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Optimisation of $SrBi_2(Nb,Ta)_2O_9$ Aurivillius phase for lead-free electrocaloric cooling

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Abstract: The influence of different substitutional mechanisms on the electrocaloric effect of a lead-free $SrBi_2(Nb_{0.2}Ta_{0.8})_2O_9$ Aurivillius phase was studied for application in electrocaloric cooling systems. The A-site substitution with barium efficiently reduced the temperature of maximum permittivity from about 300°C to 100°C. The A-site substitution induced phenomena that are typical of strong relaxor ferroelectrics such as significant broadening of the permittivity peak and an increase in its frequency dispersion and with a depolarization temperature below room temperature. These features directly influenced the electrocaloric effect. A direct measurement system, based on a modified-differential scanning calorimeter, was used to analyze the EC effect of the dense $(Sr_{0.5}Ba_{0.5})Bi_2(Nb_{0.2}Ta_{0.8})_2O_9$ ceramics. In accordance with the relaxor characteristics, the EC effect was found to increase continuously over a broad temperature range above the room temperature. This was attributed to the alignment of field induced polar nanodomains. Directions for optimization towards a high-performing EC ceramic were identified.
Keywords: Aurivillius phases; lead-free ceramics; ferroelectric relaxor; electrocaloric materials

1. INTRODUCTION

Among lead-free electrocaloric (EC) materials Aurivillius phases have been recognized as promising candidates for mid- and large-scale EC refrigeration [1] due to very high values of dielectric strength. It was reported to be around 280 MV/m for $SrBi_2Ta_2O_9$.[2] The dielectric strength has been recognized as one of the key properties that enable achieving the high EC effects.[3] For this reason, thin films that can sustain much higher fields than bulk, give much higher EC effects. However, their application potential is limited due to a very low heat capacity and, consequently, a low cooling power. In addition, the Aurivillius phases exhibit a low leakage current and a high resistance to fatigue. [4,5] This is important for the refrigeration devices where high alternating electric fields are expected to be applied for as much as 10^9 cycles.

To exploit the benefits of the Aurivillius phases for the EC refrigeration their ferroelectric transition should be moved from as high as 400°C down towards or even below room