Observation of Positive and Negative Magnetodielectric Effects in Relaxor $PbCo_{1/3}Nb_{2/3}O_3$ Ceramic

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Lead cobalt niobate ($PbCo_{1/3}Nb_{2/3}O_3$: PCN) is a multifunctional electroceramic belongs to the class of relaxor ferroelectrics which are very popular due to their extraordinary properties and applications. To the best of our knowledge magnetoedielectric study of PCN ceramic has not been studied. With this motivation we performed magnetodielectric measurement on PCN ceramic synthesized by two step columbite precursor method using high purity chemicals: PbO (99.9%), CoO(99.99%), Nb_2O_5(99.9%). The room temperature crystal structure and microstructure is examined using x-ray diffraction (XRD) and scanning electron microscope (SEM) respectively and dielectric constant as a function of frequency as well as temperature is measured in 0T and 9T magnetic field using high performance frequency analyzer (NOVO-CONTROL). Microstructure images revealed closely packed grains with grain size ~8-10 μm. Room temperature X-ray diffraction (XRD) pattern suggests single phase pseudo cubic crystal structure having Pm3m symmetry, where Co and Nb randomly sitting at B-site with lattice constant ~4.0496(2) Å. Rietveld Refinement on XRD data yields larger value of thermal parameters, implying Pb and O are disordered along <111> and <110> directions respectively. The temperature dependent dielectric properties revealed re-entrant relaxor behaviour (T_m ~130 K and 210 K for 1 kHz) along with a high temperature diffused phase transition, T_c ~270 K. The low temperature frequency dependent dielectric maximum, $f(T_m)$ is analyzed using generalized glass model (also called critical slowing down model). The magnetodielectric effect (MDE(%)=(ε'(9T)-ε'(0T))/ε'(0T)*100) depicts positive and negative MDEs implying coupling between mangnetic and dielectric properties. Such positive and negative MDEs are also observed in disordred double perovskite Pr_2CoMnO_6 ceramic whereas single MDE peak is reported in B-site ordered phase. Therefore, it is believed that the re-entrant relaxor behaviour and multiple positive-negative megnetodielectric effects are associated with the quenched disorder as well as multiple heterogeneities present in PCN ceramics and details will be discussed.