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Low-temperature fabrication of $SrTiO_3$-based core/shell nanostructure for high performance organic/inorganic composite dielectrics

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[Introduction]

Dielectric capacitors are always considered a key component in recent electronic devices, electric vehicles, and high-power system for controlling their dielectric characteristics and storing charge. Among available dielectric materials, organic polymers with high breakdown strength and inorganic ceramics with high dielectric constant are regarded as representative materials. This highlights that ceramic/polymer nanocomposite dielectrics can achieve high dielectric properties and withstand voltage toward future capacitors if both materials are properly utilized.

To overcome above conventional issues, a new Nb-$SrTiO_3$/Mn-$SrTiO_3$ (Nb-@Mn-STO) core/shell nanoparticle, where donor and acceptor are doped in ceramics, respectively, was designed and synthesized by hetero-coagulation technique at room temperature to prevent dopant diffusion between core and shell. In other words, core generates high dielectric constant by donor-induced interfacial polarization, whereas shell can effectively block the movement of charge carriers, eventually contributing to high dielectric performances. The effect of core/shell nanoparticles on dielectric performances in polymer film was evaluated by comparing various ceramic/polymer composite samples.

[Results & Discussion]

Nb-@Mn-STO core-shell nanostructures were basically fabricated by hetero-coagulation approach at room temperature to thoroughly prevent dopant interdiffusion between core and shell during synthesis. Hetero-coagulated core-shell nanostructure is achieved mainly by (1) coagulation of plenty of small-sized shells onto large-sized cores with (2) different surface charge and thus making raspberry-like core-shell structure. Thus, for huge gap in particle sizes,

a low temperature synthesis method was utilized for Nb- and Mn-doped $SrTiO_3$ particles with a wide range of particle sizes that is conducted at temperatures close to room temperature. The cation-doped $SrTiO_3$ particles were synthesized under diverse solution conditions using facile and low-cost methods, leading to huge gap in particle size between core and shell particles. After that, the dielectric performances of Mn-doped $SrTiO_3$ nanoparticles are optimized by demonstrating the influence of post-annealing temperature on their phases transitions as well as oxidation states of transition metal Mn. Especially, the best annealing condition for high breakdown strength was discovered by carefully investigating dielectric performances with ceramic/polymer composite films. On the basis of above results, a new class of Nb-@Mn-STO core-shell nanostructures was fabricated via hetero-coagulation technique at room temperature. A series of experiments were conducted by investigating various synthesis conditions, such as huge gap in both particle sizes, appropriate amount of shell particles, repetitive centrifugation with low speed, and control of solution pH, which make high coverage at approximately 80%. Finally, using Nb-@Mn-STO particles, the dielectric properties and breakdown strength were evaluated after fabricating inorganic/organic composite films. Compared to pure polymer film and comparative ceramic/polymer composite films, the incorporation of Nb-@Mn-STO core-shell nanostructures improve the major dielectric performances and thus makes high energy density.

[Reference]

- M.Y. Cho, S. Yasuhara, T. Tsurumi, T. Hoshina*, Low temperature synthesis of Nb-doped and Mn-doped SrTiO3 with easily controllable particle size. *Journal of Crystal Growth*, 626 (2024) 127485.