Heating magnetic fluid with alternating magnetic field

R.E. Rosensweig*

Exxon Research and Engineering Co. (ret.), 34 Gloucester Rd., Summit, NJ 07901, USA

Abstract

This study develops analytical relationships and computations of power dissipation in magnetic fluid (ferrofluid) subjected to alternating magnetic field. The dissipation results from the orientational relaxation of particles having thermal fluctuations in a viscous medium.
© 2002 Elsevier Science B.V. All rights reserved.

Keywords: Magnetic fluid; Ferrofluid; Heating; Medical hyperthermia

1. Introduction

It appears that heating of colloidal magnetic fluid (ferrofluid) due to time-varying magnetic induction has not been significantly studied. Heating ferrofluid to achieve hyperthermia in medical treatments is an emerging area of importance [1,2]. Heating effects in ferrofluids may also be important in loudspeakers where temperature rise adversely affects performance [3]. Other applications exploit the ability to heat the fluid magnetically, as in the thermal actuation of a polymer gel in contact with a ferrofluid.

This work develops dissipation relationships based on rotational relaxation of single domain magnetic particles dispersed in a liquid matrix. Eddy current heating is assumed negligible due to the small size of the particles (<15 nm). The SI system of units is employed.

2. Power dissipation

From the first law of thermodynamics for a constant density system of unit volume $dU = \delta Q + \delta W$, where U is the internal energy, Q the heat added and W the magnetic work done on the system. For an adiabatic process $\delta Q = 0$ with the differential magnetic work, given in general [4] by $\delta W = \mathbf{H} \cdot d\mathbf{B}$, the

result is

$$dU = \mathbf{H} \cdot d\mathbf{B}, \tag{1}$$

where $\mathbf{H}$ (A m^{-1}) is the magnetic field intensity and $\mathbf{B}$ (T) the induction, both in the sample. Because the fields are colinear the relationship reduces to $dU = H\,dB$ where H and B are magnitudes. $B = \mu_0(H + M)$, where M (A m^{-1}) is the magnetization and $\mu_0 = 4\pi \times 10^{-7}$ (T m A^{-1}) is the permeability of free space. Substitution into Eq. (1) followed with integration by parts shows that the cyclic increase of internal energy can be written as

$$\Delta U = -\mu_0 \oint M\,dH. \tag{2}$$

When magnetization lags the field, the integration yields a positive result indicating conversion of magnetic work to internal energy. It will be convenient to express the magnetization in terms of the complex ferrofluid susceptibility $\chi = \chi' - i\chi''$. Then with an applied magnetic field of the form

$$H(t) = H_0 \cos \omega t = Re[H_0 e^{i\omega t}], \tag{3}$$

the resulting magnetization is

$$M(t) = Re[\chi H_0 e^{i\omega t}] = H_0(\chi' \cos \omega t + \chi'' \sin \omega t), \tag{4}$$

where it is seen that χ' is the in-phase component, and χ'' the out-of-phase component of χ. Substituting for M and H in Eq. (2) from Eq. (4) leads to

$$\Delta U = 2\mu_0 H_0^2 \chi'' \int_0^{2\pi/\omega} \sin^2 \omega t\,dt, \tag{5}$$

*Tel.: +1-908-277-2846; fax: +1-908-277-1084.
E-mail address: rerosen@comcast.net (R.E. Rosensweig).

where only the component χ'' survives, hence its name, the loss component. Integrating and multiplying the result by the cyclic frequency $f = \omega/2\pi$ gives the following expression for volumetric power dissipation:

$$P = f\,\Delta U = \mu_0 \pi \chi'' f H_0^2. \tag{6}$$

To make use of this result χ'' must be related to material parameters of the ferrofluid.

3. Relationship to material parameters

The relaxation equation of Shliomis [5] reduces to the following form for motionless fluid in an oscillatory field:

$$\frac{\partial M(t)}{\partial t} = \frac{1}{\tau}(M_0(t) - M(t)), \tag{7}$$

where τ is the relaxation time, $M_0 = \chi_0 H_0 \cos \omega t = Re(\chi_0 H_0\,e^{i\omega t})$ is the equilibrium magnetization in the applied field whose value is given in Eq. (3), and χ_0 is the equilibrium susceptibility. Substituting the complex representations of M_0 and $M(t)$ into Eq. (7) yields

$$\chi = \frac{\chi_0}{1 + i\omega\tau}, \tag{8}$$

which gives the dependence of complex susceptibility on frequency and from which the components of susceptibility are

$$\chi' = \frac{\chi_0}{1 + (\omega\tau)^2}, \tag{9a}$$

$$\chi'' = \frac{\omega\tau}{1 + (\omega\tau)^2}\chi_0. \tag{9b}$$

These relationships are identical to the Debye spectra of polar molecules in the absence of a constant field [6]. The spectra for monodisperse particles are plotted in Fig. 1 which shows that the real and imaginary components have values that cross at the peak value of χ''. The data of Fanin [7] display this trend. As it follows from Eq. (9b), the predicted cross-over occurs at $\omega\tau = 1$. Also shown for comparison is the computed spectra for a polydispersion of magnetic particles assuming a log normal distribution of particle size with standard deviation $\sigma = 0.1$ as discussed in Section 5. The polydisperse curves are shifted greatly showing why measurements of actual magnetic fluids, which are nearly all polydisperse, vary from the monodisperse predictions. Ferromagnets exhibit magnetic resonance at frequencies $\sim 10^8$–10^{10} Hz yielding a change of sign to negative value of $\chi'(\omega)$ and a sharp peak of $\chi''(\omega)$ [8]. Here interest is limited to the megahertz frequency range and below; hence, no further discussion is devoted to the higher frequency aspects.
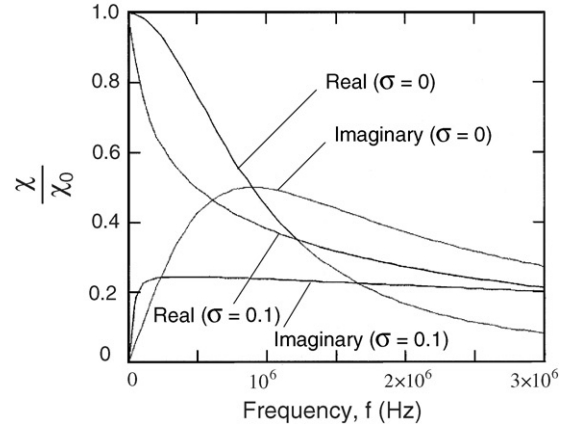

Fig. 1. Susceptibility components of monodisperse particles compared to that of a polydispersion. $B_0 = 0.06$ T, $R = 4.45$ nm.

4. Time constants

With the Brownian mechanism of relaxation the magnetic moment is locked to the crystal axis and when the magnetic moment aligns with the field, the particle rotates as well. A second mechanism exists (Néel relaxation) in which the magnetic moment rotates within the crystal. To achieve high heating rates the Néel relaxation must not be allowed to dominate.

The Brownian time constant is given by the following relationship [9]:

$$\tau_B = \frac{3\eta V_H}{kT}, \tag{10}$$

where η is the viscosity coefficient of the matrix fluid, k the Boltzmann constant (1.38×10^{-23} J K^{-1}), and T the absolute temperature (K). V_H is taken as the hydrodynamic volume of the particle which is larger than the magnetic volume $V_M = 4\pi R^3/3$ for a particle of radius R. As a model for V_H, it is assumed that $V_H = (1 + \delta/R)^3 V_M$, where δ is the thickness of a sorbed surfactant layer.

The Néel relaxation time, denoted τ_N, is given by the following expression due to Brown [10],[1] where $\Gamma = \mathscr{K} V_M/kT$ with $\mathscr{K}$ as the anisotropy constant which may be of magnetocrystalline or shape origin:

$$\tau_N = \frac{\sqrt{\pi}}{2}\tau_0\frac{\exp \Gamma}{\Gamma^{1/2}} \tag{11}$$

or in the equivalent form

$$\tau_N = \frac{\sqrt{\pi}}{2}\tau_D\frac{\exp \Gamma}{\Gamma^{3/2}} \tag{12a}$$

$$\tau_D = \Gamma\tau_0. \tag{12b}$$

---

[1] The author thanks M.I. Shliomis for pointing out the alternate expressions.

Because $\tau_0 \sim \mathscr{K}^{-1}$ while $\tau_D \sim V_M$, Eq. 11 is used when determining the size dependence of τ_N, where V_M enters only in Γ. Alternatively, when considering how τ_N depends on $\mathscr{K}$, Eq. (12) is used where $\mathscr{K}$ is contained only in Γ.

A typical ferrofluid has a broad distribution of particle sizes with a mean size of ~ 10 nm. Because the Brownian and Néel processes take place in parallel, the effective relaxation time τ is given by

$$\frac{1}{\tau} = \frac{1}{\tau_B} + \frac{1}{\tau_N}. \tag{13}$$

The plot shown in Fig. 2 illustrates that the shorter time constant tends to dominate in determining the effective relaxation time for any given size of particle.

From Eqs. (6) and (9b) the power dissipation is expressed as

$$P = \pi \mu_0 \chi_0 H_0^2 f \frac{2\pi f \tau}{1 + (2\pi f \tau)^2}. \tag{14}$$

Eq. (14) expresses the power dissipation density for a monodispersion, assuming susceptibility is constant. Actual susceptibility χ_0 is magnetic field dependent. As a conservative estimate giving a lower bound of P, it is assumed that χ_0 is the chord susceptibility corresponding to the Langevin equation $L(\xi) = M/M_s = \coth \xi - 1/\xi$ where $\xi = \mu_0 M_d H V_M / kT$. $M_s = \phi M_d$ is saturation magnetization of the ferrofluid, M_d the domain magnetization of a suspended particle, and ϕ the volume fraction solids. Thus,

$$\chi_0 = \chi_i \frac{3}{\xi} \left(\coth \xi - \frac{1}{\xi} \right), \tag{15}$$

where the initial susceptibility is $\chi_i = (\partial M / \partial H)_i = \mu_0 \phi M_d^2 V_m / 3kT$ as determined from differentiation of the Langevin relationship. Temperature rise for the monodispersion is computed as $\Delta T = P \Delta t / c$, where c is the ferrofluid specific heat and Δt is the duration of the heating. Specific heat is calculated as the volume average of solid and liquid constituents.

5. Polydispersions

It is found that the log normal particle size distribution $g(R)$ provides a reasonably good fit to the measured distribution for ferrofluids [11]. $g(R)$ is obtained by substituting $\ln(R)$ for R in a Gaussian distribution function, thereby circumventing the unphysical aspect of contributions from negative particle sizes associated with the Gaussian:

$$g(R) = \frac{1}{\sqrt{2\pi}\sigma R} \exp \left[\frac{-(\ln R/R_0)^2}{2\sigma^2} \right], \tag{16a}$$

$$\int_0^\infty g(R)\,\mathrm{d}R = 1, \tag{16b}$$

where $\ln R_0$ is the median and σ the standard deviation of $\ln R$. The most probable radius is given by $R_0 \exp(-\sigma^2/2)$ and the mean radius is $R_0 \exp(\sigma^2/2)$.

Numerical integration over the distribution function transforms P of Eq. (14) to $\bar{P}$, the volumetric heat release rate of a polydispersion:

$$\bar{P} = \int_0^\infty P g(R)\,\mathrm{d}R. \tag{17}$$

The corresponding temperature rise of an adiabatic sample is $\Delta \bar{T} = \bar{P} \Delta t / c$.

6. Calculated results

The integrand of Eq. (17) is a huge expression when the expressions for $g(R), P, \chi$ and τ are substituted in it, and analytical integration is not desirable. Instead, numerical values of temperature rise are calculated as examples. These are based on physical property values listed in Table 1. A value of $\tau_0 = 10^{-9}$ s is employed throughout.

Fig. 3a illustrates the heating rate for monodisperse magnetite samples in a hydrocarbon carrier as a function of particle radius and field intensity $(B_0 = \mu_0 H_0)$. *Very substantial heating rates are achievable.* An optimum particle size yielding maximum heating exists that is nearly independent of the applied field intensity. Fig. 3b shows the influence of carrier viscosity on the heating rate with frequency as parameter for a sample with a fixed particle size. It is seen that heating rates peak at viscosity values lower than the usually available ones.
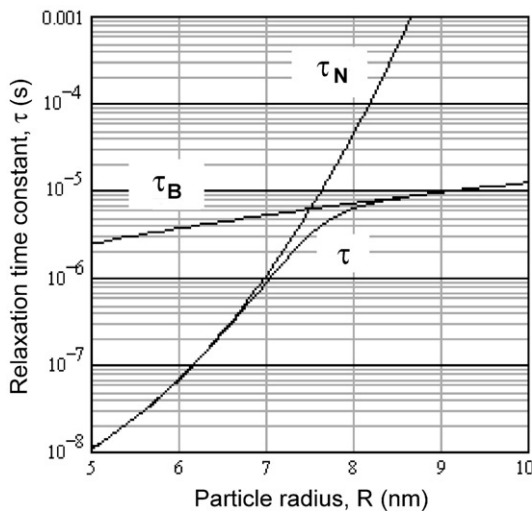

Fig. 2. Time constants vs. particle size for magnetite particles.

Table 1
Physical properties of magnetic solids

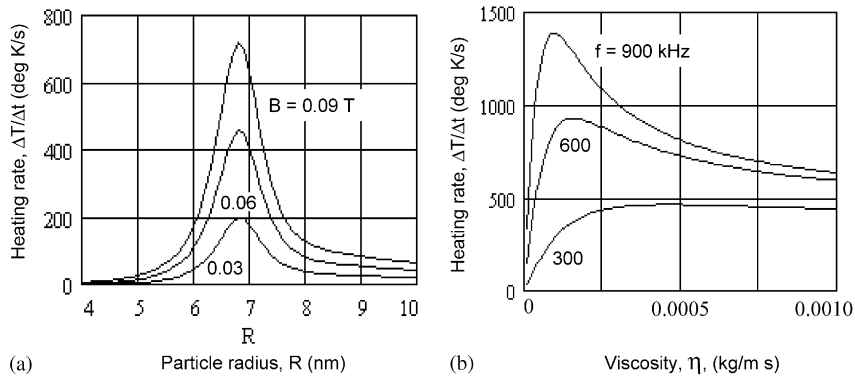| Magnetic solid	Chemical formula	M_d (kA m^{-1})	K (kJ m^{-3})	c (J kg^{-1} K^{-1})	ρ (kg m^{-3})
Maghemite	γ-Fe$_2$O$_3$	414	−4.6	∼746	4600
Magnetite	FeO·Fe$_2$O$_3$	446	23–41	670	5180
Cobalt ferrite	CoO·Fe$_2$O$_3$	425	180–200	700	4907
Barium ferrite	BaO·6Fe$_2$O$_3$	380	300–330	∼650	5280

Fig. 3. Temperature rise rate in monodisperse ferrofluid based on magnetite with $\phi = 0.071$: (a) Tetradecane carrier, frequency 300 kHz; (b) Magnetic induction 0.06 T, $R = 7$ nm.
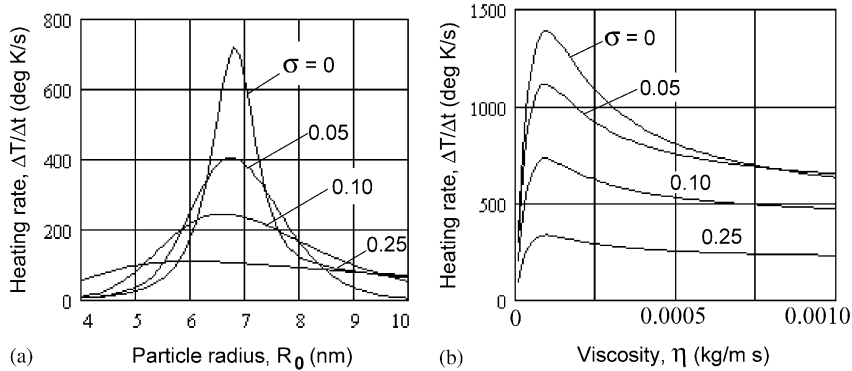

Fig. 4. Temperature rise rate in polydisperse ferrofluid based on magnetite with $\phi = 0.071$: (a) Tetradecane carrier, $f = 300$ kHz; (b) $B_0 = 0.06$ T, $f = 900$ kHz, $R = 7$ nm.

The degradative influence of polydispersity is illustrated in Fig. 4 for magnetite particles in hydrocarbon carrier, showing the incentive to utilize highly monodisperse samples. The calculations are done for a tetradecane carrier fluid having specific heat of 2080 J kg^{-1} K^{-1}, mass density 765 kg m^{-3} and viscosity 0.00235 kg m^{-1} s^{-1}. Surfactant layer thickness is set at 2.0 nm.

Fig. 5 compares heating rates for monodispersions of the various magnetic solids listed in Table 1 assuming $\tau_0 = 10^{-9}$ s. The carrier liquid is tetradecane in all cases. The main differences between the performance of the resultant ferrofluids are due to the anisotropy constants and the domain magnetizations. From the figure it can be seen that barium ferrite and cobalt ferrite yield the largest heating rates in the size range of typical
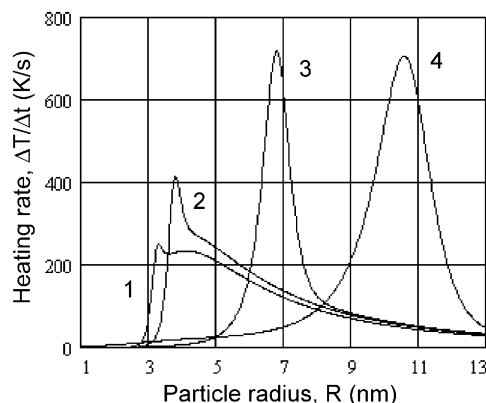
Fig. 5. Comparative heating rates for various magnetic solids. 1—Barium ferrite, 2—Cobalt ferrite, 3—Magnetite, 4—Maghemite. ($\phi = 0.071$, $\eta = 0.00235\,\mathrm{kg\,m^{-1}\,s^{-1}}$, $f = 300\,\mathrm{kHz}$, $B = 0.09\,\mathrm{T}$, $\delta = 2\,\mathrm{nm}$).

ferrofluids ($R \sim 4 - 5\,\mathrm{nm}$). The highest heating rates are provided by magnetite and maghemite; however, the size range where this occurs is larger than that found in stable ferrofluids.

7. Conclusion

This study utilizes a collection of well-known principles in magnetic fluid science to formulate and compute heating rate in samples subjected to an alternating magnetic field. The methodology should be of interest to investigators in numerous fields of technology.

References

[1] A. Jordan, P. Wust, H. Fahling, W. Hohn, A. Hinz, R. Felix, Int. J. Hyperthermia 9 (1) (1993) 51.
[2] R. Hiergeist, W. Andra, N. Buske, R. Hergt, I. Hilger, U. Richter, W. Kaiser. J. Magn. Magn. Mater. 201 (1999) 420 (volume issued out of sequence).
[3] K. Raj, in: B. Berkovski (Ed.), Magnetic Fluids and Applications Handbook, Begell House Inc., New York, 1996, pp. 716–724.
[4] J.A. Stratton, Electromagnetic Theory, McGraw-Hill, New York, 1941, p. 124.
[5] M. Shliomis, Sov. Phys. Uspekhi (Engl. transl.) 17 (2) (1974) 153.
[6] P. Debye, Polar Molecules, The Chemical Catalog Co., Reprinted by Dover Publications, New York, 1929.
[7] P.C. Fanin, in: B. Berkovski (Ed.), Magnetic Fluids and Applications Handbook, Begell House Inc., NewYork, 1996, p. 170.
[8] Y.L. Raikher, M.I. Shliomis, in: W. Coffey (Ed.), Relaxation Phenomena in Condensed Matter, Advances in Chemical Physics Series, Vol. LXXXVII, Wiley, New York, 1994, pp. 596–751.
[9] J. Frenkel, The Kinetic Theory of Liquids, Dover Publications, New York, 1955.
[10] W.F. Brown, Jr., Phys. Rev. 130 (1963) 1677.
[11] R.W. Chantrell, J. Popplewell, S.W. Charles, IEEE Trans. Magn. MAG-16 (1978) 975.