Liu. J Powder Metall Min 2013, 2:5
http://dx.doi.org/10.4172/2168-9806.1000e122

Powder Metallurgy & Mining

Editorial Open Access

Carbon Sphere Obtained Via Hydrothermal Carbonization as Hard Template: Preparation of Hollow Metal Oxide Sphere

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As semiconductor materials, metal oxides with specific physical and chemical characters are widely used as catalysts, co-catalysts and carriers. Hollow spheres with specific structure and morphology has attracted increasingly interest because of its unique properties such as well monodispersity and stability, high specific area and low density. In addition, particles of hollow spheres itself can be seen as a nano-reactor [1], which can improve the metal oxide performance by varying the internal micro reaction environment. Recently, various chemical and physicochemical methods, including templates [2], self-assembly techniques [3], chemical induction [4], ultrasonic and solvent thermal growth [5] methods have been developed for the preparation of hollow metal oxide spheres. Among these techniques, hard template methods using carbon spheres from monosaccharide hydrothermal carbonization exhibited great promising [6].

Hydrothermal carbonization is proved apt to generate monodispersed carbonaceous spheres [7]. Carbon spheres obtained via hydrothermal carbonization is a promising hard template for the preparation of hollow metal oxide spheres with following advantages: (1) the HTC reaction facilities are simplicity and carbon raw materials are usually monosaccharide, cellulose, lignocelluloses and such biomass that are environmental friendly; (2) reaction proceeds in a mild condition (T=100-300) and the templates can be facile removal by calcinations; (3) the carbon spheres are rich in oxygen-containing hydrophilic functional groups such as hydroxyl groups, carboxyl groups and carbonyl groups which are benefit for binding with metal; (4) the size, structure and properties of the templates can be controlled produced through change the reaction conditions, in turn determine the properties of hollow metal oxide spheres.

Until now, preparation of hollow metal oxide sphere using as-synthesized carbon spheres as hard templates mainly through two ways. The general synthesis method mainly includes the adsorption of metal ions from solution to the surface layer of carbonaceous spheres and subsequent removal of the carbonaceous cores via calcinations [8-10]. Nearly all common metal oxide hollow spheres have been successfully synthesized, such as TiO_2, SiO_2, $Cr2O_3$, MnO_2, ZnO, SiC, CaO and so on, especially those are not accessible by other processes. It is worth mentioning that the sizes and structures of different metal oxide hollow spheres are predominantly determined by the templates, and showing well performance as catalysts, energy storage, optical and electrochemical materials. In our previous report [11], size and morphology controllable carbon spheres was prepared via glucose hydrothermal carbonization, they exhibited well performance as hard templates for the preparation of TiO_2 hollow spheres with high activity. Zhang [12] reported the fabrication of size controllable ZnO hollow nanospheres using carbon from sucrose hydrothermal carbonization as template. Ming [13] synthesized finely dispersed hollow TiO_2 nanoparticals using the same method, which exhibited excellent performance as anode materials with high capacities and stable cyclability in Li-ion battery applications.

Another method is all-in-one pathway to metal oxide hollow spheres through hydrothermal treatment of mixtures of carbohydrates with different metal salts in water in a sealed autoclaves. Carbon spheres with the metal oxide precursors tightly embedded in the microspheres were obtained. The removal of carbon directly resulted in hollow spheres of the corresponding metal oxide. The obtained products exhibit high surface areas as well as porosity that are brought about by the interstitial porosity between the metal nanoparticals. Porosity can provide more active sites for the next practical application. Demir-Cakan [14] prepared mesoporous SnO_2 microspheres by hydrothermal treatment in the presence of glucose. After the removal of template through calcination, the hollow spheres with nanosized building block and high electrochemical performance can be obtained. Wang [15] synthesized CO_3O_4 hollow spheres with two-level hierarchical pores and high areas, the mesoporous can be tuned by pre-treatment of the carbon precursor, the CO_3O_4 show high catalytic activity and durability for the combustion of CH_4. Carbon spheres from HTC as hard templates have opened an era for the preparation of hollow metal oxide sphere, while we expect some further study next. 1) Make use of more low cost and environmental friendly raw materials such hemicelluloses, lignin and some by-products. 2) Establish control system of reaction, find out the regularity between reaction conditions, specific area and porous structure for the control preparation.

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Received October 14, 2013; **Accepted** October 15, 2013; **Published** October 16, 2013

Citation: Liu S, (2013) Carbon Sphere Obtained Via Hydrothermal Carbonization as Hard Template: Preparation of Hollow Metal Oxide Sphere. J Powder Metall Min 2: e122. doi:10.4172/2168-9806.1000e122

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