Study of the heating characteristics and mechanisms of magnetic nanoparticles over a wide range of frequencies and amplitudes of an alternating magnetic field

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Abstract. An effective approach to gaining an understanding of the mechanism of heat generation for magnetic hyperthermia from nanomagnet suspensions is to perform heating tests over a wide frequency range. We constructed a heating test apparatus by using ferrite field cores with air gaps for low frequencies and solenoids for high frequencies. Magnetic field amplitudes up to 600 Oe (400 Oe) can be generated for frequencies lower than 500 kHz (800 kHz). Heat generation tests were performed for ferromagnetic nano-platelets and Co-doped Fe_3O_4 nanoparticles over a wide range of frequencies.

1. Introduction

In the biomedical field, application of nanomagnets has attracted attention especially for cancer therapy. For use in magnetic hyperthermia cancer therapy, magnetic nanoparticles are required to have strong heating power. In general, employing an alternating magnetic field (AMF) with high frequencies and amplitudes results in high heat generation; however, there is a limitation on the AMF, as it has a considerable effect on living matter due to eddy currents.[1] Therefore, effective use of the AMF is necessary and we have demonstrated the advantage of using ferromagnetic nanoparticles with cubic symmetry.[2] The major mechanisms of heat generation by magnetic nanoparticles, which include ferromagnetic hysteresis, superparamagnetic (SPM) relaxation (Néel relaxation), and Brownian relaxation, should be analyzed and combined to schieve effective use of AMF.

The heating characteristics of a magnetic nanoparticle suspension over a wide range of AFM frequencies and amplitudes are worth investigating in order to understand and differentiate between these heating mechanisms because the responses of the nanoparticles depend on each mechanism. In this paper, a high-performance magnetic field generator (up to 400–600 Oe) with a wide frequency range (50–800 kHz) was built, and the mechanism of heat generation in several different types of water-based nanoparticle suspensions was examined experimentally.

2. Magnetic field generator

An AMF was generated using series resonant LC circuits. A magnetic field, $H(t) = H_0 \sin(2\pi f_0 t)$, with an amplitude of H_0 was produced at a resonance frequency of f_0. Ferrite cores[3] and solenoids[4] were used as field generators. Commercial ferrite cores (TDK, PC-95) with a 40-mm inner diameter, a 60-nm outer diameter, and a 10-mm air gap were used for frequencies under 200 kHz, and Litz-wire windings with 10–20 turns were formed. For frequencies higher than 200 kHz, solenoid coils with inner diameters of 16 mm were used, and Litz wires (660 wires, each 0.04 mm in diameter) were used to make windings with 100 turns.

The circuit diagram is shown in Fig. 1. The sinusoidal wave from a signal generator was amplified by a 200 W power amplifier (Thamway, T145-5314A). To achieve better impedance matching, a transformer with a 12-turn primary coil and 1-turn secondary coil was used between the power supply and the field generator. The current flowing in the coil was monitored by a Hall-probe current sensor (U-RD, HSC-20-SC-A2.5) with a wide current range.

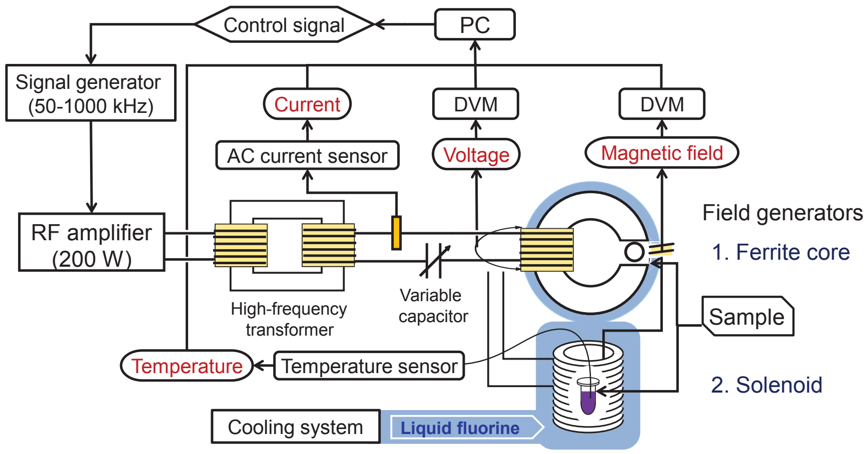

Figure 1. Block diagram of the AMF generator system.

The performance of the field generator system is shown in Fig. 2. The maximum amplitude of the AMF, H_0, decreased almost linearly with increasing operating frequency and was about 600 Oe for frequencies lower than 400 kHz and was 400 Oe for a frequency of 800 kHz. The maximum amplitude is limited by the Joule heating at the coil even though we used Litz wires. The solid line in the figure corresponds to a constant AMF frequencyamplitude product of $f_0 \cdot H_0 = 6.5 \times 10^7$ Hz·Oe; this value represents a recommended safety guideline.[1]

The heating characteristics were measured by setting water suspension samples and a fluorescent thermometer in the space where the AMF existed, and the specific loss power (SLP) was deduced from the initial change in the time variation of the temperature.

3. Heating characteristics of nanoparticles

Two kinds of samples were examined for the heat generation test: dimple-contained nanoparticles (DINP)[5] and Co-doped Fe_3O_4 nanoparticles(SCF)[2]. The DINP have an oval shape with approximate dimensions of 80 x 40 x 10 nm for the long and short axes and the thickness, respectively. The coercive force of a dried sample is around 150 Oe, mostly exhibiting bulk ferromagnetic properties. The SCF particles have a particle size of 20 nm and a coercive force of 145 Oe in a dry state. Water–based suspensions of these particles were made with a particle concentration of 0.9 and 3.7 wt% for DINP and SCF, respectively. These two particles have almost the same coercive force; however, the heating characteristics are different (see Fig. 3).

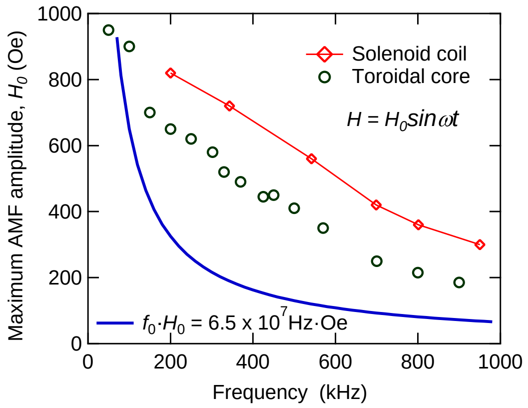

Figure 2. Performance of the AMF generator with a wide frequency range. The solid line shows the safety limit corresponding to a constant value of $f_0 \cdot H_0$.

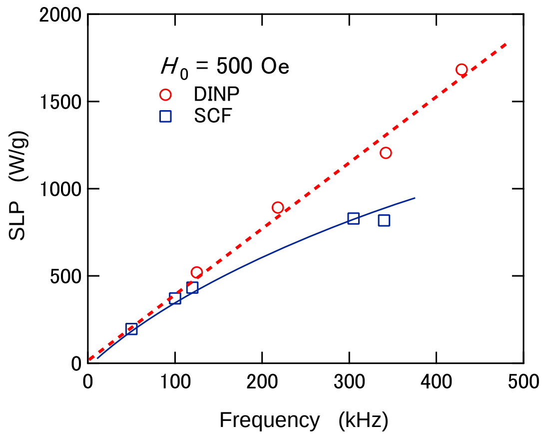

Figure 3. Frequency dependence of the specific loss power for ferromagnetic nano-platelets (DINP)[5] and Co-doped Fe_3O_4 nanoparticles (SCF).

For typical ferromagnetic particles with sufficient coercive force, the major heating mechanisms are hysteresis loss and/or Brownian relaxation. For the case of the ferromagnetic nano-platelets (DINP), because the particle size and the hydrodynamic radius $V_{\rm H}$ are large, the characteristic time for Brownian relaxation, $\tau_{\rm B} = 3\eta V_{\rm H}/k_{\rm B}T$ and $f_{\rm B} = 1/\tau_{\rm B}$ are estimated to be 2×10^{-4} s and 5 kHz, respectively, which is much lower than the test frequencies. For the present case, the viscosity of the suspension, η, is 1 mPa·s. Thus, it was concluded that the heating mechanism was governed by the magnetic hysteresis loss. The power loss Q is simply proportional to the hysteresis area S and the frequency, f_0, such that $Q = f_0 S$. The experimental data[5] clearly shows the linear dependence on the AFM frequency and supports the idea that the ferromagnetic hysteretic mechanism is responsible for the heat generation. This mechanism might be stabilized as a result of the dipolar interaction between ferromagnetic particles even though the concentration is 0.9 wt%

On the other hand, because the SCF sample has a small diameter and a spherical shape, the hydrodynamic radius $V_{\rm H}$ is small, and the characteristic time for Brownian relaxation, $\tau_{\rm B}$, is estimated to be 5×10^{-6}s suggesting that the deviation of the SLP curve from a linear relationship with f_0 may take place at around 200 kHz. This tendency can be seen in Fig. 3 and Co-doped Fe_3O_4 nanoparticles (SCF) can be explained by the combination of magnetic hysteresis and Brownian relaxation, respectively.

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