doi:10.1017/S1431927616000374

Statistical analysis of grain size distributions in pressure-assisted $BaLa_4Ti_4O_{15}$ microstructures

Manuela A.C. Fernandes[1], João P. Bóia[1], Nivas B. Selvaraj[1], João C.C. Abrantes[2], Ana M.R. Senos[1] and Paula M. Vilarinho[1]

[1.] Department of Materials and Ceramic Engineering/CICECO, University of Aveiro, 3810-193 Aveiro, Portugal
[2.] UIDM, ESTG, Instituto Politécnico de Viana do Castelo, Apartado 574, 4901-908 Viana do Castelo, Portugal

High permittivity (ε_r), high quality factor (Q) and a near zero temperature coefficient of resonant frequency (τ_f) are the physical properties of $BaLa_4Ti_4O_{15}$ (BLT) that make it a suitable material for microwave applications in communications devices [1,2]. BLT presents a perovskite structure with the lattice parameters of the hexagonal cell a = 5.5671 Å and c = 22.4602 Å. Related to this structure, the tendency for anisotropic microstructures is high. Anisotropic grain growth can lead to anisotropic dielectric response.

On the other hand, sintering conditions, such as temperature, time, atmosphere and external pressure, are key parameters to control the microstructure. There are some few studies about the effect of the applied pressure in ceramics microstructure evidencing the influence of the applied pressure in the density and grain size of alumina [3,4] and the influence in the dielectric properties of copper titanate (CCTO) [5]. A previous work performed by our group in BLT shows that pressure-assisted sintering increases texturized grain growth with the development of abnormal grains [6]. For such a complex microstructure it is not trivial to find representative parameters of the grain size distribution and this work is focused on the statistical analysis of those distributions.

Conventional sintering, without any external pressure (CS), hot isostatic pressing (HIP) with 65 MPa and hot pressing (HP) with 60 MPa were used to design microstructures with different grain size distributions of BLT ceramics, having all the samples the same thermal history. Polished cross-sections were characterized by Scanning Electron Microscopy (SEM) and the micrographs analyzed using Image J. The grain size was evaluated by measuring the area of the elongated grains and the distributions were divided into two populations, normal (NGs) and abnormal grains (AGs, 6.25 times larger than the average area, following the Hillert criterion [7]) to be analyzed using statistical parameters as average, median and mode.

Figure 1 presents representative micrographs of BLT sintered with different pressure cycles and Table 1 the results of the statistical analysis of those microstructures. All the ceramics show elongated grains and the percentage of AGs is larger in samples sintered with external pressure, see Table 1. Generally, the calculated statistical parameters show equivalent variation for the different microstructures, i.e, a slight increase in size of NGs for samples with HP or HIP, whereas a huge increase for the size of AGs is observed. Despite the fact that the analysis with the tested statistical parameters leads to similar conclusion, we assumed as a more representative parameter of the grain size distribution the median, namely for the AG population. In fact, for the sample HP+HIP, with favored AG growth, the most frequent value for the mode calculation was not achieved due to the limited number of largest grains and the average value, on the right side of the distribution, is much larger than the median and the mode. Taking the median value as the most representative parameter, we can clearly conclude from the statistical analysis of the grain size distributions that the main effect of external pressure is on triggering AG growth, increasing both the number and size of AGs and leading to a more anisotropic grain growth.

References:

[1] T. Okawa, K. Kiuchi, H. Okabe and H. Ohsato, Ferroelectrics **272** (2002) p. 345.

[2] H. Yamada, T. Okawa, Y. Tohdo and H. Ohsato, J. Eur. Ceram. Soc. **26** (10-11) (2006) p. 2059.

[3] J. Besson and M. Abouaf, Acta Metall. Mater. **39** (10) (1991) p. 2225.

[4] J. Besson and M. Abouaf, Mater. Sci. Eng. A **109** (1989) p. 37.

[5] B.K. Kim, H.S. Lee, J.W. Lee, S.E. Lee and Y.S. Cho, J. Am. Ceram. Soc. **93** (9) (2010) p. 2419.

[6] N.B. Selvaraj, M. Fernandes, T. Denneulin, E. Snoeck, P.M. Vilarinho and A.M.R. Senos, Microsc. Microanal. **21** (S5) (2015) p. 47.

[7] M. Hillert, Acta Metall. **13** (1965) p. 227.

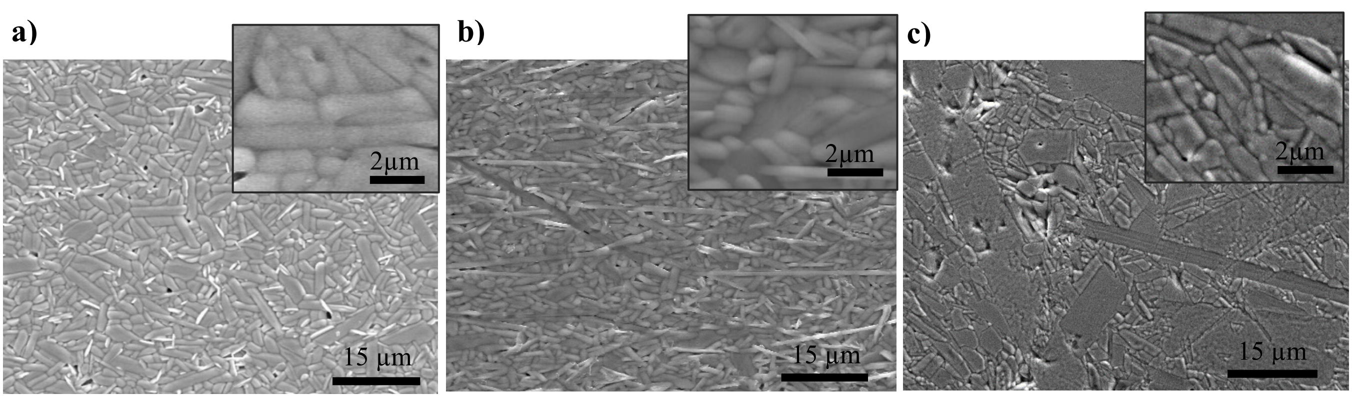

Figure 1. Microstructure of BLT samples without applied pressure, a) CS+CS, and with external applied pressure, b) HP+CS and c) HP+HIP.

Table 1. Statistical parameters for the area of normal and abnormal grains of CS+CS, HP+CS and HP+HIP samples.

	Normal grains (NGs)				Abnormal grains (AGs)			
	Average (μm^2)	Median (μm^2)	Mode (μm^2)	% (in number)	Average (μm^2)	Median (μm^2)	Mode (μm^2)	% (in number)
CS + CS	2.6	1.7	0.9	98	19	17	13	2
HP + CS	4.3	3.1	1.8	97	40	36	40	3
HP + HIP	3.9	2.2	2.3	95	200	73	n. d.	5

https://doi.org/10.1017/S1431927616000374 Published online by Cambridge University Press