Preparation and Size Control of Hollow Silica Nanoparticles with High Dispersibility using Vesicle Template Method

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Hollow silica nanoparticles (HSNPs), having low specific gravity and high specific surface area, demonstrate several unique properties compared to normal solid particles. Recently, these HSNPs have received increased attention in designing new type of low-refractive index materials and low dielectric constant materials. In our previous study, we successfully prepared highly dispersible HSNPs with an average particle size of 20–30 nm using cationic vesicles as soft templates[1]. However, for practical applications, HSNPs with larger particle sizes and higher porosity are required to achieve improved performance. Therefore, in this study, we aimed to synthesize HSNPs with larger particle sizes than those previously reported by using the vesicle template method.

The vesicles were prepared with a mixture of a double-chained cationic surfactant, didodecyldimethylammonium bromide (DDAB), and a single-chained cationic surfactant, dodecyltrimethylammonium bromide (DTAB), with DDAB/DTAB molar ratios of 9/1 or 7/3. The vesicle dispersions were prepared in ammonia basic solutions at pH 11.5 or 11.7 by ultrasonic irradiation treatment. After adding the silica precursor (tetraethyl orthosilicate : TEOS) to this dispersion under stirring for 2 hours, the pH was adjusted from basic to neutral (pH 8.0) condition. Following additional stirring for 16 hours, hydrothermal treatment at 150 ℃ was carried out to obtain the HSNPs.

When the reaction of TEOS was maintained at pH 11.5 for the first 2 hours and the pH was subsequently decreased to 8.0, highly dispersible HSNPs with particle sizes of 20–30 nm were obtained after hydrothermal treatment when the composition of DDAB/DTAB = 9/1 [1]. When the pH was increased to pH 11.7, the accelerated sol-gel reaction produced HSNPs with a particle size of 40 nm, though particle aggregation was significantly observed. This is likely because increasing the pH accelerates the sol–gel reaction, causing the silica shell to form before the vesicles are destabilized and shrunk under the shear force of stirring. Next, the pH was fixed at 11.7, and the molar ratio of DDAB/DTAB was varied. As a result, well-dispersed HSNPs with a particle size of 46 nm were obtained at a composition of DDAB/DTAB =7/3 . Furthermore, the effect of using tetrapropyl orthosilicate (TPOS) as an alternative silica precursor on the structure and dispersibility was also investigated.

Keywords:
Vesicle Template Method
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References:

[1] K.Tsujii, et al., Abstract of the *9 th International Colloids Conference* (2019).