

Synthesis of monodispersed iron oxide particles in a large-scale microwave reactor

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Abstract

Well defined, monodispersed iron oxide particles can be synthesized in conventional hydrolysis approaches, but the synthesis generally takes 2-7 days. Microwave hydrothermal approaches offer the possibility to significantly reduce the synthesis time. This work demonstrates the feasibility of microwave synthesis in large scale. The batch size was five liters, and the holding time was 2 hours. The crystallinity, the shape and size of the synthesized particles were comparable to those produced from conventional approaches. The only problem was a relatively long period for cooling down of the product solution. When this period can be eliminated or significantly shortened, a production facility with laboratory size but industrial productivity will be realized.

Introduction

γ - Fe_2O_3 based magnetic materials have been extensively used as recording materials because of their good magnetic properties. Conventionally, γ - Fe_2O_3 powders with monodispersed particles are synthesized first by means of forced hydrolysis of ferric nitrate or ferric chloride in aqueous solutions, which produces α - Fe_2O_3 , and then conversion of the α - Fe_2O_3 into γ - Fe_2O_3 in a high-temperature process^{1,2}. To effectively control the particle shape and particle size, which are essential for achieving the desired magnetic properties, the hydrolysis solution must be very dilute, usually within a range around 0.02M^{3,4}. Furthermore, the conventional hydrolysis has to be carefully controlled and it generally requires 2-7 days. This means that, even based upon the shortest processing time and highest concentration, the rate for producing the nanosized iron oxide powders would be less than 0.014 gram per liter per hour. In order to make sense for industrial applications of the nanosized magnetic materials, appreciable acceleration of the process is needed. Although previous researches have demonstrated that microwave-assisted hydrolysis was capable of accelerating the formation of crystalline iron oxide powders^{5,6}, there is lack of efforts to utilize this acceleration approach in large scale producing monodispersed iron oxide powders.

Experimental

Microwave-assisted hydrolysis of ferric nitrate and ferric chloride was performed in a self-built microwave reactor. The reaction chamber was 7 liters in volume and the microwave power was 6 kW. Ferric nitrate and ferric chloride was reagent grade

purchased from Aldrich Chemical Company, Inc. The hydrolysis was carried out with the iron salts solutions of various ferric ion concentrations. The temperature, pressure and holding time were 160°C, 120 psi and 2 hours, respectively.

XRD was used to characterize the crystallinity and the crystallography of the produced powders. SEM was used to examine the particle characteristics.

Results and discussion

Under our experimental conditions, all of the powders produced from the microwave-assisted hydrolysis of ferric nitrate were α -Fe₂O₃, despite of ferric ion concentrations in the hydrolysis solutions. However, the physical appearances were distinctly different as the ferric ion concentration varied. Powders prepared from 1M ferric ion solutions appeared black in color and had large agglomerates, which can be seen under an optical microscope. Powders prepared from 0.025-M ferric ion solutions were red. SEM images (Fig.1) revealed that the powders consist of uniformly distributed spherical particles of ~0.2 μ m in diameter.

The crystallography of powders prepared from ferric chloride solutions depends on the ferric ion concentration. While the 0.025-M Fe³⁺ solutions yield uniform submicron α -Fe₂O₃ spheres, the 0.1- and 1-M solutions yield yellow colloids, which were characterized as β -FeOOH.

The results obtained from this study demonstrate the feasibility to scale up the microwave-assisted hydrolysis approach. The microwave acceleration capability remains the same as the batch size increased from milliliters to liters, and the powders produced from the large scale have the same quality as those produced from small scales. The significant kinetic acceleration in large-scale synthesis shows a promise for chemical plant miniaturization. Consider that, if the job of a plant can be done by a workshop that is just one hundredth of the plant, the savings in energy, labor and facilities investment will be tremendous.

The only problem was the relatively long period for cooling down of the product solution. For a 5-liter batch, this period takes 3-4 hours. We anticipate that the period will be longer with the increase of the batch size. In conventional process, such a period can be neglected. However, when we want to utilize microwave effect to miniaturize the chemical plant, we should employ appropriate approaches to eliminate or significantly shorten such a period.

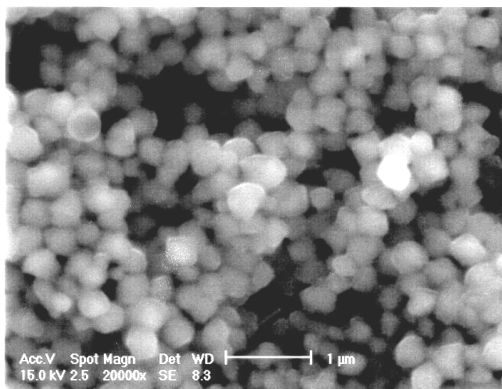


Fig.1 Monodispersed iron oxide particles synthesized by microwave wet chemical approach in multi-liter scale

Conclusion

The study on synthesis of iron oxide powders shows that the microwave acceleration capability can be retained as the process increased from milliliter batches into liter batches. Compared to conventional hydrolysis approaches, the microwave-assisted hydrolysis is able to shorten the process from several days to two hours. It shows a promise to miniaturize a large plant into a small workshop, which has significant potentials in saving energy, labor and facilities investment, and may also have great advantages in production management. For a miniaturized plant, appropriate approaches to eliminate or significantly shorten the product-cooling period are to be employed.

Reference:

1. M. Ozaki, S. Kratochvil and E. Matijevic; "Formation of monodispersed spindle-type hematite particles"; *Journal of Colloid and Interface Science*, 102 [1] 146-151 (1984).
2. M. Ozaki, S. Kratochvil and E. Matijevic; "Preparation and magnetic properties of monodispersed spindle-type γ - Fe_2O_3 particles"; *Journal of Colloid and Interface Science*, 107 [1] 199-203 (1985).
3. M.P. Morales, T. Gonzalez-Carreno and C.J. Serna; "The formation of α - Fe_2O_3 monodispersed particles in solution"; *J. Mater. Res.*, 7 [9] 2538-2545 (1992).
4. P.D. Sawant; "Characterization of hematite sols: correlation of size, shape and percentage yield"; *Bull. Mater. Sci.*, 20 [1] 27-35 (1997).
5. S. Komarneni, V.C. Menon and Q.H. Li; "Synthesis of ceramic powders by novel microwave-hydrothermal processing"; *Ceramic Transactions*, 62 1042-1122 (1996).
6. P. Rigneau, K. Bellon, I. Zahreddine and D. Stuerger; "Microwave flash-synthesis of iron oxide nanoparticles"; *The European Physical Journal Applied Physics*, 7 41-43 (1999).