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PII: S0955-2219(17)30512-5
DOI: http://dx.doi.org/10.1016/j.jeurceramsoc.2017.07.026
Reference: JECS 11383

To appear in: *Journal of the European Ceramic Society*

Received date: 23-5-2017
Revised date: 21-7-2017
Accepted date: 22-7-2017

Please cite this article as: Legallais M, Fourcade S, Chung U-C, Michau D, Maglione M, Mauvy F, Elissalde C.Fast re-oxidation kinetics and conduction pathway in Spark Plasma Sintered ferroelectric ceramics.*Journal of The European Ceramic Society* http://dx.doi.org/10.1016/j.jeurceramsoc.2017.07.026

Fast re-oxidation kinetics and conduction pathway in Spark Plasma Sintered ferroelectric ceramics

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Abstract
The re-oxidation kinetics of $BaTiO_3$ ceramics sintered by Spark Plasma Sintering (SPS) was investigated using in-situ impedance spectroscopy. Thanks to the flexibility of the SPS process, the grain size of the dense ceramics was tuned from 0.5µm to 10µm. The re-oxidation kinetics are found to be very fast regardless of the grain size and a full re-oxidation of the ceramics are achieved after 20 hours of exposure to an ambient environment at only 600°C. The residual density of charge carriers is reduced when using finer starting powders. SPS ceramics made with micrometer size grains demonstrate a residual charge-carrier density that is one tenth that of ceramics made from 10µm particles. Grain-boundary conduction is dominant through fine-grain SPS ceramics. This latter feature is similar to $BaTiO_3$ sintered using the conventional route with 10µm size grain. Finally, the critical grain size for optimal dielectric permittivity is found to shift from 0.7µm in standard ceramics to 1.5µm in SPS ceramics.

Keywords: $BaTiO_3$, SPS, dielectric properties, Impedance Spectroscopy, oxidation process

1. Introduction

The quest for improved functionalities in ferroelectric ceramics requires a control of composition, microstructure and defect chemistry. Ferroelectric materials which display the highest dielectric permittivities are very sensitive to defects because of the long scale correlation of their lattice features. As a result, a large density of chemical, structural or charged defects can strongly impact the bulk properties, artificially raising the permittivity and strongly increasing the dielectric losses [1, 2]. The sintering step is critical with respect to