## Effects of Mn concentration on the ac magnetically induced heating characteristics of superparamagnetic $Mn_xZn_{1-x}Fe_2O_4$ nanoparticles for hyperthermia

Minhong Jeun,<sup>1</sup> Seung Je Moon,<sup>1</sup> Hiroki Kobayashi,<sup>2</sup> Hye Young Shin,<sup>3</sup> Asahi Tomitaka,<sup>2</sup> Yu Jeong Kim,<sup>4</sup> Yasushi Takemura,<sup>2</sup> Sun Ha Paek,<sup>3</sup> Ki Ho Park,<sup>4</sup> Kyung-Won Chung,<sup>5</sup> and Seongtae Bae<sup>1,a)</sup>

 <sup>1</sup>Department of Electrical and Computer Engineering, Biomagnetics Laboratory (BML), National University of Singapore, Singapore 117576
<sup>2</sup>Department of Electrical and Computer Engineering, Yokohama National University, Yokohama 240-8501, Japan
<sup>3</sup>Department of Neurosurgery, Cancer Research Institute, Ischemic/Hypoxic Disease Institute, Seoul National University College of Medicine, Seoul 110-744, Republic of Korea
<sup>4</sup>Department of Ophthalmology, Seoul National University College of Medicine, Seoul 110-744, Republic of Korea
<sup>5</sup>Daion Co. Ltd., Incheon 405-846, Republic of Korea

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The effects of  $Mn^{2+}$  cation concentration on the ac magnetically induced heating characteristics and the magnetic properties of superparamagnetic  $Mn_xZn_{1-x}Fe_2O_4$  nanoparticles (SPNPs) were investigated to explore the biotechnical feasibility as a hyperthermia agent. Among the  $Mn_xZn_{1-x}Fe_2O_4$  SPNPs, the  $Mn_{0.5}Zn_{0.5}Fe_2O_4$  SPNP showed the highest ac magnetically induced heating temperature ( $\Delta T_{ac,mag}$ ), the highest specific absorption rate (SAR), and the highest biocompatibility. The higher out of phase susceptibility ( $\chi''_m$ ) value and the higher chemical stability systematically controlled by the replacement of  $Zn^{2+}$  cations by the  $Mn^{2+}$  cations on the A-site (tetrahedral site) are the primary physical reason for the promising biotechnical properties of  $Mn_{0.5}Zn_{0.5}Fe_2O_4$  SPNP. © 2010 American Institute of Physics. [doi:10.1063/1.3430043]

Magnetic hyperthermia (MH) using a superparamagntice nanoparticle agent (SPNA) has recently drawn huge attraction due to its clinical promises.<sup>1,2</sup> Accordingly, the interests to utilize the ternary phase of SPNPs, MFe<sub>2</sub>O<sub>4</sub> (M=Fe, Co, Ni, and Mg), with a mean diameter below 10 nm for a MH agent has increased dramatically. However, despite their promising chemical, physiological, biotechnical, and physical properties suitable for SPNA applications,<sup>3</sup> an insufficient  $\Delta T_{ac,mag}$  and low specific absorption rate (SAR) at the physiologically tolerable range of frequencies and magnetic fields  $(f_{appl} < 100 \text{ kHz}, H_{appl} < 200 \text{ Oe})$  are still revealed as the technical challenges to be overcome for a real clinical MH.<sup>4,5</sup> Thus, a great deal of research activity is being conducted in order to develop a functional SPNA and to improve the efficiency of currently used ferrite SPNAs. The quarterly phase of bulk  $Mn_xZn_{1-x}Fe_2O_4$ , which is one of the softest (or the highest permeability) ferrite materials, is considered to be a potential material for MH agent. The main physical reason is that it has a good electrical field absorption and a large power loss  $(500-200 \text{ W/m}^3)$  at a low frequency (<100 kHz) which is due to its low electrical resistivity  $(0.02-20 \ \Omega \text{ m})$ .<sup>6,7</sup> Furthermore, its magnetic properties, i.e., saturation magnetization, M<sub>s</sub>, and magnetic susceptibility  $(\chi_{\rm m} = \chi'_{\rm m} + i\chi''_{\rm m}$ , particularly  $\chi''_{\rm m}$ ), which are directly relevant to the  $\Delta T_{\rm ac,mag}$  characteristics, can be easily controlled by adjusting the relative concentration of  $Mn^{2+}$  and  $Zn^{2+}$  cations in the  $Mn_xZn_{1-x}Fe_2O_4$ .<sup>6,8,9</sup> However, all of the works relevant to the  $Mn_xZn_{1-x}Fe_2O_4$  done so far were entirely focused on magnetoelectronics device applications.<sup>10</sup> There have been no systematic studies on the magnetic properties,  $\Delta T_{ac,mag}$  characteristics, and the biocompatibility of  $Mn_xZn_{1-x}Fe_2O_4$  SPNPs for MH agent applications until now.

In this paper, we report on the effects of  $Mn^{2+}$  cation concentration on the magnetic properties and the  $\Delta T_{ac,mag}$ characteristics of  $Mn_xZn_{1-x}Fe_2O_4$  SPNPs to explore its feasibility as a MH agent. The physical correlation between the magnetic properties of  $Mn_xZn_{1-x}Fe_2O_4$  SPNPs controlled by the  $Mn^{2+}$  cation concentration and  $\Delta T_{ac,mag}$ , characteristics including power loss mechanism were systematically investigated at different ac  $H_{appl}$  and  $f_{appl}$ . In addition, the cell viability of  $Mn_xZn_{1-x}Fe_2O_4$  SPNPs with different  $Mn^{2+}$  cation concentrations were quantitatively analyzed to evaluate the biocompatibility for *in vivo* MH agent applications.

The spinel ferrite  $Mn_xZn_{1-x}Fe_2O_4$  nanoparticles (NPs) with different Mn<sup>2+</sup> cation concentration were synthesized using a modified high temperature thermal decomposition (HTTD) method, where ramping up rate and heat treatment time were changed to 8.5 °C/min and 25 min, respectively, compared to a conventional HTTD method.<sup>11</sup> The Mn<sup>2+</sup> cation concentration of  $Mn_xZn_{1-x}Fe_2O_4$  NPs was systematically controlled from x=0.2 to 0.8 during the synthesis. The crystal structure, the particle size, and the distribution of  $Mn_xZn_{1-x}Fe_2O_4$  NPs were analyzed by using a Cu-k $\alpha$  radiated x-ray diffractometer (XRD) and a high resolution transmission electron microscopy (HRTEM). The  $\Delta T_{ac,mag}$  characteristics and the ac magnetic hysteresis of the solid state Mn<sub>x</sub>Zn<sub>1-x</sub>Fe<sub>2</sub>O<sub>4</sub> NPs were measured by using an ac solenoid coil system, which is connected to a capacitor. The  $f_{appl}$  and the  $H_{appl}$  were varied from 30 kHz to 210 kHz and from 60 Oe to 140 Oe, respectively. The dc magnetic properties of the

<sup>&</sup>lt;sup>a)</sup>Author to whom correspondence should be addressed. Electronic mail: elebst@nus.edu.sg.