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Title(English)	Low-temperature fabrication of SrTiO3-based core/shell nanostructure for high performance organic/inorganic composite dielectrics
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Category(English)	Doctoral Thesis
種別(和文)	論文要旨
Type(English)	Summary

（博士課程）
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論 文 要 旨

THESIS SUMMARY

系・コース： Department of, Graduate major in	材料 系 材料 コース	申請学位（専攻分野）： Academic Degree Requested	博士 (Engineering) Doctor of
学生氏名： Student's Name	Myung-Yeon Cho	審査員主査： Chief Examiner	保科　拓也

要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

This thesis is entitled "Low-temperature fabrication of $SrTiO_3$-based core/shell nanostructure for high performance organic/inorganic composite dielectrics" consisted of 6 chapters.

Chapter 1 is the introduction of this study, which mentioned the importance of developing a new ceramic core-shell structure for application to high performance (i.e., high dielectric constant and high breakdown strength) ceramic/polymer composite film. Till date, numerous efforts have been made for high dielectric performances via ceramic/polymer composite dielectrics, but the most chronic issue in this composite is considered a paradox between dielectric constant and breakdown strength because local electric field is always concentrated at the interface between polymer and ceramic under high voltage environment due to their difference in conductivity. To solve this conventional issue, the interface can be significantly improved through core-shell ceramic structure with both high dielectric constant and high breakdown strength. For this purpose, a new core-shell ceramic structure where Nb-doped $SrTiO_3$ (Nb-STO) and Mn-doped $SrTiO_3$ (Mn-STO) are utilized as core and shell, respectively, was designed in this thesis. This structure can solve the paradox in ceramic/polymer composite films because donor (Nb)-doped STO core induces high dielectric constant by interfacial polarization, while acceptor (Mn)-doped STO shell makes high breakdown strength by trapping the charge carriers through oxygen vacancies, eventually enabling to improve both dielectric performances in ceramic/polymer nanocomposite dielectrics. Especially, this core-shell structure was fabricated by hetero-coagulation technique at room temperature to thoroughly prevent dopant interdiffusion between core and shell during synthesis. This method is generally achieved 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.

In Chapter 2 entitled "Low temperature synthesis of Nb-doped and Mn-doped $SrTiO_3$ with easily controllable particle size", large-sized Nb-STO core and small-sized Mn-STO shell particles were synthesized under liquid-phase system at low temperature below 40℃. Both particles were synthesized under diverse solution conditions using facile and low-cost methods. The particle sizes were simply controlled by changing the ratio of H_2O/ethanol solvents and their difference in viscosity, two-step aging processes, and the alkali concentration in aqueous solution. Consequently, a wide range of particle sizes (25 to 600 nm) was obtained with a facile method even under low temperature environment. Finally, 600 nm is selected as a size of core particle, while 25 nm is selected as a size of shell particle.

In Chapter 3 entitled "Thermal annealing-induced oxidation state transition of Mn in Mn-doped $SrTiO_3$", the effect of post-annealing temperature on Mn-STO powder was investigated by disclosing the origin of change in powder color with annealing temperature and its impact on dielectric performances. The annealing process caused the color change by oxidation state transition of Mn, but it also accompanied decrease in H_2O and OH^- onto the particle surface as well as phase transition. Therefore, the dielectric performance was evaluated to determine which annealing temperature leads to the

optimal dielectric performances as a role of shell in target core-shell structure. Consequently, the 300°C-annealed Mn-STO powders provided the most desirable performances such as medium dielectric constant, low dielectric loss, and high breakdown strength in PVDF polymer. This outcome is achieved through charge trapping effect of oxygen vacancies and effective elimination of H_2O and OH^- onto the particle surface by optimizing post-annealing temperature of Mn-STO powder.

In Chapter 4 entitled "Room temperature fabrication of Nb-$SrTiO_3$/Mn-$SrTiO_3$ core/shell nanostructure via hetero-coagulation technique", a new class of Nb-@Mn-STO core-shell nanostructures was fabricated via hetero-coagulation technique at room temperature to completely prevent atomic diffusion between core and shell. On the basis of hetero-coagulation theory, the starting materials were selected as a 600 nm-sized Nb-STO core and 25 nm-sized Mn-STO particles obtained in Chapters 2 and 3, respectively. The shell coverage onto core was gradually improved by 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. Consequently, based on a series of optimization processes, Nb-STO core particles were well-encapsulated by Mn-STO shell particles, showing a high coverage at approximately 80%.

In Chapter 5 entitled "High performance dielectric capacitor with organic/inorganic composite films comprised of Nb-$SrTiO_3$/Mn-$SrTiO_3$ core/shell nanostructure", using Nb-@Mn-STO particles, the dielectric properties and breakdown strength are evaluated after fabricating inorganic/organic composite films. When the dielectric constant of various comparative samples was compared, Nb-@Mn-STO/PVDF composite had significantly high dielectric constant mainly due to donor-derived interfacial polarization. Moreover, its breakdown strength was highly enhanced, accompanying low dielectric loss, through well-wrapped shell particles onto core. Consequently, the incorporation of Nb-@Mn-STO core-shell nanostructures improve the major dielectric performances and thus makes high energy density.

In chapter 6 "Conclusion", a series of research processes and their outcomes obtained from previous chapters were summarized. This thesis aimed to fabricate and evaluate high performance ceramic/polymer nanocomposite dielectrics by a design of a new Nb-@Mn-STO core-shell ceramic structure, unique hetero-coagulation approach, and evaluation of material/dielectric properties. This new approach suggested in this dissertation provides various design paradigm and synthesis approach for ceramic/polymer nanocomposites with high dielectric performances.