論　文　の　要　旨

題目 SYNTHESIS AND CHARACTERIZATION OF HOLLOW AND POROUS SILICA FINE PARTICLES VIA ORGANIC TEMPLATE ASSISTED WET-CHEMICAL PROCESS

（有機テンプレートを用いた液相法による中空、ポーラスシリカ微粒子の合成と特性評価）

氏　名　LUSI ERNAWATI

Silica nanoparticles are promising applications in many current and emerging areas of technology because of their nature advantages. The synthesis, properties and applications of silica particle have become a quickly expanding field of research. Nowadays, special attention has been paid to synthesis, control morphology and characterization of hollow and porous silica nanostructures in micron and/or submicron sized. This dissertation is focused on the development of synthesis and nanostructurization of hollow and porous silica particles using tetramethyl-orthosilicate (TMOS) instead of tetraethyl-orthosilicate (TEOS) as silica source through organic-template assisted wet-chemical process. This work is concerned in the development of synthetic route to prepare hollow and porous silica based on polystyrene as template particle. Being the application of silica the most important point to consider, was to improve synthetic methods through wet-chemical process. In this term, the most important point of this work was not to find the formation mechanism of materials. This aim drove the work through different procedures, different reactants, and different conditions of synthesis. Because the morphology and size of nanostructured silica particles is of both great significance and challenge, this dissertation was focused to prepare self-organized nanostructured silica particles through organic template assisted wet-chemical method. Additionally, other research finding on the synthesis of polystyrene (PS) with diameter as small as 50 nm was also presented in this current research. It is suggested that the present research will be gained and can bring to the improvement of particle technology, especially concerned to the fields of chemical and material engineering. This dissertation is mainly divided into five chapters, in which some section comprises several sections. The brief descriptions of each chapter in this dissertation are shown below.

The background and the motivation of the current research are described in **Chapter 1**. Basic explanation and review of previous researches on the synthesis and application of hollow and porous silica fine particles were also provided.

Numerous studies of the synthesis of mesoporous silica (MPS) particles with tailored properties have been published. Among those studies, tetraethyl orthosilicate is commonly used as a silica source, but tetramethyl orthosilicate (TMOS) is rarely used because its reaction is fast and difficult to control. In **Chapter 2**, an improved synthesis route of hollow silica particles using TMOS and their applications as an optically and thermally insulating polymer additive are explained. The results showed that as synthesized particles were successfully implemented into polymer films and permitted maintaining optical transparency while significantly improving the heat barrier properties of composite film. The shell-thickness of particle was controlled from 6.2 to 17.4 nm by increasing TMOS concentration and the diameter of particle from 95 to 430 nm through use of the different sizes of polystyrene particles. Hollow silica particle with the shell-thickness about 6.2 nm displayed a high light transmittance intensity up to 95%. Poly-ethersulfone (PES)/hollow silica composite films (35 ± 5 μm thick) exhibited a much lower thermal conductivity (0.03 ± 0.005 W $m \cdot K^{-1}$) than pure polymer films. The prepared particle has promising for cost and energy effective optical devices requiring thermal insulation.

Emulsion based synthesis of porous particles is a straightforward and relatively mild process to prepare a wide variety of materials with well controlled particle size, shape, composition and spatial arrangement. **Chapter**

3, the synthesis of submicron sized spherical mesoporous silica using TMOS via an oil-water (O/W) emulsion process. The characterization of the particles and how their morphology depends on the emulsions formed were detail studied. An important part of the present work was to understand the mechanism behind the formation of these unusual structures and an explanation based on hydrolysis-condensation reaction of TMOS double template emulsion process.

Polystyrene (PS) nanoparticles, both anionic and cationic, have attracted much interest owing to their excellent properties such as low density, high hydrophobicity, large specific surface area, good mechanical and chemical stability. They have many potential applications as templates, calibration standards, film coatings, ink toners, or polymer fillers, or in chromatographic separation and drug delivery. The size and distribution of PS particle are considered to be the most important parameters that succeeded in these applications. Hence, how to synthesize and control highly dispersed PS particles with the size around 50 nm or less becomes a critical issue. In **Chapter 4**, a modified emulsion polymerisation synthesis route for preparing highly dispersed cationic polystyrene nanoparticles is described. The combined use of 2,2′-azobis[2-(2-imidazolin-2-yl) propane] di-hydrochloride (VA-044) as the initiator and acetone-water as the solvent medium afforded successful synthesis of cationic PS particles as small as 31 nm in diameter. This study provides important insights and a new methodology for further research and application, especially for synthesis and design of nanostructure materials.

Chapter 5 contains the summary of all chapters and direction for further research.