

Investigation of Structural Hierarchy of Nanoceramics Compacted by Dry Pressing under Powerful Ultrasound Action

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Keywords: nanoceramics, zirconia, dry compacting, ultrasound action, AFM, microstructure.

Abstract. AFM techniques were used to investigate the microstructure, internal grain structure and intergrain interfaces of zirconia nanostructured ceramics sintered from green compacts being compacted by the common uniaxial dry compacting of nanopowders and under powerful ultrasound action. It was confirmed an essential influence of nanopowder compacting conditions on microstructure of sintered ceramics.

Introduction

Structural or functional nanocrystalline ceramics has improved mechanical and functional properties (fracture toughness, strength, hardness, thermal conductivity, specific electronic or magnetic properties) caused by the reduction of grain size to the nanometer range. However, the microstructure of nanoceramics have an impact on these properties [1].

It has been shown that among various techniques of nanoceramics production the method of the uniaxial dry compacting of initial nanopowders under powerful ultrasound (PU) action is effective to sinter homogeneously dense ceramics having nanostructured grains. Using SEM and XRD analysis we observed the structural-scale hierarchy and phase composition of zirconia nanoceramics ($\text{ZrO}_2 - 5.4\text{wt}\%\text{Y}_2\text{O}_3$) fabricated from the plasma chemically synthesized zirconia nanopowder compacted with PU-action and without it. PU-action at compacting leads to the decrease of grain sizes and grain aggregate sizes in sintered ceramics. Nanoscale grains with mean size of 300 nm in such ceramics are layered packs of subgrains (around 20 nm x 200 nm x 200 nm), with the scaly particles of initial nanostructured powder as their nuclei. The ceramics consist of cubic and tetragonal phases of zirconia [2].

The aim of this work is AFM investigation of microstructure of the zirconia ceramics prepared using the above compacting method at varying PU power ($W = 0, 1, 2, 3$ kW) and compaction pressure ($P = 50, 100, 150, 200$ MPa).

Experimental Methods and Specimens

Nanoceramics were sintered from green compacts of $\text{ZrO}_2 - 5\text{wt}\%\text{Y}_2\text{O}_3$ nanopowder, produced by plasma-chemical synthesis [2]. The powder particles had the shape of polycrystalline plates (scaly morphology) with the average linear size of 150 nm; the mean crystallite size in such a plate was 23 nm. The elemental composition of the powder under study was measured by means of the electron-probe microanalysis using the EPMA-1400 (Shimadzu Inc.) installation. According to the measurements the powder contained 74.13% of Zr, 18.16% of O, 4.28% of Y, and 3.43% of C.

The uniaxial dry compacting of green compacts 50 mm in diameter and with an aspect ratio of 0.05 was carried out under PU-action (PU-samples) and without it (O-samples) by the techniques described in [3,4]. Then compacts were sintered in air at $T_s = 1650^\circ\text{C}$ up to density of 88%.

The AFM measurements of the fracture surface of ceramics were performed in semi-contact mode using a scanning probe microscope P47-MDT (NT-MTD, Russia). We used the AFM studies in a mode of phase contrast (PC) images constructed by monitoring the phase shift of cantilever oscillation during the scan. The PC images contain information related to micromechanical properties of the sample materials (elastic modulus, viscoelasticity, or damping) and adhesion

generated between the probe tip and the sample surface (surface energy, capillary forces). Recent works conducted by several investigators showed that the phase contrast images were highly effective for estimating structural inhomogeneity of materials [5]. The chemical composition of the fracture surfaces was studied by techniques of XPS-analysis described in [6].

Results and Discussion

The microstructure of ceramics sintered from non-sonicated compacts has been determined to be different from that of the sonicated samples.

By AFM investigation of the O-ceramics ($W = 0$ kW), the samples compacted at $P=50$ MPa had aggregates close to $2\ \mu\text{m}$ involving grains of ~ 500 nm (Fig. 1a). At $P=100$ and 150 MPa scaly subgrain structure is formed with less subgrain sizes of $100\text{--}150$ nm (Fig. 1b). The scaly structure becomes less pronounced and grains grow in size with the increase of P up to 200 MPa (Fig. 1c). Apparently, particle deformation at compacting results in the fusion of scales.

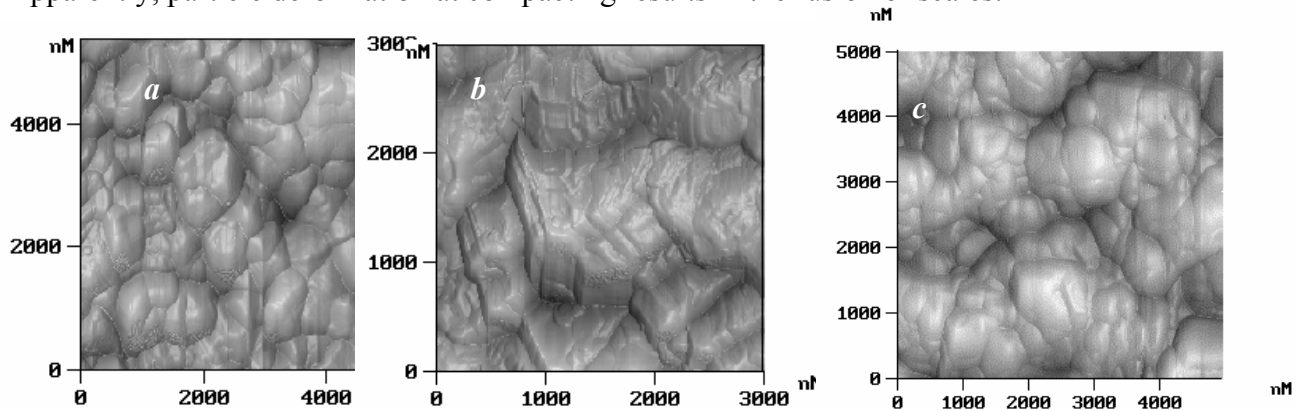


Fig. 1. AFM image for O-ceramics ($a - P=50$ MPa, $b - P=150$ MPa, $c - P=200$ MPa).

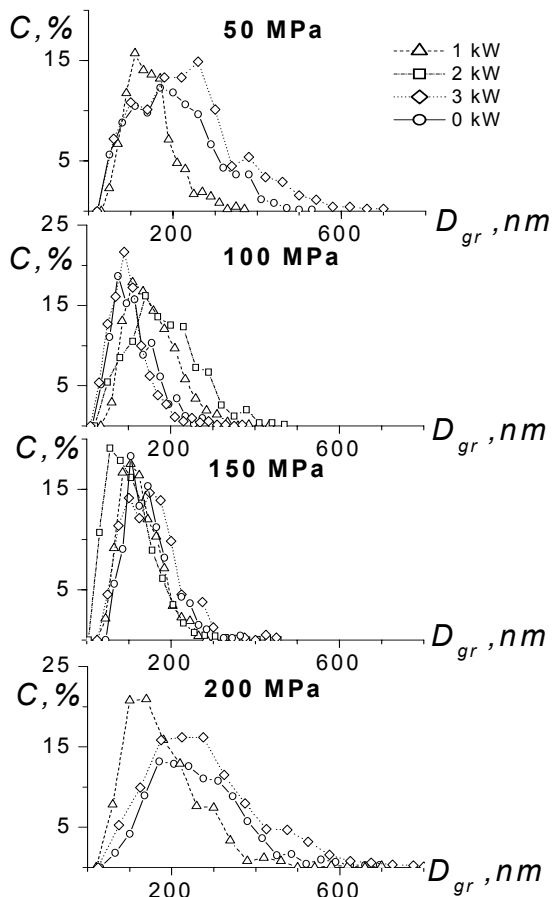


Fig. 2. Ceramics grain size histograms.

In support of above the histograms of grain size D_{gr} distribution in ceramic samples are shown in Fig. 2. They were calculated and plotted from AFM images using ImageJ 1.28 software.

Regarding the PU-ceramics, all samples preserve in grains the scaly structure of initial powder (Fig. 3), because of the sonication of green compacts enables a lower rate of plastic deformation of particles during densification. At the same time the particle structure varies due to a mechanochemical activation under PU vibrations.

Subgrains of $25\text{--}35$ nm detected in the AFM-phase contrast mode for PU-samples are likely to correspond to the regions of stress concentration, that emerge at the secondary recrystallization in sintering process of the samples. The size of the detected structure is similar to the grain size of the initial powder.

According to the AFM-data on size distribution of grains, the increase of W together with the increase in P sets a grain size limit, after which a reverse process – coagulation – becomes dominant. In this case grains can be as large as 1500 nm, which is comparable with the agglomerate sizes (Fig. 4a).

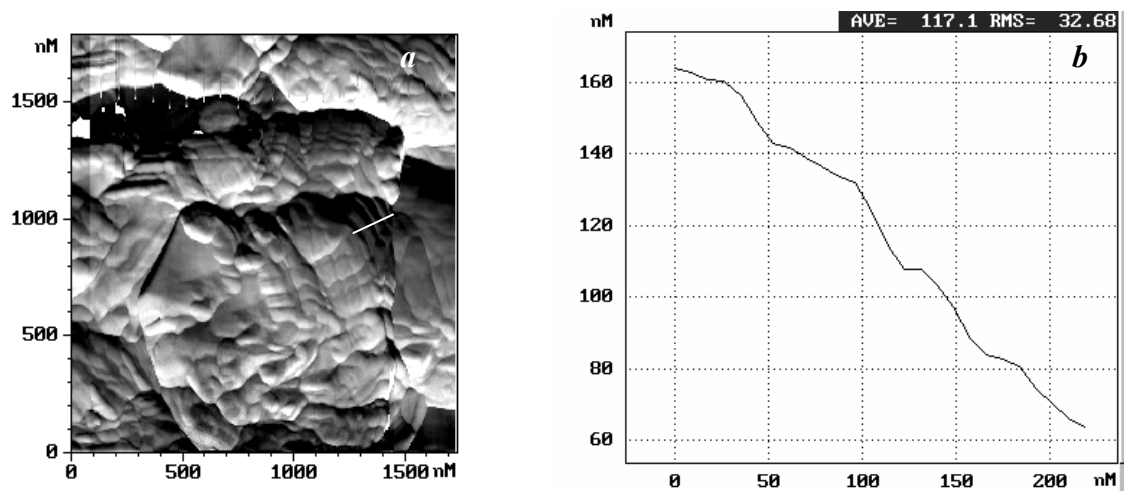


Fig. 3. AFM topography image for PU-ceramics (*a*; $W=1$ kW, $P=150$ MPa) and cross-section along white line showing the plate-scale subgrains with a thick of 20-25 nm (*b*).

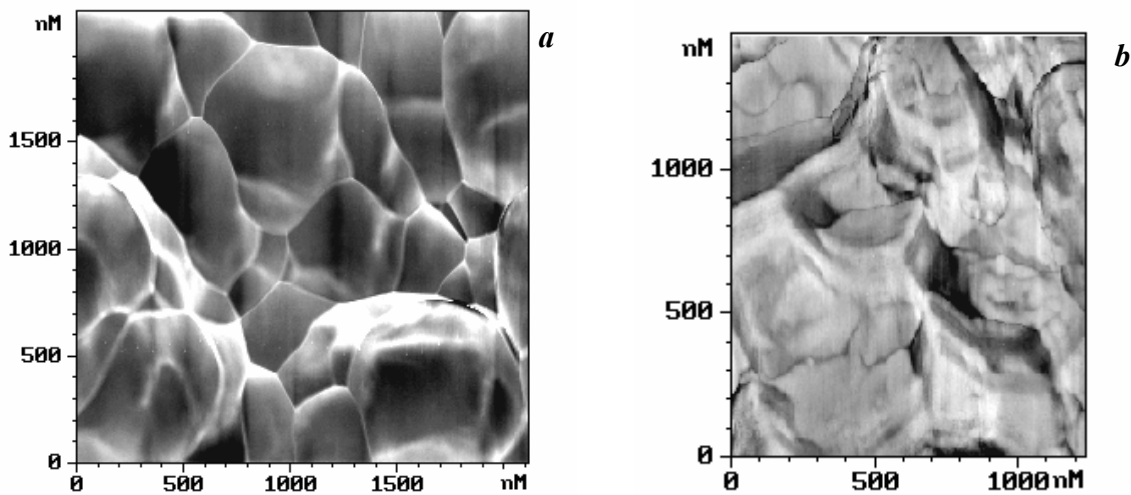


Fig. 4. AFM topography images for PU-ceramics compacted under $W=3$ kW ($P=200$ MPa - *a*; 150 MPa - *b*).

The fracture of O-ceramics in all the cases occurs by an intercrystalline mechanism, which points to the instability of grain boundaries. For PU-ceramics the transcrystalline mechanism of ceramics fracture is sometimes observed (Fig. 4*b*).

The rising of W up to 3 kW while powder compacting leads to an additional diffusion activation during sintering. It may cause element segregation (admixture, for example) at intergrain interfaces, which was observed (Fig. 4*a*). According to the XPS data the relation of Y and Zr content changed for the samples with the intercrystalline mechanism of breakdown. Content of Y is twice as much as in the initial powder and ceramics. Such a high-temperature diffusion of Y from a grain bulk to its surface [7] results in the appearance of an interfacial region of 4-6 nm in width. This region is different from the matrix in its elastic characteristics, and this is especially evident in ceramics compacted at $W=2$ kW, $P=150$ MPa; at $W=3$ kW, $P=100$ and 200 MPa due to the structural changes in particle surfaces under PU-action. The enrichment of these regions in Y brings forward a high local stiffness, as is evidenced by the AFM-data measured in the phase-contrast mode, and the stabilization of the interfaces.

Summary

1. Zirconia ceramics sintered from green compacts pressed by common uniaxial static pressing consists of agglomerates involving grains and subgrains, which sizes non-monotonically depend on compaction pressure. The fracture of such ceramics occurs by an intercrystalline mechanism, which points to the instability of grain boundaries.

2. Ceramics sintered from green compacts pressed under PU-action, preserve in grains the scaly structure of initial powder. For PU-ceramics the transcrystalline mechanism of ceramics fracture is sometimes observed.

3. Most narrow grain size distribution was observed at $P = 150$ MPa – optimal level to prevent intensive particle deformation under stress and grain growth during sintering.

4. PU activation of powder particles at compacting leads to an additional diffusion activation during sintering. It may cause impurity segregation at intergrain interfaces.

5. The perfection of the internal grain structure and the optimization of the grain interface structure using proposed method of nanopowder compacting - these are the ways to improve properties of nanoceramics.

Acknowledgements

The work is supported by the RFBR grants 01-02-96463; 01-03-32360; 03-02-16233.

The AFM investigations were performed using the equipment of the Centre of Joint Usage of Physical Technical Institute UrB of RAS (Research Centre for the investigation of the surface and nanosized systems).

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