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Dielectric relaxation and charged domain walls in (K,Na)NbO_3-based ferroelectric ceramics

D.O. Alikin[1,2], A.A. Esin[1], A.P. Turygin[1], A.S. Abramov[1], J. Hreščak[3], J. Walker[4], T. Rojac[3], A. Bencan[3], B. Malic[3], A.L. Kholkin[1,2], V.Ya. Shur[1]

[1]*School of Natural Sciences and Mathematics, Ural Federal University, 620100, Ekaterinburg, Russia denis.alikin@urfu.ru*

[2]*Department of physics & CICECO, University of Aveiro, Portugal*

[3]*Electronic ceramic department, Jožef Stefan institute, Ljubljana, Slovenia*

[4]*Materials research institute, Pennsylvania state university, PA, USA*

We report on the evidence of significant contribution of charged domain walls to low frequency dielectric permittivity in KNN ferroelectric ceramics in the frequency range 10-10^6 Hz. The effect has been attributed to the Maxwell-Wagner-Sillars relaxation.

The piezoelectric device market is dominated by lead containing $Pb(Zr_{1-x},Ti_x)O_3$ (PZT) based materials due to their versatility and robust functional properties. The toxicity of lead, however, has raised health and environmental concerns and in the last two decades legislative changes have stimulated intensive research into suitable lead-free PZT alternative materials [1]. Among the numerous investigated lead-free oxides and solid solutions, $K_{1-x}Na_xNbO_3$ (KNN) based systems have received enormous attention after publications by Saito et al. in 2004, who reported piezoelectric constants for KNN comparable to PZT [2,3].

Despite the significant focus on KNN-based ceramics and the subsequent wave of scientific publications, commercial realization of its piezoelectric properties has not been forthcoming due to difficult process of the material synthesis in the bulk ceramic form, low bulk density, irreproducibility of electric properties, essentially smaller in comparison with PZT piezoelectric coefficient [3]. One of the strategy have been used to overcome this problem is doping by Sr^{2+}. Together with increasing of the ceramics relative density up to 96% and improvement of its functional response dielectric permittivity shows a non-monotonic dependence with great increase (above 3 times) within 0.5-3% doping interval and further sharp decrease in high degrees of doping (6-15 %) [4,5].

In this contribution, we studied relation between dielectric relaxation, grain and domain structure in Sr^{2+} doped KNN ceramics. The influence of the domain walls to the macroscopic properties of ferroelectric materials is known for a long time. Oscillations of the domain walls in the AC electric field are considered as a mechanism for additional dielectric and piezoelectric responses [6]. However, separation of the vibrational and stationary contributions of the domain walls to dielectric permittivity is still under discussion. As for another lead-free ceramic material, $BiFeO_3$, it was shown that the domain walls in it could influence the macroscopic properties via nonlinear Maxwell-Wagner mechanism [7] due to their conductivity.

We report on the evidence of significant contribution of charged domain walls to low frequency dielectric permittivity in KNN ferroelectric ceramics in the frequency range 10-10^6 Hz (Figure 1). We have found an apparent relationship between the density of charged domain walls, which are expected to exhibit enhanced conductivity, and the increase of the dielectric permittivity at low frequencies. We showed that the effective dielectric response increases with increasing domain wall density. Theoretical consideration in frame of Maxwell-Wagner-Sillars model postulates that conductive inclusions in the ferroelectric media with conductivity only three orders higher the bulk one could have great impact on resulting dielectric permittivity and losses. The results thus suggest a possible role of charged domain walls in the dielectric dispersion through Maxwell-Wagner mechanism. Thus, we identified an interesting possibility to modify macroscopic electromechanical properties of ferroelectric ceramics using approaches of the domain wall engineering.

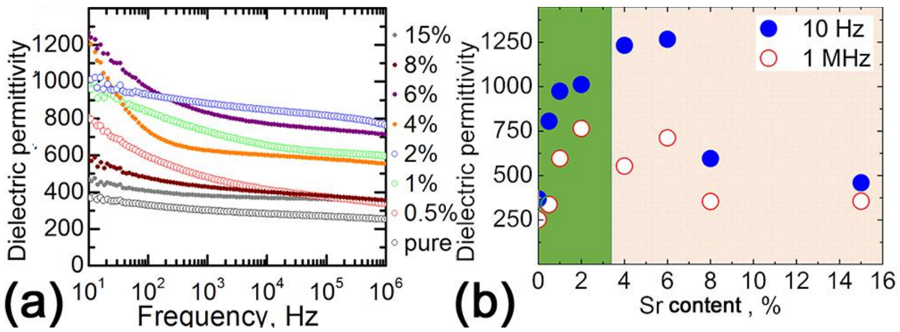

Figure 1. Dielectric properties of the Sr^{2+} doped KNN ceramics. (a) Frequency dependence of dielectric permittivity KNN ceramics with different compositions (imaginary part is presented in supplementary), (b) dependence of dielectric permittivity on Sr^{2+} content. Orange region marks the range of compositions with “single grain – single domain region”.

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