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Study of specific loss power of magnetic fluids with various viscosities



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ABSTRACTS

Using hydrothermal method, $CoFe_2O_4$ (hard ferrite) and $MnFe_2O_4$ (soft ferrite) nanoparticles of size up to 20 nm were synthesized and the viscosities were controlled using various concentrations of agar. The hydrodynamic diameter of these particles was measured and fitted to a lognormal distribution and the results showed their polydispersity is very narrow. From the calorimetric measurements of the particles stabilized in agar solutions, we have demonstrated that at a given frequency, the dependence of the specific loss power of magnetic fluids on the viscosity is in good agreement with the theoretical predictions made in the earlier studies.

1. Introduction

Magnetic nanoparticles (MNPs) are of great interest because of their potential applications in various fields of science and technology [1–3]. One of the most promising directions of magnetic nanoparticle studies is to synthesize new magnetic systems for the use in fields ranging from high-density data storage [1] to biomedical applications [2-4]. Magnetic fluid hyperthermia (MFH) is a cancer treatment in which only the tumor cells are locally heated if magnetic nanoparticles are injected into tumor cells and the patient is placed under the external alternating magnetic field. This leads to hyperthermia, which damages and kills the tumor cells with minimal damage to the surrounding normal tissue[5]. In order to be applicable as mediators of nanohyperthermia, nanoparticles must meet a number of requirements, such as single domain, being stable, weakly agglomerated, and small in size[6]. For biomedical applications, MNPs should have superparamagnetic properties[6], which prevents the nanoparticle from aggregation and to assure their elimination after the magnetic field is switched off[7].

Considering the other condition, however, superparamagnetic nanoparticles (NPs) may not be the best choice for mediators of hyperthemia. It was reported that heating power was maximized in large ferromagnetic NPs with low anisotropy [5] and furthermore, the optimum size for the maximum power loss changes with the amplitude of the applied magnetic field [6]. Therefore, the choice between superparamagnetic and ferromagnetic NPs for hyperthermia is not a simple task but depends on several experimental conditions [8]. *In addition, the concentrations of MNPs must be controlled as low as possible to maximize the heat dissipation efficiency under the AMF*[9]. Furthermore, the MNPs should have narrow size-distribution since polydispersity impair the heating rates achieved [10]. Finally, MNPs should be rendered soluble in physiological fluids and biocompatible in order to interact with tissues.

The most commonly used physical quantity concerning the calorific power of MNPs under AC magnetic field (i.e. MFH) is the specific absorption rate (SAR), which can also be called specific loss power (SLP)[11]. There are several ways to lose energy, which could contribute to the SLP such as hysteresis loss, eddy current, Brown relaxation, and Néel relaxation [2,3,6]. According to the Néel relaxation and the Brown relaxation, SLP is dependent on several physical and magnetic parameters including particle size (D), size distribution, saturation magnetization (M_s), magnetic anisotropy constant (K), as well as the viscosity of magnetic fluid [3–15].

Rosensweig first reported that the SLP of Magnetite, Maghemite,

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Barium ferrite and Cobalt ferrite is dependent on the particle size, the size distribution and the viscosity of magnetic fluid [12]. Furthermore, he also showed that the dependence of SLP on D of hard ferrite (Barium ferrite and Cobalt ferrite) was different from that of soft ferrite (Magnetite and Maghemite) and the main difference between the resultant nanofluids is due to their anisotropy constants. As mentioned above, the heating rate is also dependent on the viscosity of magnetic fluid. However, there have been not many reports of influence of viscosity on the specific loss power of magnetic fluid. In a recent publication, SLP of monodispersed maghemite ($K=16 \text{ kJ/m}^3$) and cobalt ferrite ($K=123 \text{ kJ/m}^3$) particles was investigated in two different solvents, water and glycerol [16,17]. Under the applied field amplitude of 24.8 kA/m, it was observed that SLP decreased with the increase in viscosity and its effect was more pronounced in cobalt ferrite than in maghemite nanoparticles. Besides, Yolanda Pineiro-Redondo et al.[18] found that SAR of PAA-coated magnetite ferrofluids (preventing aggregate formation) increases slightly from 36.5 to 37.3 W/g when the solvent viscosity increases from $\eta=1$ mP s (water) to $\eta=17$ mP s (ethylene glycol). Most recently, Jeun et al.[15] studied the AC magnetically-induced heating characteristics of various viscous nanofluids, either soft ferrite (Fe₃O₄) or hard ferrite (CoFe₂O₄) superparamagnetic nanoparticles. They empirically found that the Brown relaxation loss power was greatly affected by the surrounding viscosity and its contribution to the total AC heat generation and the SLP was drastically degraded with increasing the viscosity up to 4×10^{-3} Pa s. By contrast, the contribution of the Neel relaxation loss power to the total AC heat generation and the SLP does not depend on the variation of surrounding viscosity of nanofluids. However, the theoretical calculation results have not been used yet to interpret the experimental results. Therefore, in our opinion, the impact of the fluid viscosity performance on the magnetic induced heating (MIH) also needs to be studied in more detail both in calculation and experiment. We believe that these results not only explain the competition between Néel and Brown relaxation in SLP but also establish direction of application in MIH. Moreover, understanding the impact of viscosity on specific loss power is important in making accurate predictions of materials performance under applied AMF, and in determining the various experimental parameters that can be tuned to achieve optimal heat dissipation in tissue.

So, in the present work, the CoFe₂O₄ (CFO) and MnFe₂O₄ (MFO) was synthesized and controlled to have uniformity in size, crystallinity and shape with particle size up to 20 nm. Then, the viscosity of the two nanofluids was varied from 1×10^{-3} to 2×10^{-3} Pa s to study the effects of surrounding viscosity on the empirical SLP characteristics of nanofluids under AC magnetic field. The theoretical SLP was calculated by the Rosensweig equation [12]. By comparing SLP obtained from calorimetric measurement with SLP from theory, we demonstrated that at a given frequency, SLP of magnetic fluids depends on surrounding viscosity by two different ways, in agreement with theoretical predictions.

2. Experimental

CFO and MFO nanoparticles were synthesized by co-precipitation of <u>hexahydrate</u> cobalt chloride (MnCl₂·6H₂O) and tetrahydrate manganese chloride (MnCl₂·4H₂O), ferric chloride hexahydrate (FeCl₃· 6H₂O), ammonium hydroxide (NH₄OH), and hydrochloric acid (HCl) in alkaline. The synthesis procedure is similar to that described in our previous paper[19], except for one condition, namely that the fabrication temperature was 100 °C, 12 h with using autoclave as reaction chamber.

Agar was used as an activator to stabilize the MNPs that were dispersed in water with a concentration of 3 mg/ml. To create the magnetic fluids with different viscosity, 6 ml of the MNPs suspension was mixed with various agar concentrations (0, 0.5, 1, 1.5, and 2 ml for CFO nanoparticles and 0, 0.4, 0.8, 1.2, 1.6, and 2 ml for MFO nanoparticles, respectively), under ultrasonic conditions at room temperature. The mixture of NMPs and agar was then magnetically stirred for 48 h. Rheological characterization of nanofluids was performed by a Sine wave Vibro Viscometer SV 10, featuring the vibrating tuning fork measurement method. It measures viscosity by detecting the driving electric current necessary to resonate two sensor plates at constant frequency of 30 Hz and amplitude of less than 1 mm. The measured viscosities obtained for the 5 CFO and 6 MFO specimens varied in the range from 1 to 2.12 mPa s, as will be shown in Table 2. The structural characterizations of MNPs were determined by X-ray diffraction (XRD) equipment (Siemens, D-5000) using Cu-Ka radiation with wave length λ =1.5406 Å. The diffraction patterns were collected with 2θ in the range of $20^{\circ}-70^{\circ}$. The morphology, particle size and distribution of MNPs were examined with a JEOL JEM-1010 transmission electron microscopy (TEM) operated at an acceleration voltage of 80 kV. The magnetic properties of the magnetic nanoparticle powder were determined by a homemade vibrating sample magnetometer (VSM). The hydrodynamic diameters $(D_{\rm H})$, the poly dispersity index (PDI) of the two nanofluids, CFO and MFO nanoparticles, were measured using a dynamic light scattering (DLS) system. All the calorimetric experiments were carried out using a commercial generator (RDO HFI 5 kW) providing an alternating magnetic field of amplitude 65 Oe, and frequency of 178 kHz. The dependence of SLP of two nanofluids on the viscosity was quantitatively analyzed based on the theoretical and experimental results.

3. Results and discussion

The XRD patterns of the CFO and MFO nanoparticles are shown in Fig. 1. The diffraction peaks at the planes of (220), (311), (222), (400), (422), (511), and (440) confirm the presence of single-phase facecentered cubic structure. The patterns in (a) and (b) are in good agreement with their corresponding standard patterns of $CoFe_2O_4$ (cubic, space group: *Fd3m*, Z =8; ICDD PDF: 22–1086) and MnFe₂O₄ (cubic, space group: *Fd3m*, Z=8; ICDD PDF: 73–1964) [20,21],



Fig. 1. XRD patterns for (a) CFO and (b) MFO nanoparticles.

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