Control of the particle morphology and particle outer diameter (from nano and submicron sizes) has increasingly captured the attention of researchers for decades. The exploration of unique sizes and shapes as they relate to various properties has become a great quest for large field applications. To meet these demands, this study covered our recent developments in an aerosol-assisted self-assembly technique (a spray method) for particle processing. The particle processing of several morphologies (sphere, doughnut, encapsulated, porous, hollow, raspberry, and hairy shapes) was discussed in terms of the selection of material types, the addition of supporting materials, and the change of process conditions. Controllable particle outer diameter was discussed in terms of the adjustment of the droplet size and concentration, and the addition of specific techniques. A theoretical mechanism was also simplified described, especially to describe how particles are designed with various sizes and morphologies. The performance of various particle morphologies was also demonstrated, which was essential for an understanding of the importance that shape could exert on practical use. Because the method outlined here can be broadly applied to the production of various types of functional materials, we believe that this report contributes new information to the field of chemical, material, environmental, and medical engineering.

**Keywords:** Nanomaterial processing, Spray Pyrolysis route, Functional nanostructure, Composite material, Morphological control of particle

1. Introduction

Recently, control of particle morphology has received a tremendous amount of attention. The change of shapes has caused unprecedented chemical and physical properties that differ markedly from those of bulk or dense material. Great potential for use in various applications could be achieved with morphological control: electronics, catalysts, drug carriers, sensors, pigments, and magnetic and optical materials, etc.

In this report, a spray method was used to produce particles with different morphologies. Several parameters, which could alter morphologies, were investigated.

2. Experimental method

Inorganic particles with controllable size and morphology were prepared using a mixture of main
and supporting components in the specific solvent with different initial concentrations. The mass ratio of main and supporting components was fixed at the optimum ratio. The mixed solution was sonicated initially in an ultrasonic bath for several minutes to obtain a homogenous solution. Then, the mixed solution was sprayed using a spray-equipment system. In briefly, the apparatus consists of ultrasonic nebulizer (Omron Corp., NE-U12, freq. 1.7 MHz) for generating droplets from the precursor, laminar flow tubular furnace (a ceramic tube; D = 13 mm, L = 1 m) and electrostatic precipitator or filter. The generated droplets were then heated with two heating zones in the tubular furnace. The first zone (T = 200°C) was used to evaporate the solvent in the droplet, resulting in large particles of composite main/supporting component particles. Then, the second zone (T = 500°C) was used to evaporate the remained solvent and change the particle morphology. In addition, flow of N2 gas, as the carrier gas, is approximately 1 L/min. Filter or electrostatic precipitator was used for collecting the particles. Then, the morphology and particle size of prepared particles were characterized using a scanning electron microscope (SEM, S-5000, Hitachi, Tokyo, Japan, operated at 2 kV) and a transmission electron microscope (TEM, JEM-3000F, JEOL, Tokyo, Japan, operated at 30 kV).

3. Result and discussion

The spray-drying method has great potential for the synthesis of particles that are rich in desirable properties. A simple process can be achieved, which involves only solvent evaporation and self-assembly of materials inside the droplet system. The control of particle shape is also possible by adding some technical modifications. The features of the initial raw material (e.g., initial particle size, type of material, physical and chemical properties, and surface charge) and process conditions play an important role in producing various products.

Fig. 1 shows a conclusion of our work. The effectiveness of this method in controlling particle morphologies that include dense, hollow, encapsulated, hairy, and doughnut forms had been

![Diagram of various particle morphologies produced by spray method](image-url)
described. An explanation of the various morphologies could be simplified as follows:

(i) The spherical particles are basically prepared using the spray method because the particles are produced from the droplets with a spherical shape, and the most stable shape for a droplet is the spherical form.

(ii) The particles with doughnut and hollow shapes could be prepared by changing the process conditions (e.g., flow rate, temperature process, addition of surfactant).

(iii) The production of particles with a multi-component was possible when multi-components were added to the initial precursor. Well-mixed component particles could be created when the component fractions (i.e., size) were almost the same, while the possibility of the particle encapsulation phenomenon could be prepared when the component fractions were different.

(iv) The hairy particle could be produced when the specific material (i.e., a CNT catalyst) was added to the precursor.

(v) The particles with porous structures could be formed when the template component was added to the precursor.

The various morphologies of particles exhibit many potential applications for their uses in future technology (Table 1). Enhancement of particle performance could be achieved by a change in particle morphology.

| Particle morphology | Precursor | Density | Porosity | Enhancement Properties |
|---------------------|-----------|---------|----------|------------------------|
| Completely Spherical Dense Particle | Solution type (e.g., zirconate) | $\rho_p = \rho_{app}$ | $\varepsilon = 0$ | — |
| Small Rough Spherical Dense Particle | Nanoparticles ($d_p << d_w$) | $\rho_p < \rho_{app}$ | $\varepsilon > 0$ | Near to Normal |
| Highly Rough Spherical Dense Particle | Nanoparticles ($d_p < d_w$) | $\rho_p < \rho_{app}$ | $\varepsilon > 0$ | Near to Normal |
| Hollow Particle | Effect of Fast Evaporation rate | $\rho_p < 0.1 \rho_{app}$ | $\varepsilon > 30\%$ | Ultra low refractive index |
| Doughnut Particle | Effect of Hydrodynamic | $\rho_p << \rho_{app}$ | $\varepsilon > 30\%$ | High Surface Area Low Refractive index |
| Porous Particle | Additional template component | $0.3 \rho_{app} < \rho_p < \rho_{app}$ | $0 < \varepsilon < 70\%$ | Very high surface area Very low Refractive index |
| Encapsulated Particle | Multi component ($d_p < 3 d_w$) | $\rho_p \sim \rho_{app}$ | $\varepsilon = 0$ | Protect Core material |
| Mixed Particle | Multi component ($d_p < 3 d_w$) | $\rho_p \sim \rho_{app}$ | $\varepsilon = 0$ | Specific Properties (e.g., dope material) |
| Hairy Particle | Multi component ($d_p < 3 d_w$) + Additional CNT catalyst component | $\rho_p \ll \rho_{app}$ | $\varepsilon >>>$ | CNT-related Properties |

Note: *depend on the CNT population and tube size; $\rho_p =$ particle density; $\rho_{app} =$ appearance/real density; $\varepsilon =$ porosity; $d_{p1} =$ size of component 1; $d_{p2} =$ size of component 2
4. Summary

Critical issues, associated with the development of the aerosol-assisted self-assembly technique (using the spray method) to obtain effective strategies in designing particle morphology, had been discussed in this study. The types and the concentrations of precursors, the selection of process conditions, and the addition of supporting materials all played significant roles in the production of particles with various sizes and morphologies. Exhibition of unique performance could also be obtained by the management of particle morphologies. We believe this report contributes new information to the fields of chemical, material, environmental, and medical engineering because the method had shown that broad application could be born from the management of particle morphology. Finally, size- and morphological-controllable synthesis of particles is of both great significance and challenge, and need to be further explored.

5. Reference of published paper and proceeding information that supported by Hosokawa Micron Foundation

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