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Evaluation of Relaxational and Hysteretic Heat Losses in Concentrated Magnetic Fluid under Influence of Alternating Magnetic Field

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Abstract This article describes the magnetic and hyperthermic investigation of a concentrated magnetic fluid on the basis of a transformer mineral oil with magnetite particles. The structure of a ferrofluid and general magnetic parameters of a medium were determined from vibrating sample magnetometer measurements. The hyperthermic experiments were carried out at a frequency $f = 1500\,\mathrm{kHz}$ for various magnetic field amplitudes. The analysis of calorimetric results allowed an estimate of the contribution of relaxational and hysteresis loss mechanisms in total energy losses in the hyperthermic effect under the influence of an alternating magnetic field. Additional calorimetric measurements were also performed (at $H_C = 1500\,\mathrm{A \cdot m^{-1}}$) versus frequency in the range from 50 kHz to 2 MHz which indicated that for $f_o = 653\,\mathrm{kHz}$ the heating process is the most effective.

Keywords Hyperthermia · Magnetic fluid characterization · Relaxation and hysteresis losses · Specific absorption rate · Vibrating sample magnetometer (VSM)

1 Introduction

For the case of placing a magnetic fluid (MF) into an external alternating magnetic field, the magnetic vector M in that sample undergoes reorientation in space following the magnetic field intensity vector H. As a result of the phase delay of the magnetic vector M in relation to the vector H, a part of the magnetic field energy undergoes irreversible conversion into thermal energy. It leads to heating of the MF sample, which can be used in biomedicine for therapy that destroys cancerous cells. An important

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medical application of a biocompatible MF is so-called hyperthermia, which relies on the heating of specific body organs or tissues to temperatures of 41 °C to 46 °C.

An MF is even applicable to the power industry, particularly in high-voltage devices (in construction of transformers, high-voltage switches). In that case, an MF based on a high-quality mineral (or synthetic) oil with a small [1,2] or average [3] concentration of magnetic particles is used. The purpose of the MF application in transformers is the assurance of good electric insulation and the facilitation of heat removal from windings to surroundings. In practice, there exist a few reasons for alternating magnetic field energy loss and its exchange for thermal energy. If the fluid contains only superparamagnetic particles, magnetic relaxation appears and such a sample magnetizes itself according to two mechanisms: by Brown and by Néel mechanisms. Each of these mechanisms is defined by the appropriate average relaxation time according to well-known expressions,

$$\langle \tau_{\mathrm{B}} \rangle \;=\; \frac{3\langle V_{\mathrm{H}} \rangle}{k_{\mathrm{B}} T} \eta_{\mathrm{S}} \tag{1a}$$

and

$$\langle \tau_{\mathrm{N}} \rangle \;=\; \tau_{\mathrm{o}} \exp\left(\frac{K \langle V \rangle}{k_{\mathrm{B}} T} \right), \tag{1b}$$

where V_{H} is the hydrodynamic volume of the magnetic grain, $k_{\mathrm{B}} = 1.38 \times 10^{-23}\,\mathrm{J \cdot K^{-1}}$ is the Boltzmann constant, T is the absolute temperature, η_{S} is the shear viscosity coefficient of mineral oil (at $T = 298\,\mathrm{K}$, $\eta_{\mathrm{S}} = 0.01\,\mathrm{Pa \cdot s}$), τ_{o} is the time constant ($\cong$1 ns), $K = 23 \times 10^3\,\mathrm{J \cdot m^{-3}}$ is the anisotropy constant for magnetite, and V is the volume of the magnetic grain.

The averaging is a result of magnetic particle polydispersion, which most often is described by a distribution function of the log-normal type:

$$f(d) \;=\; \frac{1}{\sqrt{2\pi}\beta d} \exp\left[\frac{-\ln^2\left(\frac{d}{d_{\mathrm{o}}}\right)}{2\beta^2} \right], \tag{2}$$

where d_{o} and β are parameters of the function $f(d)$.

Because of MF polydispersion, both mechanisms appear most often simultaneously, and in effect, the resultant relaxation time is

$$\tau_{\mathrm{eff}} \;=\; \frac{\tau_{\mathrm{B}} \tau_{\mathrm{N}}}{\tau_{\mathrm{B}} + \tau_{\mathrm{N}}}. \tag{3}$$

The mean volumetric power dissipation at frequency f is given by [4]

$$P = \pi \mu_{\mathrm{o}} \chi^{//} f H_{\mathrm{o}}^2 \tag{4}$$

where $f = \omega/2\pi$ is the frequency of the alternating magnetic field, $\chi^{//}$ is the imaginary part of the magnetic susceptibility, and $\mu_{\mathrm{o}} = 4\pi \times 10^{-7}\,\mathrm{H \cdot m^{-1}}$.

From Eq. 4, it follows that in the sample containing only the superparamagnetic particles, at a given frequency f, the density of the releasing thermal power should be proportional to the square of the magnetic field intensity amplitude: $P \propto H^2$.

The imaginary part of the magnetic susceptibility is a function of both the frequency and magnetic field intensity H_o according to the following equation [5]:

$$\chi^{//} = \chi_o \frac{\omega\tau}{1+(\omega\tau)^2} = \frac{\mu_o \phi M_S^2 V}{k_B T} \frac{L(\xi)}{\xi} \frac{\omega\tau}{1+(\omega\tau)^2}, \tag{5}$$

where $\chi_o = 3\chi_i(\coth\xi - \xi^{-1})\xi^{-1}$ is the equilibrium susceptibility of a magnetic fluid, $\chi_i = (\partial M/\partial H)_i$ is the initial susceptibility determined from differentiation of the Langevin relationship [4], ϕ_V is the volume fraction of particles in the suspension, M_S is the saturation magnetization of the magnetic material, and $\xi = \mu_o M_S V H/(kT)$ is the parameter of the Langevin function $L(\xi)$.

From Eq. 5, it follows that for $\omega\tau = 1$, the imaginary part of the magnetic susceptibility $\chi^{//}$ achieves the maximal value.

Because of polydispersion, magnetic particles with small sizes are located in the MF sample, being the source of relaxation losses, but there are also particles of larger sizes, which are the source of energy losses for hysteresis. From Refs. [6–9], it follows that for particle systems with ferromagnetic behavior (i.e., hysteresis), a power of three is found at low field amplitudes (for Rayleigh losses). Thus, hysteresis losses for so-called Rayleigh loops may be well described by a third-order power law. In that case, it can be written as $P \propto H^3$.

The purpose of the present research was determination of what part in releasing thermal energy have the losses been caused by magnetic relaxation and what part of energy losses are caused by magnetic hysteresis in an MF with a large concentration of magnetic particles. Similar research has been described previously [10], but that research concerned fluids on the basis of water with a small concentration of magnetic particles $CoFe_2O_4$.

2 Magnetic Properties

Magnetic investigations of an MF sample were carried out with the aid of the vibrating sample magnetometer (VSM) technique, which provided a range of valuable information about the MF structure on the basis of transformer mineral oil with magnetite grains of Fe_3O_4. The surfactant of the fluid was a single layer formed from oleic acid particles.

For lower magnetic field intensities, the measurements were carried out with the aid of electromagnet coils. In Fig. 1a and b there are shown, for example, magnetic hysteresis loops obtained in both sets at values of the magnetic field with an induction amounting to $\Delta B = \pm 20\,\text{mT}$ and $\pm 100\,\text{mT}$. The surface area enclosed by the magnetic hysteresis loop is a measure of the thermal energy W_h released in the magnetic material during a single magnetization cycle.

In turn, in Fig. 2 the curve of primary magnetization is shown along with the distribution function of magnetite grain sizes.

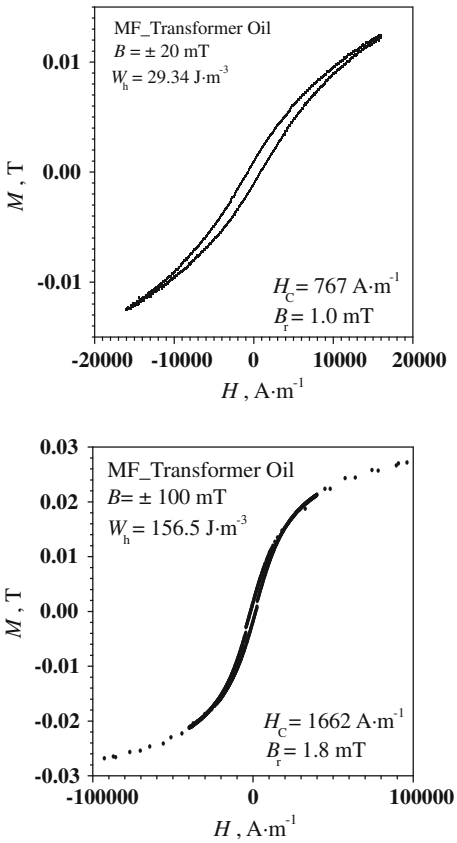

Fig. 1 Examples of magnetic hysteresis loops obtained during sample magnetization. (**a**) In Helmholz's coils. (**b**) In the gap of an electromagnet

From a fit of the log-normal function to the experimental data, the values of the parameters $d_o = 10.6\,\text{nm}$ and $\beta = 0.5$ were obtained. Having these values, the average grain size $\langle d \rangle = 12\,\text{nm}$ and the standard deviation $\sigma = 6.4\,\text{nm}$ were calculated from [11]

$$\langle d \rangle = d_o e^{\frac{\beta^2}{2}} \quad \text{and} \quad \sigma = d_o \exp\left(\frac{\beta^2}{2}\right)\sqrt{\exp^{\beta^2} - 1}. \tag{6}$$

For the mean-spherical model [12], the asymptotic formula for magnetization at $\xi \gg 1$ is expressed as

$$M(H) = M_S\left(1 + \frac{P}{3\mu_o H^2}\right) - \frac{P}{\mu_o H}, \tag{7}$$

where M_S is the saturation magnetization of the ferrofluid and $P = nkT$ is the partial pressure of the colloid particles.

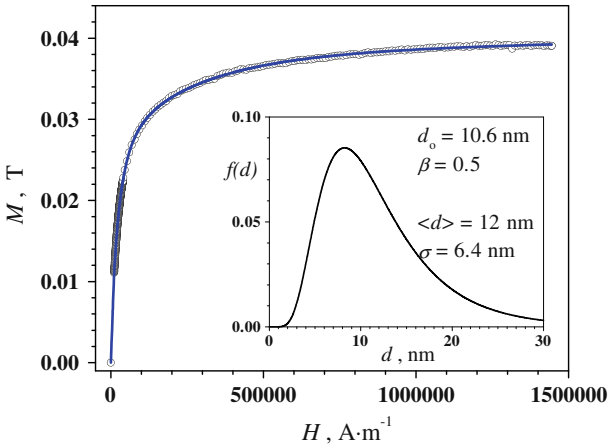

Fig. 2 Primary magnetization curve of an MF sample along with the distribution function of magnetite grain sizes

In that case, from the fit of this function to the experimental data (for $\xi \gg 1$), the following values were obtained: $M_S = 29.1\,\mathrm{kA \cdot m^{-1}}$ and $P = 1630\,\mathrm{Pa}$. For a temperature $T = 298\,\mathrm{K}$, the number of magnetic grains in a unit volume equals $n = 4 \times 10^{23}\,\mathrm{m^{-3}}$.

In turn, for the high-temperature approximation model [12],

$$M(H) = \left(M_S - \frac{P}{\mu_o H}\right)\left(1 + \frac{P}{3\mu_o H^2}\right), \tag{8}$$

the author obtained similar values: $M_S = 29.17\,\mathrm{kA \cdot m^{-1}}$, $P = 1751\,\mathrm{Pa}$, and $n = 4.3 \times 10^{23}\,\mathrm{m^{-3}}$.

Taking into account that the spontaneous magnetization of magnetite grains $M_{gr} = 446\,\mathrm{kA \cdot m^{-1}}$ [4], and the measured value $M_S \cong 29.13\,\mathrm{kA \cdot m^{-1}}$, the magnetite concentration in the MF was calculated as $\phi_V \cong 6.5\,\%$. The value of $M_{gr} = 446\,\mathrm{kA \cdot m^{-1}}$ determining the magnetic properties of magnetite nanoparticles is lower than $477\,\mathrm{kA \cdot m^{-1}}$. The latter is appropriate for a bulk solid sample. The spontaneous magnetization decrease of the magnetite grains results from the fact that there is an external layer around the nanoparticles, which is magnetically neutral.

3 Results of Calorimetric Experiments and Analysis

Figure 3 presents time changes of the magnetic fluid sample temperature, for a sample subjected to an alternate magnetic field of different intensity H_{AC} and frequency of $f = 1500\,\mathrm{kHz}$, over a range of time (0 to 10 min).

The revealed heating effect is very distinct, although the values of the applied magnetic field intensity were relatively low. It seems that the quite high frequency and concentration of magnetite in the MF are of great importance here.

The heating system consisted of a sine wave power oscillator, an induction coil (length of 78 mm), and an oscilloscope (Philips). A glass tube containing the sample

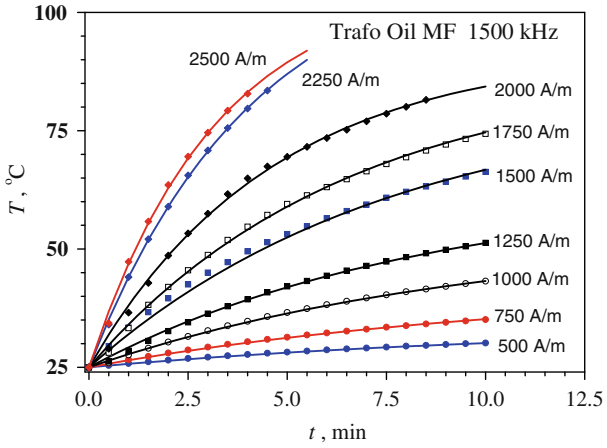

Fig. 3 Measured temperature increase versus time for various intensities of magnetic fields at a frequency $f = 1500\,\text{kHz}$

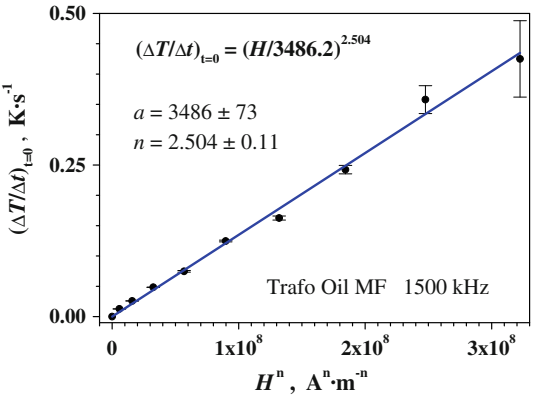

Fig. 4 Rate of temperature increase $(\mathrm{d}T/\mathrm{d}t)_{t=0}$ at the beginning of the heating as an exponential function of magnetic field H^n. *Circles* (•) stand for experimental values, and the *solid line* (–) is the function (Eq. 9) of the fit

was thermally isolated by a layer of material from the solenoid winding supported on a plastic sleeve. The temperature of the sample was monitored using a thermocouple (LT Lutron TM-917) [10,13].

The slope of the curve $T(t)$ is a measure of the power release in a unit volume of the MF.

Figure 4 presents the rate of temperature increase $(\mathrm{d}T/\mathrm{d}t)_{t=0}$ at the beginning of the heating process as a function of the magnetic field intensity H in the form of an exponential function:

$$\left(\frac{\mathrm{d}T}{\mathrm{d}t}\right)_{t=0} = \left(\frac{H}{a}\right)^n, \tag{9}$$

where a and n are the parameters obtained from the fit of the exponential function, Eq. 9, to the experimental data. From the fitted function, it was possible to determine the values of $a = 3486$ and $n = 2.504$.

The latter value $n \cong 2.5$ can be interpreted either as a result of the size of magnetic particles in the magnetic fluid studied being much greater than that of typical superparamagnetic molecules or as a consequence of strong interactions in the colloidal solution of this high concentration. The greater size of the magnetic particles leads to a release of thermal energy as a result of the additional loss caused by the hysteresis.

Based on the principle of energy additivity, we can then write that the released power of losses proportional to $(\mathrm{d}T/\mathrm{d}t)_{t=0}$ consists of two components:

$$\left(\frac{\mathrm{d}T}{\mathrm{d}t}\right)_{t=0} = \left(\frac{H}{a}\right)^n = \left(\frac{H}{r}\right)^2 + \left(\frac{H}{h}\right)^3, \tag{10}$$

where r and h are parameters from the fit, which are adequate to describe losses for the relaxational mechanism and hysteresis.

Thus, Eq. 10 can be represented in the following numerical form:

$$\left(\frac{\mathrm{d}T}{\mathrm{d}t}\right)_{t=0} = \left(\frac{H}{3486.2}\right)^{2.504} = \left(\frac{H}{5604.5}\right)^2 + \left(\frac{H}{4057}\right)^3. \tag{11}$$

In Fig. 5, Eq. 11 is expressed by the curve of function $(\mathrm{d}T/\mathrm{d}t)_{t=0}$.

It follows from the shape of the function that for $H_{AC} > 2\,\mathrm{kA}\cdot\mathrm{m}^{-1}$ the losses related to magnetic hysteresis definitely start dominating in the total energy losses. The reason for the domination of the hysteresis among the total loss mechanisms can be explained by the dependences of the magnetic remanence and the coercive field intensity H_c, which are presented in Fig. 6a and b.

It is evident from these functional shapes that both values increase distinctly together with the growth of H_AC to the value $H_\mathrm{AC} \cong 20\,\mathrm{kA}\cdot\mathrm{m}^{-1}$. After reaching a magnetic field intensity of $20\,\mathrm{kA}\cdot\mathrm{m}^{-1}$, the saturation tendency is revealed. It is known that the losses for hysteresis are related to the surface area of a magnetic hysteresis loop, and the loop is larger the larger are the values of M_r and H_C, which result from Preisach's model [14].

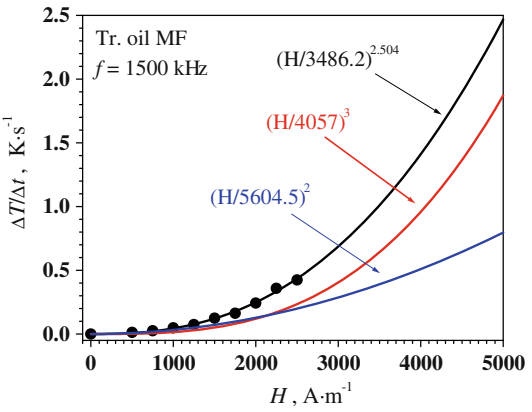

Fig. 5 Dependence $(\mathrm{d}T/\mathrm{d}t)_{t=0}$ as a function of the alternating magnetic field amplitude H_AC at a frequency $f = 1500\,\mathrm{kHz}$

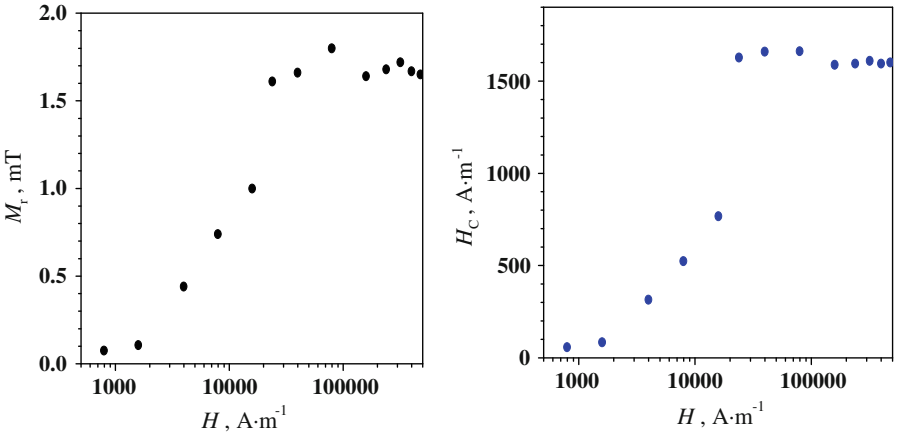

Fig. 6 Shape of magnetic remanence and of coercive field intensity as functions of alternating magnetic field amplitude ($f = 1500\,\text{kHz}$)

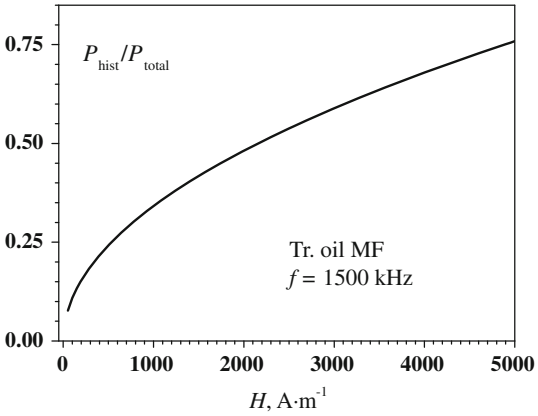

Fig. 7 Shape presenting the contribution of the energy losses for magnetic hysteresis in relation to total losses

From Eq. 11, it can be concluded also that together with the growth of the magnetic field intensity amplitude the contribution of energy losses for hysteresis increases, which was presented graphically in Fig. 7.

The specific absorption rate (SAR) defined as the thermal power dissipation divided by the mass of a magnetic crystal can be expressed as [15]

$$\text{SAR} = \frac{C_S \rho_S}{m_{Fe}} \frac{dT}{dt} \left[\frac{W}{g_{Fe}} \right], \tag{12}$$

where $C_S = 1.2834\,\text{J} \cdot \text{g}^{-1} \cdot \text{K}^{-1}$ is the specific heat capacity of the sample obtained with a DSCQ2000 calorimeter at $T = 25\,^\circ\text{C}$, $\rho_S = 1.6635\,\text{g} \cdot \text{cm}^{-3}$ is the MF density, $m_{Fe} = \phi_V \rho_{Fe} = 0.3367 g_{Fe} \cdot \text{cm}^{-3}$ is the mass of the magnetic material in the $1\,\text{cm}^3$ sample, and $\rho_{Fe} = 5.18\,\text{g} \cdot \text{cm}^{-3}$ is the density of magnetite.

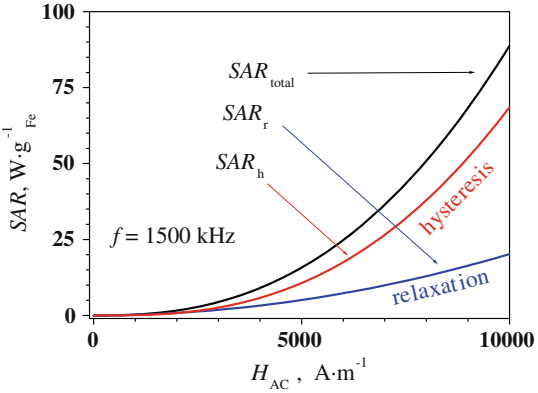

Fig. 8 Dependence of SAR on the magnetic field intensity ($f = 1500\,\text{kHz}$) together with the contributions from the relaxational losses and for hysteresis

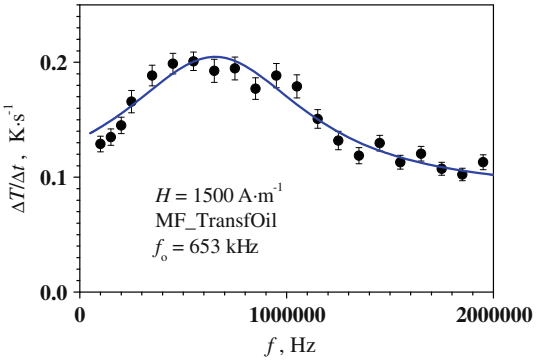

Fig. 9 Frequency dependence of $(\mathrm{d}T/\mathrm{d}t)_{t=0}$ determined for $H = 1500\,\mathrm{A}\cdot\mathrm{m}^{-1}$. *Circles* (•) stand for experimental values, and *solid line* (–) is the Lorentz function fit allowing an estimation of the frequency ($f_\mathrm{o} = 653\,\text{kHz}$) ensuring the maximum yield of heating $\mathrm{d}T/\mathrm{d}t_\mathrm{max}$

Taking into account Eqs. 11 and 12 and the experimental data, the following numerical form for SAR as a function of the magnetic field intensity is obtained:

$$\mathrm{SAR} = 6.34\left(\frac{H}{3486}\right)^{2.504}\left[\frac{W}{g_\mathrm{Fe}}\right], \tag{13}$$

which is also presented in Fig. 8.

The rates of temperature changes obtained at a constant magnetic field amplitude for different frequencies are shown in Fig. 9. For the frequency range from 420 kHz to 900 kHz at constant intensity ($H_\mathrm{AC} = 1500\,\mathrm{A}\cdot\mathrm{m}^{-1}$), the heating process is the most effective. The thermal energy loss reaches a maximum for a frequency $f_\mathrm{o} = 653\,\text{kHz}$. This maximum is probably related to the course of Eq. 5, which describes the imaginary part of the magnetic susceptibility as a function of frequency. In the limits of the range of frequency, $\mathrm{d}T/\mathrm{d}t$ reaches 90 % of the value obtained at a middle frequency, $f_\mathrm{o} = 653\,\text{kHz}$. Additionally, due to the values in Eq. 1a and b, the Brown and Néel average relaxation times of $\tau_\mathrm{B} \cong 15.7\,\mu\text{s}$, and $\tau_\mathrm{N} \cong 157.6\,\text{ns}$, respectively, were calculated. Therefore, $f_\mathrm{B} \cong 10\,\text{kHz}$ for $(2\pi f_\mathrm{B}\tau_\mathrm{B}) = 1$, and $f_\mathrm{N} \cong 1.01\,\text{MHz}$ for $(2\pi f_\mathrm{N}\tau_\mathrm{N}) = 1$.

The resultant relaxation time of both mechanisms is $\tau_{BN} \cong 156\,\text{ns}$, what matches the frequency $f_{BN} \cong 1.02\,\text{MHz}$. However, this frequency f_{BN} is bigger than f_o. It may be caused by the additional energy losses connected with the hysteresis phenomena.

4 Conclusions

1. From calorimetric measurements, it follows that the initial increase of the temperature rate $(dT/dt)_{t=0}$ is proportional to the exponential function (H/a^n).
2. The revealed H^n—law-type dependence of the temperature increase rate, $(dT/dt)_{t=0}$, on the amplitude of the magnetic field indicates the presence of superparamagnetic and partially ferromagnetic particles in the tested sample since $n > 2$.
3. With an increase of the magnetic field amplitude H, the contribution of energy losses related to magnetic hysteresis also rises, which results from a rise of the magnetic remanence B_r as well as from the coercive magnetic field H_c.
4. The heating process is the most effective and reaches a maximum for a frequency $f_o = 653\,\text{kHz}$.

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