1884
doi:10.1017/S1431927613011410
Microsc. Microanal. 19 (Suppl 2), 2013

Lead Zirconate Titanate – Zirconia Composites: Microstructural Evaluation of the Homogeneity Using the Voronoi-Diagram Approach

Andreja Benčan[1,2], Gregor Trefalt[1,2], Mitja Kamplet[1], Barbara Malič[1], Yohan Seo[3], Kyle. G. Webber[3]

[1.] Jožef Stefan Institute, Electronic Ceramics, Ljubljana, Slovenia.
[2.] Center of Excellence NAMASTE, Ljubljana, Slovenia.
[3.] Technische Universität Darmstadt, Institute of Materials Science, Darmstadt, Germany.

Ceramic piezoelectric materials based on the lead zirconate titanate solid solution [$(Pb,Zr)TiO_3$] are commercially important due to their excellent electromechanical properties. Unfortunately, as all ceramics, these materials are brittle, which presents severe limitations on some applications, e.g., diesel fuel injectors and bending actuators. [1] A possible way to toughen the originally brittle $(Pb,Zr)TiO_3$ ceramic is by introducing zirconia particles. In our previous work we investigated the effect of introducing tetragonal yttria stabilized zirconia (TZ) particles in soft $[Pb_{0.98}Ba_{0.01}][(Zr_{0.53}Ti_{0.47})_{0.98}Nb_{0.02}]O_3$ (PZT) ceramics. PZT- xTZ, (x = 0, 2, 5, 10, 20 vol%) composites were prepared by solid state synthesis (for details see [2]) and sintered at 1275^{o}C, 2h. We have shown that zirconium diffuses into the PZT, which resulted in a shift of the matrix phase composition towards the zirconia-rich rhombohedral phase. The addition of zirconia hinders the matrix grain growth and changes the fracture mode. Lower dielectric and piezoelectric properties of composites were related to the observed changes in phase composition and microstructure. [2]

Furthermore, we presented that the addition of zirconia leads to transformation toughening and reduced ferroelastic toughening of PZT–TZ composites.[3] It was found, however, that the second phase zirconia particles coalesced within the PZT matrix when using classical solid state synthesis, forming agglomerates that could affect the mechanical properties of the composites. In order to achieve a homogeneous distribution of TZ grains within the PZT matrix, a modified solid state synthesis procedure, which included pre-milling, pH adjustment and modification of the surface of the TZ powder, was used.

The aim of this work was to quantify the distribution of zirconia particles, and therefore the degree of homogeneity within the composites, using Voronoi-diagram analyses. In this study the analysis was performed as follows: (i) scanning electron micrographs (SEM) of the polished-and-etched PZT-TZ composites (Figure 1a) were transformed into binary images using the Image Tool program; (ii) the center of each zirconia grain was marked with a black point (Figure 1b); (iii) the distributions of these points were converted into a Voronoi diagram by the Image J program (Figure 1c). Each Voronoi polygon contains exactly one generating (TZ) point and every point in a given polygon is closer to its generating point than to any other. Finally, the area size distributions of Voronoi polygons which provide information about the regularity of the zirconia particles distribution in the PZT matrix were calculated. The narrower is the area distribution, the more homogeneous is the TZ distribution in PZT.
According to the Voronoi-diagram analysis of the SEM microstructures the modified solid-state synthesis, resulted in a more homogeneous TZ distribution in the PZT matrix (Figure 2a) compared to the non-modified composites (Figure 2b). The presence of large Voronoi area indicates some degree of clustered behavior in the distribution of zirconia particles in non-modified PZT-TZ composites.
At the end of the presentation the impact of particle homogeneity on the fracture behavior of PZT-TZ composites will be discussed.

References:

[1] CA Randall, A Kelnberger, GY Yang, RE Eitel and TR Shrout, J. Electroceram. **14** (2006), p. 177.
[2] A Bencan, B Malic, S Drnovsek, J Tellier, T Rojac, J Pavlic and D Damjanovic, J. Am. Ceram. Soc. **95** (2012), p. 651.
[3] J Rödel, Y Seo, A Bencan, B Malic, M Kosec and KG Webber, Eng. Fract. Mech (2012) in press.
[4] The work was initiated by Prof. Dr. Marija Kosec, who died on the 23rd December 2012. The authors would like to thank for the financial support of the Research Program "Electronic Ceramics, Nano, 2D and 3D Structures" P2-0105 (Slovenian Research Agency) and the European Union under the Seventh Framework project HIgh-PERformance Piezoelectric Actuators- HIPERAct. Authors thank to Ms. Jena Cilenšek and Mr. Silvo Drnovsek for technical support.

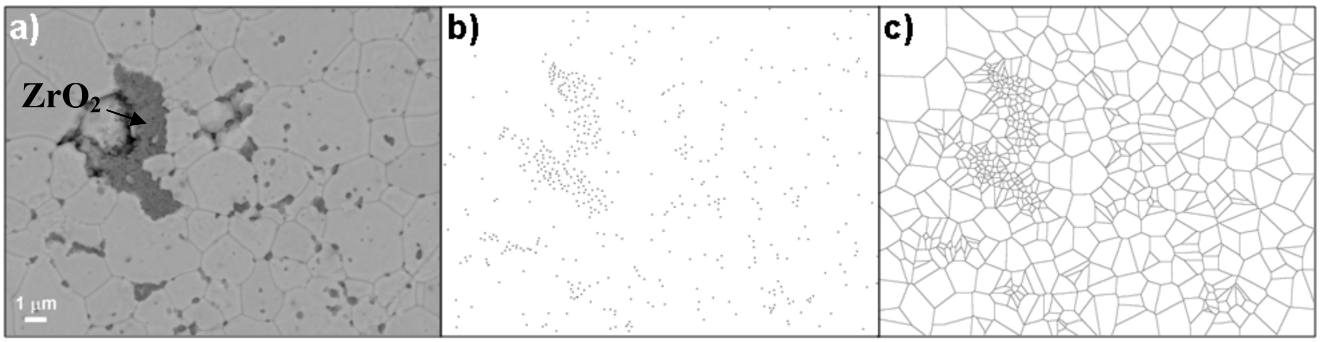

Figure 1. SEM-BE micrograph of the PZT-TZ, after sintering at 1275 °C, 2h (a), points representing the zirconia grains (b) and the Voronoi diagram representing the zirconia distribution (c).

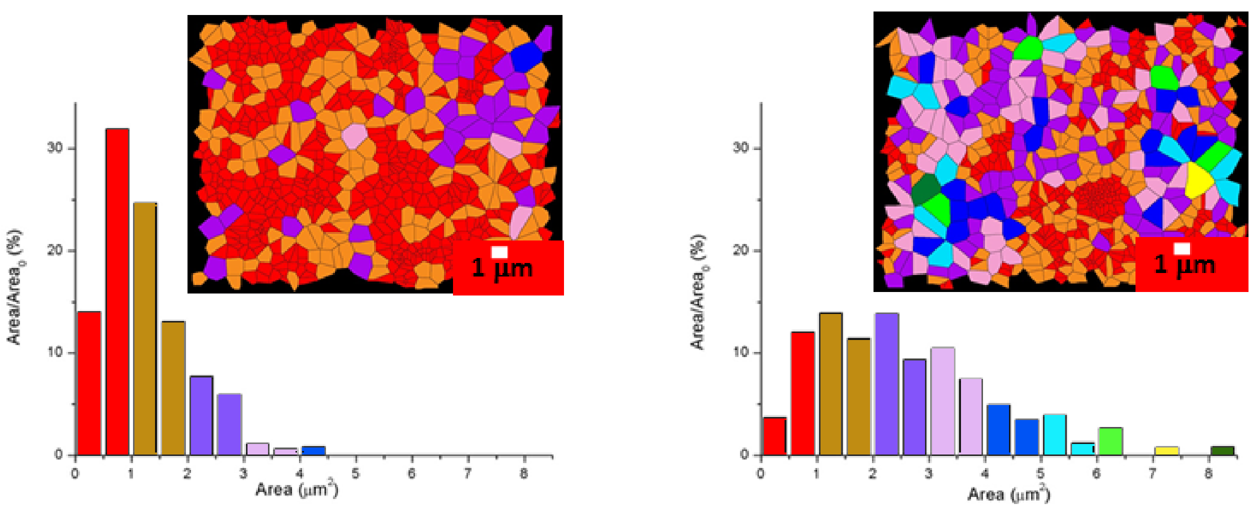

Figure 2. The area size distribution of the Voronoi polygons of PZT-TZ composites prepared by modified (a) and non-modified (b) syntheses. The insets in both figures present the Voronoi diagrams.

https://doi.org/10.1017/S1431927613011410 Published online by Cambridge University Press