HRTEM studies of stress assisted sintered $BaLa_4Ti_4O_{15}$

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The role of interfaces on the grain growth of polycrystals during sintering has been recently recalled into attention and important theoretical work has been developed, supported by experimental results on ceramic oxide materials [1,2]. It was shown that by controlling the nature of the interfaces one can tune the grain growth during sintering and, therefore, the subsequent control of related properties is envisaged. One way to control the interface nature is by changing the sintering conditions and there are many studies proving the importance of the sintering temperature, holding time, heating rate and atmosphere, but very few about the effect of an external pressure applied during the consolidation process [3, 4].
In this work we are presenting a HRTEM study of $BaLa_4Ti_4O_{15}$ (BLT) ceramics sintered under compressive stresses applied by Hot Pressing (HP) and Hot Isostatic Pressing (HIP) and the comparison with a free sintered sample. This study clearly reveals that the external pressure has a say on the nature of the grain boundaries in BLT.
BLT powders were prepared by conventional solid state route by calcining the stoichiometric proportion of the precursors at 1330°C for 3 h. Free sintered samples were prepared by uniaxial pressing of BLT powders at 100 MPa, followed by sintering at 1530°C for 180 min in conventional (CS) way. Second set of samples were prepared by HP at 1250°C, for 120 min, with an external pressure of 65 MPa. Another set of samples were prepared by HIP at 1530°C, for 90 min, under a pressure of 60 MPa, the previously HP samples. Cross section and top view Scanning Electron Microscopy (SEM) photographs (XL 30 FEG, Philips Electronic Instruments, Mahwah, NJ and S4100, Hitachi, Tokyo, Japan) were acquired for microstructure evaluation. Both set of samples were thinned down to 20 µm using precision ion polishing system (PIPS-Model 691; Gatan, Pleasanton, CA) for transmission electron microscopy (TEM) studies (SACTEM TECNAI-F20 and Hitachi HF3300 I2TEM).
SEM (Fig.1) revealed that the microstructure developments of CS samples are distinct from the stress assisted ones (HP and HP+HIP). In the stress assisted samples abnormal grain growth (AGG) was observed to have occurred during sintering, together with textured effects. Grain elongation was found to be enhanced in those samples, as well, by the effect of the external pressure applied during sintering. No second phases were observed in all the samples. HRTEM images (Fig. 2, 3) show clean grain boundaries for CS samples, while the stress assisted samples show crystal imperfections ie, line defects and surface defects in an unusual amount when compared to CS. Stacking faults were observed in both HP and HP+ HIP samples indicating that changes in distance between the atomic layers occurred due to the external pressure applied during sintering. The density of defects in the crystal structure was analyzed using geometrical phase analysis (GPA), which confirmed that CS samples are almost free of imperfections, while stress assisted samples are showing a quite large strain distribution.

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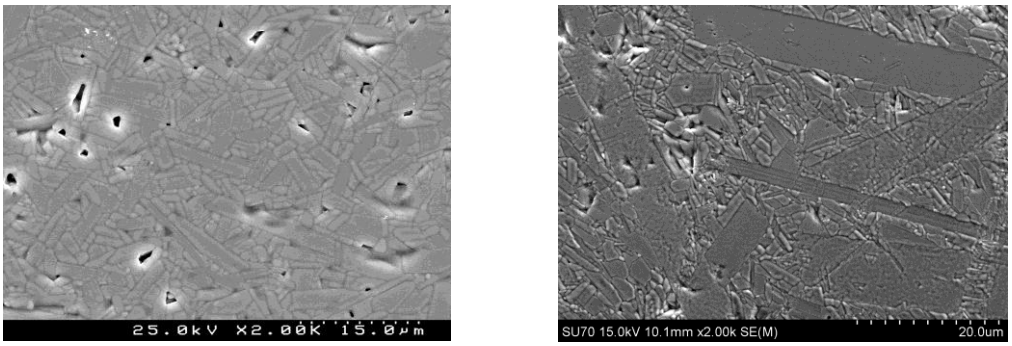

Figure 1. SEM image of CS (left) and HP+HIP sample (right).

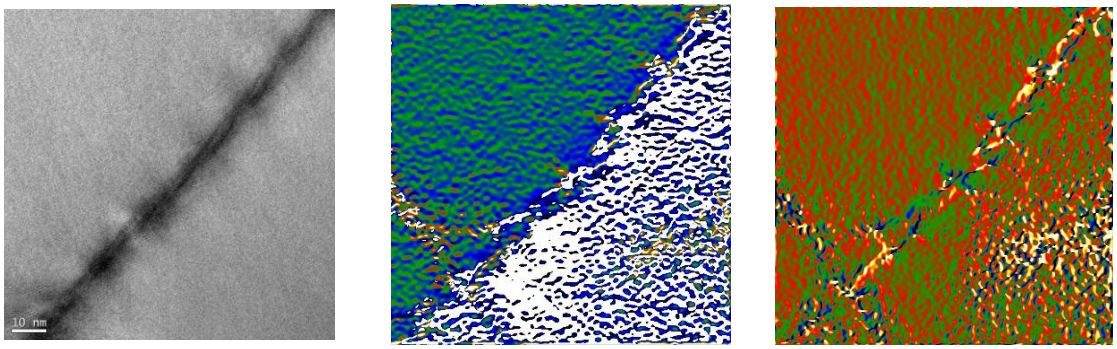

Figure 2. HRTEM image of CS sample (left), showing minimal fringe rotation (middle) and deformation (right).

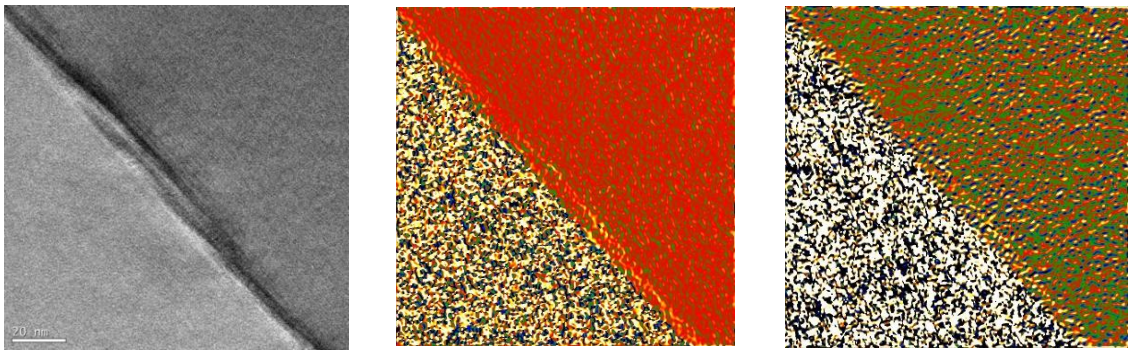

Figure 3. HRTEM image of HP sample (left), showing observable amount of fringe rotation (middle) and deformation (right).