and (c) 256 hours of stirring

4. ábra (a) 6 óráig, (b) 133 óráig és (c) 256 óráig kevert porelegyekből készített alumínium-oxid kerámiák törésfelületéről készített AFM felvételek

At this stage the density and the microhardness of the ceramics (Figure 2) is close to the maximal value (97% and 21 GPa respectively). The decrease of the ceramics quality upon maximal mixing times could be connected with the microstructure peculiarities presented in Figure 4c. The fracture surface, build up from submicron blocks is non-uniformly covered by the relatively small grains. It can be assumed that the observed grains were formed from the conglomerated particles. This distribution heterogeneity, the location and the size of these grains can decrease the stiffness of the intergranular boundaries, leading to the decrease of the mechanical properties of the ceramics.

## Conclusions

In present work the influence of stirring time of two powders (alumina and aluminum) on the properties of alumina-based

ceramics was investigated. It was shown that upon stirring in an ultra low speed ball mill up to 150 hours the homogeneity of the mixture increases with a following decrease at longer stirring times. This effect can be explained by the dominating role of aggregates at long stirring times. The optimal mixing time (around 140 hours) for the fabrication of dense (97%) and hard (21 GPa) alumina ceramics was found. The increase of microstructure homogeneity upon stirring was found as well.

## Acknowledgements

The work was supported by the Federal Principal Program “Scientific and Scientific-educational specialists of the Innovative Russia for 2009–2013”.

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### Az $Al_2O_3$ -Al porelegy keverési idejének hatása az alumínium-oxid kerámiák minőségére

Közleményünkben azt vizsgáljuk, hogy az  $Al_2O_3$ -Al porelegy keverési ideje miként befolyásolja a belőle készített alumínium-oxid kerámiák minőségét. Azt tapasztaltuk, hogy nagyon kis fordulatszámú golyósmalomban 150 óra keverési időig az elegy homogenitása nő, majd további őrlés során csökken. Tömör (97%) és kemény (21 GPa) előállításához mintegy 140 óráig kell az elegyet homogenizálni. A keverés során a mikroszerkezeti homogenitás is javult.

Kulcsszavak: alumínium-oxid, keverés, elegy, mikroszerkezet

Ref.: <http://dx.doi.org/10.14382/epitoanyag-jsbcm.2010.21>

Anton Kaygorodov – Vladimir Khrustov: *The influence of  $Al_2O_3$ +Al powders stirring time on the quality of alumina based ceramics*. Építőanyag, 62. évf. 4. szám (2010), 116–118. p.