Optical and dielectric properties of barium titanate ceramics densified by Spark Plasma Sintering.

Rosa Moronta Pérez1, Laurent Boilet1*, Philippe Aubry1, Véronique Lardot1, Luc Henrard2, Olivier Deparis2 and Francis Cambier1

1 Belgian Ceramic Research Centre, 4 avenue gouverneur Cornez, B7000 Mons, Belgium;
2 Physics Department, Namur University, 61, rue de Bruxelles, B5000, Namur, Belgium
*Corresponding author: l.boilet@bcrc.be

Highlights
- Study of the sintering behaviour of a barium titanate powder by Spark Plasma Sintering
- Translucent samples obtained by optimization of pre and post-treatments
- Highest permittivity values obtained for homogeneous microstructure and grain size ~ 1µm

1. Introduction
Barium titanate (BaTiO3) owing to its recognized dielectric, ferroelectric and piezoelectric properties, appears suitable for electro-optic devices and for substituting more toxic, lead-based electronic components as Pb(Zr,Ti)O3 (PZT) [1]. In this study, the possibility of obtaining translucent barium titanate ceramics by optimisation of SPS parameters and pre-and post-treatments was evaluated. The influence of sintering conditions, powder conditioning and air annealing on dielectric properties was also evaluated.

2. Methods
The barium titanate powder used in this study is a commercial nanometric powder (primary particle size around 50 nm) from Inframat Advanced Materials (purity higher than 99.9%). Powders were pre-compacted (30 MPa) into a graphite die of 20 mm inner diameter and consolidated by Spark Plasma Sintering (HPD10, FCT, Germany) under low vacuum (10 Pa). Powder deagglomeration was performed in isopropanol in a Turbula type T2F, with a ratio zirconia balls/suspension equivalent in volume. Densities of all samples were measured by the water immersion method. Permittivity measurements were performed with a HP4284A LCR meter (frequency range from 200 Hz to 100 kHz). The optical transmittance spectrum of the samples was measured within the wavelength region of 300-900 nm using a double-beam UV-visible spectrophotometer (Perkin-Elmer Lambda 750S).

3. Results and discussion
BaTiO3 ceramics with relative densities above 95% and different grain sizes have been prepared by Spark Plasma Sintering (SPS). Barium titanate powder appeared very sensitive to abnormal grain growth and the sintering window (temperature, pressure and dwell time) to obtain fully dense ceramics while preserving a fine microstructure was very narrow. This effect was ascribed to the high agglomeration degree of the powder, leading to differential sintering and abnormal grain growth. After deagglomeration, fully dense samples with fine microstructure were obtained with optimised SPS parameters. Adapted air annealing allowed removing the dark coloration of the samples and lead to translucent ceramics (Figure 1). Although real-in-line transmission values were very low, total forward transmission values were significant, around 35% at 600 nm, highest value observed to date for a barium titanate ceramic.
The influence of the microstructure and the density on electrical properties was also evaluated. Permittivity values were high, around 16000 in samples with full density, homogeneous microstructure and grain size close to 1 μm. The influence of grain size was investigated and a drastic drop in permittivity was observed when the microstructure presented grains above 5 μm.

Annealing in air led to a diminution of the permittivity but fortunately also reduced the dielectric loss. This drop in permittivity was already high at low annealing temperature (600°C) and it did not evolve more with the increase of the temperature. Consequently, translucent ceramics presented quite high values in the frequency range 0.2-100 kHz, with permittivity of 4000 and dielectric loss of 0.02.

4. Conclusions
Barium titanate ceramics with high permittivity values can be obtained by optimisation of powder conditioning and SPS parameters (mainly temperature and dwell time). Translucent samples elaborated in this work present the highest value of transparency obtained so far, with moderate permittivity values but low dielectric losses.

Acknowledgments
This study is supported by the FSE and the DGO6 of Wallonia region (Belgium) (Grant No. ECV320600FD017F/1217883).

References

Keywords
Barium Titanate ; Spark Plasma Sintering ; Dielectric properties ; Translucent ceramics.