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Dielectric properties of CaCu3Ti4O12-based ceramics with CuO-TiO2 phase deficiency

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$CaCu_3Ti_4O_{12}$ (CCTO) ceramics are characterized by presenting extraordinary dielectric constant values (k > 10,000), which is due to the presence of an insulating layer of CuO-TiO_2 phase segregated at the grain boundaries, according to the Internal Boundary Layer Capacitor (IBLC) model. To evaluate the influence of this phase on the structural, microstructural, and dielectric properties of these ceramics, compositions of $CaCu_xTi_yO_{12}$ (where 2.70 > x > 3.00 and 3.25 > y > 4.00 (CCxTyO) were synthesized by the coprecipitation method. Next, the calcination process was carried out in two steps: 623 K/1 h, and 1123 K/5 h. Between the first and second steps, the powder was macerated for 30 min in a mortar for its homogenation, in order to enhance the formation of the CCTO phase. After, the powders were uniaxially pressed at 180 MPa into shape discs with 12 mm in diameter and 1.5 mm in thickness. The sintering of the samples was performed at 1423 K/2 h in air. The apparent density was calculated for the sintered ceramics. The structural characterization and semiquantification of the phases present in the sintered ceramics were realized from diffractograms spectra refinement, based on the Rietveld method. The microstructural characterizations of the ceramics were performed by scanning electron microscopy (SEM) associated with energy dispersive spectroscopy (EDS), being possible to observe the evolution of the CuO-TiO_2 phase in the grain boundaries and its distribution on the CCTO ceramics microstructure. The evaluation of the dielectric properties was performed by impedance spectroscopy between 20 Hz and 5 MHz, at room temperature, to evaluate how the composition variation interfered in the values of the dielectric constant (k) and the dielectric dissipation factor (tand) of the ceramics. The CC2.90T3.75O ceramic presented the highest relative apparent density (94.6 % of the theoretical density), the largest grain size (D50 = 8.38 µm), the highest k values (27×10^3 at 1.0 kHz), and the lowest tand value of 0.063.