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Magnetic hyperthermia study in water based magnetic fluids containing TMAOH coated Fe_3O_4 using infrared thermography

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HIGHLIGHTS

- Alternating magnetic field induced heating of water based ferrofluid is studied using IRT.
- IRT based results are compared with that of conventional fiber optic data.
- The temperature rise curves show deviations above some time interval due to the convection phenomena.
- A correction methodology is developed to account for the convection losses.
- Efficacy and applicability of IRT as an alternate real time non-contact temperature measurement technique is established.

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ABSTRACT

We study the alternating magnetic field induced heating of a water based ferrofluid containing tetramethyl ammonium hydroxide coated iron oxide nanoparticles using infrared thermography and compare the results obtained from the conventional fiber optic temperature sensor. Experiments are performed on ferrofluid samples of five different concentrations and under four different external field amplitudes at a fixed frequency. The temperature rise curves measured using both the infrared thermography and fiber optic sensor are found to be very similar up to a certain time interval, above which deviations are observed, which are attributed to the internal and external convection phenomena. A correction methodology is developed to account for the convection losses. The convection corrected specific absorption rate is found to be in good agreement with the values obtained from the conventional fiber optic temperature sensor, within a maximum error of ±3.4%. The highest specific absorption rate obtained in the present study is 135.98 (±4.6) W/g_{Fe} for a sample concentration of 3 wt.%, at an external field amplitude and a frequency of 63.0 kA m^{-1} and 126 kHz, respectively. The specific absorption rate is found to decrease with increasing sample concentration, due to the enhancement of dipolar interaction with increasing sample concentration due to agglomeration. This study validates the efficacy and universal applicability of IRT as an alternate, real time, non-contact and wide area temperature measurement methodology for magnetic fluid hyperthermia experiments without any sample contamination.

1. Introduction

Magnetic fluid hyperthermia (MFH) is a therapeutic procedure, where magnetic fluids are injected into tissues containing cancerous cells and thereafter exposed to a radiofrequency alternating magnetic field, resulting in a temperature rise due to various inherent loss mechanisms such as hysteresis loss, viscous loss and Neel-Brown relaxations [1–4]. Reports show that a rise in cell temperature beyond 42 °C leads to cellular degradation and apoptosis of cancerous cells thereby aiding in cancer treatment [2,5]. MFH based cancer treatment offers several advantages like, in vitro control, external magnetic field driven target localization and high probability of escape from vasculature. Owing to its immense clinical benefits, there has been a rapid development of MFH as an alternate cancer treatment modality in recent years [6–20]. Guller et al. [21] reported that tetramethyl ammonium hydroxide (TMAOH) coated nanoparticles were non-cytotoxic to human dermal fibroblasts and slightly cytotoxic to HaCat keratinocytes, indicating the bio-compatibility of TMAOH coated magnetic nanoparticles. Vaishnava et al. [22] also studied field induced heating of TMAOH coated Fe_3O_4 nanoparticles using conventional temperature measurement probes.

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In the absence of an external magnetic field, the magnetic moments of the dispersed magnetic nanoparticles are randomly oriented along their easy magnetic axes. On application of an external magnetic field, the moments are aligned along the direction of the magnetic field, thereby storing magnetic energy, which is released in the form of heat when the applied magnetic field is withdrawn, allowing the moments to reorient themselves along their easy axes. This process is known as Neel relaxation and assuming uniaxial anisotropy, Neel relaxation time (τ_N) [23,24] can be expressed as $\tau_N = \frac{\tau_0}{2}\sqrt{\frac{\pi}{\sigma}}e^{\sigma}$, where τ_0 is the attempt frequency and can be expressed as $\tau_0 = \frac{M_s}{2\gamma_0 K}\frac{(1+\alpha^2)}{\alpha}$, where M_s, γ_0, K and α are saturation magnetization, electron gyromagnetic ratio, effective uniaxial magnetic anisotropy energy density and linear response damping function, respectively [25]. σ indicates the ratio of magnetic anisotropy energy to thermal energy and is expressed as $\sigma = KV_p/k_BT$, where V_p, k_B and T indicate, particle volume, Boltzmann's constant and absolute temperature, respectively. On the other hand, in Brownian relaxation, heating is achieved by whole-scale rotation of the magnetic particle in the surrounding medium by shear action keeping the orientation of the magnetic moments remain fixed with respect to the crystal direction. The Brownian relaxation time (τ_B) can be expressed as $\tau_B = 3\eta V_h/k_BT$, where η and V_h are the viscosity of the carrier fluid and hydrodynamic volume of the dispersed magnetic nanoparticles [24]. In practice, both relaxation mechanism occur simultaneously and the effective relaxation time (τ) is given by $\tau^{-1} = \tau_N^{-1} + \tau_B^{-1}$ [2]. In the linear response regime ($\mu_0 M_s V_p H_{max} < k_BT$, where μ_0 and H_{max} indicate magnetic permeability of free space and maximum applied field amplitude, respectively), the field induced heating of magnetic nanofluidis is due to the out of phase component of complex susceptibility. According to Rosensweig's model, power (P) dissipated per unit mass is given by the following equation [1].

$$P = \frac{1}{2}\omega\mu_0\chi_0 H_0^2 \frac{\omega\tau}{1+\omega^2\tau^2} \quad (1)$$

Here, ω ($\omega = 2\pi f$, f being frequency of the alternating magnetic field) and χ_0 indicate cyclic frequency and initial static susceptibility, respectively. The field induced heating of magnetic nanofluids are quantified in terms of a dosimetric quantity known as specific absorption rate (SAR = P/density) which is defined as the heat generated per unit mass (W/g). Experimental estimation of SAR depends on the accurate measurement of field induced temperature rise in magnetic fluid specimens as a function of time and the initial rate of temperature rise can be calculated using phenomenological Box-Lucas [12,15], exponential rise [15,26], linear regression analyses [11,17,18] or the corrected slope [27] techniques. Normally, the fluid temperature at a single location is measured using radiofrequency immune fiber optic temperature sensor, or infrared and liquid thermometers. Metallic thermometers or thermocouples are not suitable for measuring fluid temperature during MFH experiments due to the problems associated with eddy current induced self-heating [16]. Recently, we have studied the field induced temperature rise directly on an oil based magnetic fluid sample containing oleic acid coated Fe_3O_4 nanoparticles using infrared thermography (IRT), where the data was found to be in good agreement with those obtained from conventional fiber optic temperature sensor and the convection corrected SAR values obtained from IRT were found to be within ±5% of that obtained using a fiber optic temperature sensor [16]. Rodrigues et al. [28] have earlier studied the temperature rise in magnetic nanofluid during alternating magnetic field (field amplitude ~1.10 kA m^{-1} and frequency = 200 kHz) induced heating and showed the applicability of IRT in such studies.

IRT is a non-contact temperature measurement methodology where the infrared rays emitted by the surface is detected by an infrared detector and surface temperature of the object is estimated from the intensity of the emitted radiation using the following radiometric equation [29].

$$I_{cam} = T_r\varepsilon I_{obj} + T_r(1-\varepsilon)I_{env} + (1-T_r)I_{atm} \quad (2)$$

Here, I_{cam} is the radiance received by the infrared camera. I_{obj}, I_{env} and I_{atm} indicate the radiance emitted by the object under investigation, surrounding environment and atmosphere, respectively. T_r and ε are the atmospheric transmittance and surface emissivity of the object, respectively. For laboratory scale distance, atmospheric transmittance can be approximated as unity and hence, Eq. (2) can be simplified as $I_{cam} = \varepsilon I_{obj} + (1-\varepsilon)I_{env}$. The emissivity of a hypothetical perfect blackbody is unity and for real surfaces, $\varepsilon < 1$ [29]. The radiance received by the infrared detector is converted into an electrical signal and the object temperature is measured using suitable calibration curves [29]. IRT is routinely used in the field of non-destructive evaluation [29–31], condition monitoring [32] and bio-medical applications [33–35]. Though the application of IRT during MFH experiments has been reported in a few studies [27,28,36–39], the temperature is not directly measured on the magnetic fluid samples.

Owing to its advantages over the conventional point probe temperature measurement, such as simultaneous temperature measurement on several locations, non-contact and non-invasive temperature measurements, and reduced probability of sample contamination due to non-contact measurement, the application of IRT in MFH is likely to grow in future. For universal acceptance as a remote temperature measurement probe, the IRT based temperature measurement methodology needs to be validated for water based magnetic fluids, which is the prime objective of this work. Here we report the IRT based temperature measurement during field induced heating of a water based stable ferrofluid containing tetramethyl ammonium hydroxide (TMAOH) coated Fe_3O_4 nanoparticles. A convection correction procedure to match with the temperature rise curves, obtained using optical fiber temperature sensor, is developed. Variation of SAR with sample concentration and the effect of dipolar interaction is also studied.

2. Materials and experimental methods

2.1. Synthesis and characterization

Fe_3O_4 nanoparticles were prepared by chemical coprecipitation technique using 1:1 ratio of 0.8 M $FeCl_3\cdot 6H_2O$ and 0.4 M $FeSO_4\cdot 7H_2O$. The detailed synthesis method can be found elsewhere [40]. The synthesized Fe_3O_4 nanoparticles were later coated with tetramethyl ammonium hydroxide (TMAOH) surfactant to stabilize the particle electrostatically. A master solution of 8 wt.% was prepared, which was later diluted to 6, 4, 3 and 2 wt.%.

The synthesized magnetic nanoparticles were characterized using powder X-ray diffraction (XRD), dynamic light scattering (DLS) and vibrating sample magnetometer (VSM) [40]. XRD data indicated an average crystallite diameter of 8.0 ± 0.9 nm, whereas DLS showed an average hydrodynamic size of 10 (±1) nm. The saturation magnetization measured using VSM was found to be 42 emu/g (without correction to dead layer contribution) which is much lower than the saturation magnetization of bulk magnetite (92 emu/g) [40].

2.2. Hyperthermia measurements

Hyperthermia measurements were performed using a high frequency induction heating system (Five Celes, France), equipped with water cooled electrolytic copper made 50 mm diameter coils with 6 numbers of turns. The TMAOH coated magnetic nanoparti-