



Invited review paper

Development of new templating approach for hollow nanoparticles and their applications



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ABSTRACT

This review mainly focuses on simple template routes for hollow particles and introduces their applications. An inorganic template using inorganic particles is a facile technique to obtain hollow particles with various shapes, sizes, from nano to micron, and also different shell microstructures. These structure controls provide wide applications such as superior thermal insulation films, anti-corrosion films, and unexpected “easy-to-grip” volley ball coatings, etc. This technique is also an environmentally friendly route that only requires a mild condition to remove the template, and the generated byproducts can be reused for synthesis of the template. This paper describes the sol–gel synthesis which is useful to form the shell structure of the hollow particle and then how to control the particle structure using various templating routes. In addition to our achievements using hollow silica nanoparticles, the development of applications such as the lithium ion battery, biomedical products, and catalysts, will be outlined.

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1. Introduction

Many research studies in the last decade have been devoted to the development of preparing micro/nano-sized hollow particles (M/NHP) [1–4]. They have attracted tremendous interest because of their unique properties compared to other dense materials, for

instance, a high specific surface area, low density, and excellent permeation and penetration abilities. With the aid of modern science, it is already common knowledge that exceptional properties may occur by downsizing the hollow particle (HP) from micro to nano-size without changing their chemical composition which can notably affect the performance of the synthesized materials [5]. In a typical process for applications, M/NHP is dispersed in a liquid medium which is generally been the basis for having a surprising array of material applications for technological scientific value. The fictionalization of the M/NHP (with facile processing)

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can then extend the impact of the M/NHP that may generate a novel synthetic implication.

The hollow interior of the HP is said to be the second-generation two-level structural micro/nano-material which exhibits a complex chemical composition but relatively simple structures and hierarchically constructed that can show superior physico-chemical properties [3,4]. Whereas, the third-generation M/NHP with multilevel porous shell wall structures or having a complex interior architecture, have been also spurred interest in the field of nano-material science applications. Just for the purpose of discussion, the first generations are said to be a one-level structure with solid interiors (solid particles). In this case, this paper mainly focuses on the second and third generation micro/nano-material fabrications.

This review paper describes the simple preparation for the fabrication of M/NHP in an eco-friendly approach. Due to the broad scope of this topic, all research studies about M/NHP in every eco-processing area cannot be treated in detail. Instead, we focused our major existing research of M/NHP by adding some current improvements for the preparation of silicate and other inorganic M/NHPs. Many excellent articles have been published for the advancement of silicates and other inorganic M/NHP with respect to the architectural design, synthesis and bio-related applications (drug release) [5–7], but only a few deals with establishing a route that would be entail environmentally friendly. This is an important task because taking good care of our environment is a major concern, and if we can eliminate toxic waste byproducts, it would be good for humans. Also, many studies involving single-step or one-pot eco-synthesis approaches would become available and developing an eco-large-scale synthesis of HP would be a great challenge to eliminate environmental problems concerning the fabrication of HP which would provide many commercial applications.

This review paper is organized as follows: First, the basic fundamental process which is the sol-gel is assessed. This is followed by the general schematic approaches for M/NHP that our group has done and other significant reference papers about HP preparations such as the solid-template (solid-cores template method), soft-template (double emulsion method), and facile process like ammonia-hydrothermal approach (AMT). The common applications of M/NHP are then summarized. This kind of approach is primarily geared towards the sol-gel process; a wet-chemical technique usually used for the fabrication of M/NHP which can create both glassy and ceramic materials.

2. Fundamental process: sol-gel

Basically, the sol (or solution) gradually evolves towards the formation of a gel-like network containing both a liquid phase and a solid phase. The precursors used in sol-gel processing consist of a metal or metalloid element surrounded by various reactive ligands. Metal alkoxides, such as aluminates, titanates and zirconates, are the most popular precursors because of their high reactivity towards water. The most widely used non-metal alkoxides are alkoxy silanes which is mostly discussed in this section, such as tetramethoxysilane (TMOS) and tetraethoxysilane (TEOS). Although ethyl groups are the most common alkoxy groups, methoxy, propoxy, butoxy and other long-chain hydrocarbon alkoxy groups are also used in alkoxy silanes. Metal alkoxides (organometallic compounds) are used in the sol-gel process either alone or in combination with non-metal alkoxides such as TEOS or alkoxyborates. The basic structure or morphology of the solid phase can range anywhere from discrete colloidal particles to a continuous chain-like polymer network [8,9]. A well studied alkoxide, TEOS, whose chemical formula is given by $\text{Si}(\text{OC}_2\text{H}_5)_4$, or $\text{Si}(\text{OR})_4$ where the alkyl group $\text{R} = \text{C}_2\text{H}_5$. These alkoxides are ideal chemical precursors for the sol-gel synthesis because they readily react with water. The reaction is called hydrolysis, because a hydroxyl ion becomes attached to the silicon atom [10]. The process consists of a series of hydrolysis and condensation reactions of an alkoxide, which proceed according to the reaction scheme shown in Fig. 1. The alkoxy silanes are used as an example but all of the metal alkoxides similarly react. Wherein hydrolysis and condensation reactions of most metal alkoxides can be carried out without a catalyst because of the extremely fast rates of reaction, alkoxy silanes hydrolyze much more slowly, requiring the addition of either an acidic or basic catalyst. Hydrolysis is initiated by the addition of water to the silane solution under acidic, neutral, or basic conditions. Thus, the polymerization is associated with the formation of a dimensional network of siloxane $[\text{Si}-\text{O}-\text{Si}]$ bonds accompanied by the production of $\text{H}-\text{O}-\text{H}$ and $\text{R}-\text{O}-\text{H}$ species.

By definition, condensation liberates a small molecule, such as a water or alcohol. This type of reaction can continue to build large silicon-containing molecules by the process of polymerization. Thus, a polymer is a huge molecule (or macromolecule) formed from hundreds or thousands of units called monomers. The number of bonds that a monomer can form is called its functionality. The polymerization of silicon alkoxide, for instance, can lead to the complex branching of the polymer, because a fully hydrolyzed monomer, $\text{Si}(\text{OH})_4$, is tetra functional (can branch or bond in 4 different directions). Alternatively, under certain conditions (e.g., low water concentration) fewer than 4 of the OR or OH groups (ligands) can be capable of condensation, so relatively little branching would occur. The mechanisms of hydrolysis and condensation, and the factors that bias the structure toward linear or branched structures are the most critical issues of sol-gel science and technology [8,9,11,12].

Therefore, the sol-gel process has been frequently employed in coating the colloidal core-templates followed by forming M/NHP by removing the templates. This is a simple reaction that does not require unusual materials, catalysts or expensive deposition equipment. Likewise, sol-gel reactions do not employ extreme reaction conditions. The reactions can take place at room temperature and require only moderate temperatures to 'cure' the gel which can easily remove the excess water/alcohol that the reaction generates. Also, the properties of the M/NHP prepared using sol gel approaches can easily be modified by utilizing an organically modified alkoxide or a variable metalloid (for example, an alkoxyborate instead of an alkoxy silane).

HPs are commonly obtained through self-assembly using surfactants because it provides superior control over the nucleation

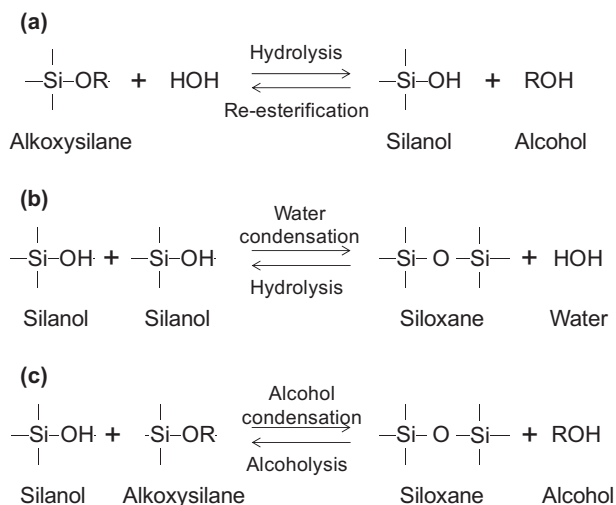


Fig. 1. Schemes of typical sol-gel reaction.

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