

Synthesis and Characterization of Nanosized $Zn_xMn_{1-x}Fe_2O_4$ Powders by Glycothermal Process

Dong-Sik Bae¹, Eun-Jung Kim², Sang-Whan Park³ and Kyong-Sop Han⁴

¹Dept. of Ceramic Sci. and Eng., Changwon National Univ., Changwon 641-773, Korea

²⁻⁴Division of Ceramics, Korea Institute of Science & Technology, Seoul 136-791, Korea
dsbae7@changwon.ac.kr

Keywords: $Zn_xMn_{1-x}Fe_2O_4$ Nanosized powders; Glycothermal process; Superparamagnetism.

Abstract. Nanosized $Zn_xMn_{1-x}Fe_2O_4$ powders were prepared in ethylene glycol solution under mild temperature and pressure conditions by precipitation from metal nitrates. The average size and distribution of the synthesized $Zn_xMn_{1-x}Fe_2O_4$ powders were 10-20 nm and narrow, respectively. The magnetic property of the synthesized $Zn_xMn_{1-x}Fe_2O_4$ powder was of superparamagnetic character at room temperature.

1. Introduction

Ferrites are important magnetic materials, which are widely used in many electronic and magnetic applications, as in transformers, choke coils, high frequency application, data storage, noise filters, and recording heads, owing to their high magnetic permeabilities and low magnetic losses [1,2]. The small sizes and the single domain nature of the nanosized ferrite powders give them super paramagnetic properties [3]. Many different ways of producing nanosize ferrite powders are described in the literature such as sol-gel processing [4], hydrothermal processing [5,6], and ion exchange resin manufacture methods [7,8]. In most of the methods listed above, the crystalline phases are frequently developed during calcinations of the amorphous room temperature phases, whereas crystalline phases may be formed during synthesis in the glycothermal technique. It is one of the good processes to produce well-dispersed nanosized particles with a narrow size distribution in solution, and also the processing costs are less compared with other production techniques of nanosized powders [9]. Some papers reported that process conditions such as solute concentration, reaction temperature, reaction time, and the type of solvent are carefully controlled, and the desired shape and size of the particles can be produced [10, 11].

The objective of this study is to prepare the nanosized $Zn_xMn_{1-x}Fe_2O_4$ powders with superparamagnetic property by the glycothermal process, and to investigate the effects of the processing conditions on the formation, morphology, and phase of the powders.

2. Experimental Procedure

The preparation sequence of $Zn_xMn_{1-x}Fe_2O_4$ powders is schematically illustrated in Fig.1. $Zn_xMn_{1-x}Fe_2O_4$ precursors were precipitated from 0.1 M $Zn(NO_3)_2 \cdot 6H_2O$, 0.1 M $Mn(NO_3)_2 \cdot 6H_2O$ and 0.1 M $Fe(NO_3)_3 \cdot 9H_2O$ solution by slowly adding 1 M KOH

solution with rapid stirring in which the pH of starting solutions varied between 5 and 9. The precipitated $Zn_xMn_{1-x}Fe_2O_4$ precursors were washed by repeated cycles of centrifugation and re-dispersion in water. Washing was performed for a minimum of five times in ethanol. Excess solution was decanted after the final washing and the wet precursor was re-dispersed in 250 ml ethylene glycol under vigorous stirring. The resulting suspension was placed in a 1000 ml stainless steel pressure vessel. The vessel was then heated to the 200 °C at a rate of 10 °C/min. The morphology of the synthesized particles was observed using transmission electron microscopy (TEM, Philips, JEM-200CX). The magnetic properties of the powder were measured by VSM (Toei Kogyo Co. Ltd).

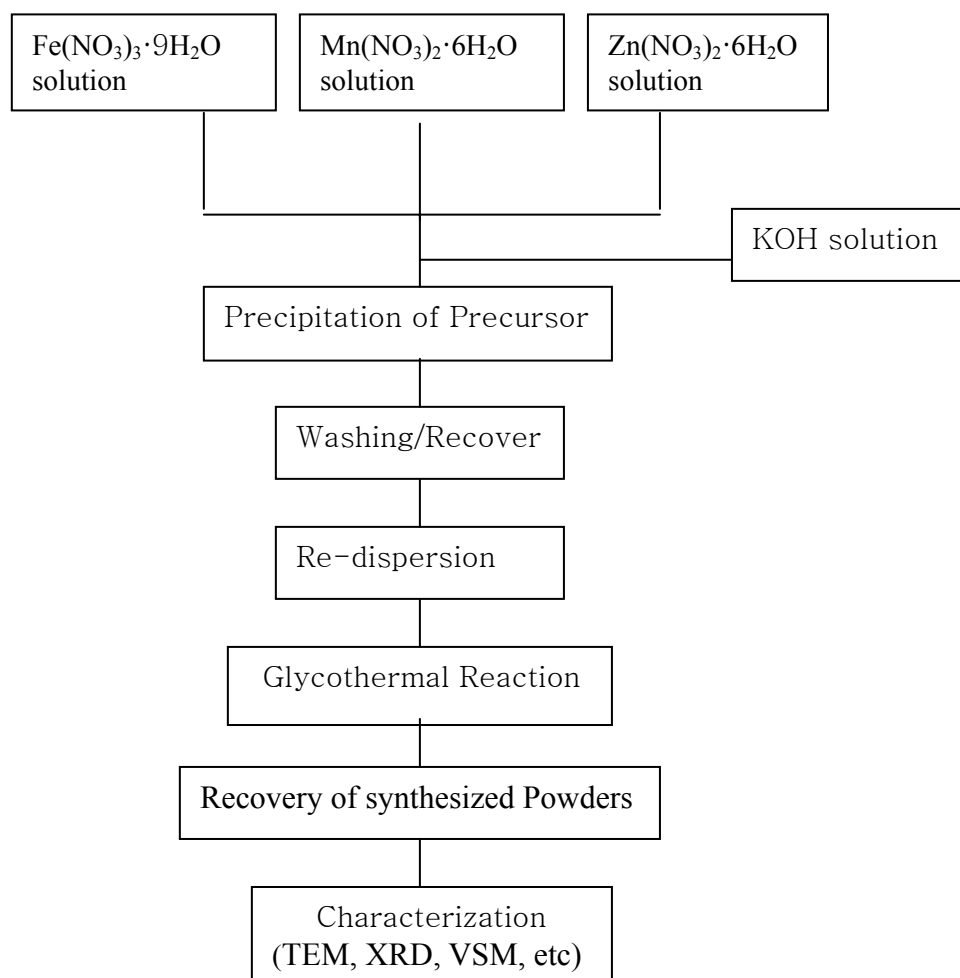


Fig. 1. Experimental flow chart of synthesized $Zn_xMn_{1-x}Fe_2O_4$ powders by glycothermal reaction in ethylene glycol solution.

3. Results and discussion

Glycothermal processing conditions such as pH, reaction temperature and time, have significant effects on the formation, phase component, morphology and particle size of $Zn_xMn_{1-x}Fe_2O_4$ powders. Fig. 2 shows the transmission electron microscopy of the synthesized particles as a function of the reaction temperature. The average size of the synthesized particles increased with reaction temperature increased. It has been shown that the average sizes of the synthesized $Zn_xMn_{1-x}Fe_2O_4$ powders are in the range of 10 to 20 nm and the size distributions was less than 25%. Fig. 3 shows the X-ray diffraction pattern of the $Zn_xMn_{1-x}Fe_2O_4$ powders synthesized in ethylene glycol solution. The X-ray powder diffraction patterns have shown that the synthesized particle has a crystalline phase. Fig. 4 shows the magnetization behavior as a function of reaction temperature. It has been shown that magnetic properties of the synthesized $Zn_xMn_{1-x}Fe_2O_4$ powders exhibiting superparamagnetic behavior. The saturation magnetization of synthesized $Zn_xMn_{1-x}Fe_2O_4$ powders was above 40(emu/g). It is possible to the application of magnetic nanoparticles for drug delivery using nanoparticulate magnetic carrier.

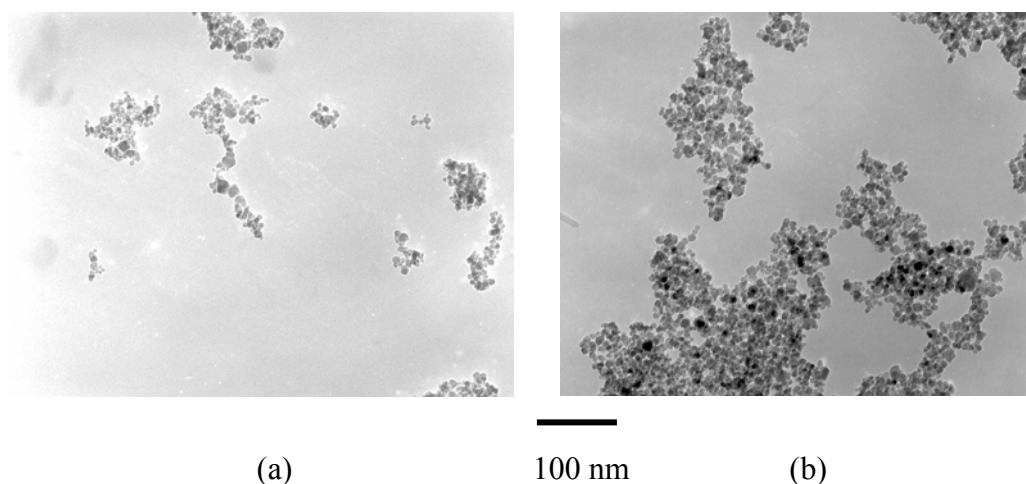


Fig. 2. TEM micrographs of the synthesized $Zn_xMn_{1-x}Fe_2O_4$ powders as a function of reaction temperature a) 200°C and b) 220°C for 6 h.

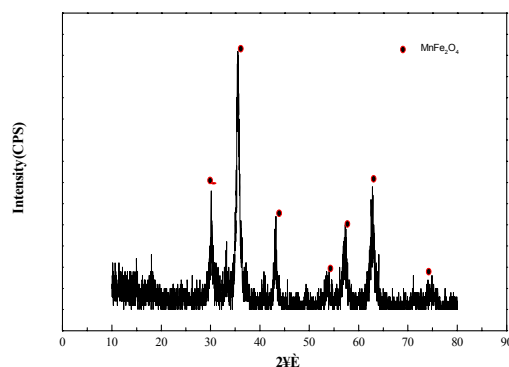


Fig. 3. X-ray diffraction pattern of the the synthesized $Zn_xMn_{1-x}Fe_2O_4$ powders by glycothermal process.

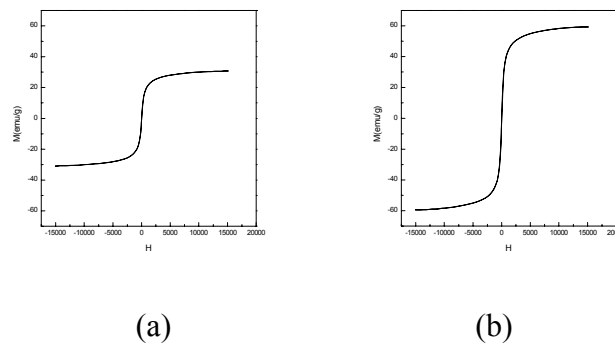


Fig. 4. Magnetic properties of the synthesized $Zn_xMn_{1-x}Fe_2O_4$ powders as a function of reaction temperature a) 200°C and b) 220°C for 6 h.

Conclusions

Nanosized $Zn_xMn_{1-x}Fe_2O_4$ powders were obtained in ethylene glycol solution reaction at 200°C for 6 h. The average particle size and distribution of the synthesized $Zn_xMn_{1-x}Fe_2O_4$ powders were in the range of 10 to 20 nm and narrow, respectively. The synthesized $Zn_xMn_{1-x}Fe_2O_4$ nanoparticles exhibited a superparamagnetic character. The saturation magnetization of synthesized $Zn_xMn_{1-x}Fe_2O_4$ powders was above 80(emu/g) at the pH=7.49.

Acknowledgment

This study was supported by the National Program for nano-structured materials technology of the Ministry of Science and Technology as one of the 21 Century Frontier Programs, Korea

References

- [1] B. R. Pieters, R. A. Williams and C. Webb, *Colloid and Surface Engineering*. (Ed. By R. A. Williams, Butterworth-Heinemann, Oxford, 1992).
- [2] M Ozaki, *Chemical Processing of Ceramics*. (Ed. By Burtrand I. Lee, Edward J. A. Pope, Marcel Dekker, New York,1994).
- [3] M. Rozman and M. Drogenik, *J. Am. Ceram. Soc.*, Vol. 78 (1995), p. 2449.
- [4] M. Seki, J. Sato, S. Usui, *J. Appl. Phys.*, Vol. 63 (1988), p.1424.
- [5] S. Komarneni, E. Freagan, and R. Roy, *J. Am. Cera. Sci.*, Vol. 71(1988), p. C26.
- [6] J. R. Ahn, D. S. Bae and J. S. Kim, *J. Kor. Ceram. Soc.*, Vol. 37(2000), p. 962.
- [7] P. Sainamthip and V. R. W. Amarakoon, *J. Am. Ceram. Soc.*, Vol. 71(1988), p. C92.
- [8] R. F. Ziolo, E. P. Giannelis, B. A. Weinstein, M. P. Ohoro, B. N. Ganguly, V. Mehrotra, M. W. Russell, D. R. Huffman, *Science*, Vol. 257 (1992), p.219.
- [9] S. Hirano, *Am. Ceram. Soc. Bull.*, Vol. 66(1987), p.1342.
- [10] W. J. Dawson, *Am. Ceram. Soc. Bull.*, Vol. 67(1989), p.1673.
- [11] S. B. Cho, S. Venigalla, and J. H. Adair, *Sci., Tech., & App., of Colloid. Susp.*, (edited by James H. Adair, Jon A. Casey, Clive A. Randall (Sridhar Venigalla),1995).