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THE EXPEIMENTAL SETUP FOR MEASURING OF THERMAL PARAMETERS OF MAGNETIC FLUIDS IN AC MAGNETIC FIELD

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Abstract. Magnetic nanoparticle heating in alternating magnetic field is the novel approach to the hyperthermia technique for cancer treatment. The accurate measuring of the specific heat absorption rate of the magnetic fluid is the critical issue in the search for efficient materials to minimize the concentration of the magnetic nanoparticles in human tissues. In this work the heat absorption measurement setup consisting of a system of the magnetic field generation, a temperature measuring system, a cooling system, and an external computer control system has been developed. The results of the calibration as well as the results of measurements on the cobalt ferrite nanoparticles are presented.

Introduction. Magnetic fluid hyperthermia is a promising cancer treatment technique that uses the heat release of the magnetic nanoparticles subjected to the ac magnetic field of radiofrequency range (1kHz-1MHz) to locally heat the tumor [1,2]. The tumor is targeted by intravenously injected nanoparticles that heat the tissue up to 42 - 46 °C (for example, due to the magnetization reversal (hysteresis) losses). This heating destroys tumor cells, which are less resistant to raised temperatures than normal cells. When selecting magnetic fluid for hyperthermia the following items are important:
- the fluid should be nontoxic,
- the fluid heating in ac magnetic field should be essential.

The better the fluid heats the less concentration of nanoparticles is necessary for treatment, which reduces the problems of toxicity and presence of the magnetic material inside the human body. Thus to minimize the concentration of the magnetic nanoparticles in human tissues it is very important to accurately measure the thermal response of the specific magnetic fluid.

When absorbing the ac magnetic field energy magnetic fluids are warming up and in such case warming its environment. Such value as specific absorption rate (SAR) is used to characterize magnetic fluids' thermal properties. SAR is also called specific loss power (SLP) and is defined as magnetic field energy that is spent for heating per second per mass unit, and can be presented by the formula, described in Eq.1:

$$SAR = C\left(\frac{\Delta T}{\Delta t}\right)\left(\frac{1}{m}\right), \quad (1)$$

where C is the ferrofluid's heat capacity, $\frac{\Delta T}{\Delta t}$ characterizes the temperature change with time, m is a mass of magnetic material in ferrofluid. As usual, SAR characterizes with power per gram W/g but also it can be measured like energy per volume unit W/m^3 [3].

There are several methods of SAR measurement: in particular nuclear magnetic resonance method and calorimetric method. Nuclear magnetic resonance method consists of heating of the sample by continuous radiation laser beam. So the time dependence of spatially averaged NMR parameters is measured using non-invasive method. For determining of the SAR the liquid sample is heated by irradiating it by two types of radiation: radio and infrared, and then the phase shift of

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the nuclear spin is measured using a gradient echo pulse sequences. If electrical conductivity of the solution and the electric field amplitude are known, SAR that defined due to radio-frequency heating can be calculated for solutions. But in practice it is very problematic. SAR is not a constant; it varies with time and coordinates, and usually adjusted to specific mass and duration of irradiation of the sample. It is used to study harmful effects of mobile phones on human health and for diagnostic of the magnetic resonance imaging equipment [4].

Also SAR can be measured by determination of $\frac{\partial T}{\partial t}$ time dependence. This is so-called calorimetric method. For this method material's heat capacity should be known. In that case, SAR is defined as shown in Eq.1 [5]. The developed experimental setup was produced for checking of SAR for different samples.

Theory part 1. If we know the magnetic fluid heat capacity C, its volume and the value of temperature it's heated during the time t, than we can define SAR by the Eq.2:

$$SAR = \frac{\Delta T * C}{t} \quad \text{(in W/g)}. \tag{2}$$

By the biological reasons ac magnetic field used in the magnetic hyperthermia should obey the inequality from Eq.3:

$$H * f < const, \tag{3}$$

where H – magnetic field amplitude, f- magnetic field frequency, const ≈ $5*10^9 Am^{-1}s^{-1}$[3,6]. This limitations follow from the requirement of minimization of harmful effect on human body arising from the magnetic field. It was defined that to conform this inequality field amplitude should not be upper then 15kA/m and field frequency not to increase 1.2 MHz [3].
When placed in an external alternating magnetic field, magnetic nanoparticles generate heat through the so-called hysteresis loss mechanism. The amount of heat is proportional to the frequency of the external magnetic field and the hysteresis loop area. This shows that the nanoparticles with higher coercetivity and remanence magnetization, are heated better [3]. There is another mechanism of heating in an alternating magnetic field as well. It is called superparamagnetic mechanism. In this case, the sample can be heated even if the coercitive force of the sample material is close to zero [3]. Ferromagnetic materials that are used nowadays shows sufficient heating when magnetic field amplitude is 100 kA/m or even more before they approach a fully saturated state. On the other hand, the superparamagnetic materials are capable of producing more heat at lower fields [3]. Hergt et al. [3, 7] gave the best meaning of SAR 600 W/g for magnetite at a field of 11 kA/m and a frequency of 400 kHz, compared with the FM materials showed only 75 W/g for the amplitude of 14 kA/m for the best specimen of magnetite.

Design of apparatus. Experimental technique. The SAR-measuring setup has been designed and constructed in the aim to have an automatic apparatus to measure SAR of magnetic fluids. Among the setup advantages its relative simplicity and automatic operation should be mentioned.

The setup consists of:
1. system of the magnetic field generation, including the power supply, transformer, generator and a magnetic coil (solenoid);
2. temperature measuring system consisting of thermocouple and controlling voltmeter;
3. cooling system represented with cooler fan and air directing sleeve;
4. external computer control system and controlling scheme switching the device.

The system of the magnetic field creation consists of current source, a transformer, generator and a magnetic coil (solenoid). The voltage from the current source is amplified by a transformer and goes to the generator that creates 100 kHz signal in the coil. That allows to reach the magnetic

field strength of about 72Oe inside the magnetic coil with working bore of 16 mm and linear size of 43 mm. The temperature measuring system consists of differential thermocouple and voltmeter Agilent 34410A.

In the experiments a test-tube with the magnetic material is put into the solenoid working bore and the magnetic field is applied during fixed time interval. The temperature change caused by the field application is measured by the temperature measuring system. Then thermal parameters of the material are calculated by model equations.

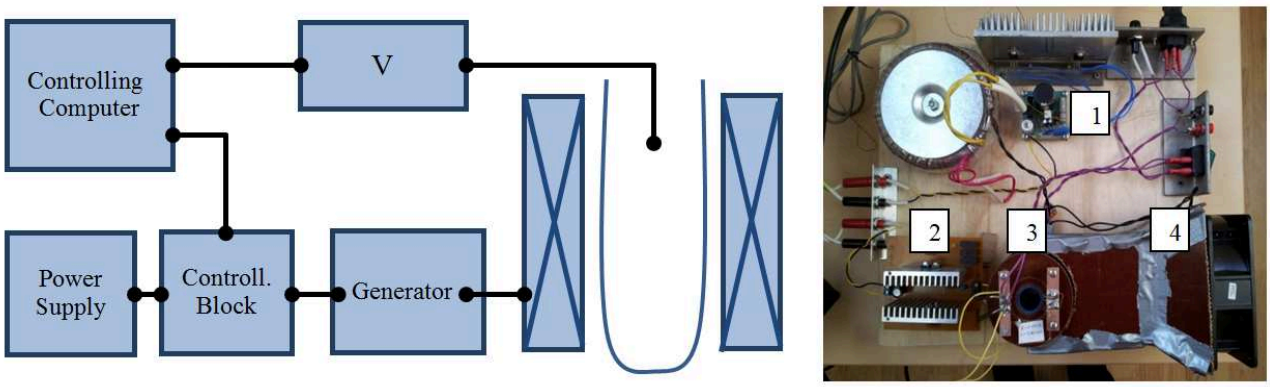


Figure 1. Left: setup block-scheme. Right: the photo of the setup. 1 is the power supply, 2 is the generator, 3 is the solenoid, 4 is the cooling system.

In the magnetic field generation system the 100 kHz signal generated by the driving generator. The solenoid consists of copper wire and has a working bore of 16 mm. The temperature measuring system consists of a differential thermocouple and a voltmeter. Voltmeter is connected to the control computer with the control software written in LabView8.0 via USB-interface. The control program sets the time interval during which the magnetic fluid is exposed to ac electromagnetic field. The program takes (from the thermocouple) the temperature values at the beginning of the experiment and every previously defined period of time before turning the ac field off. This temperature difference is directly proportional to the SAR value for this magnetic field amplitude. Heat capacity of the magnetic fluid was calculated from the heat capacity of the liquid and particles material taking into account the condition that the fluid is a solution of magnetic nanoparticles in a solvent (water, polymer).

The setup has been tested on the model object and on the sample of solid cobalt ferrite ($CoFe_2O_4$) nanoparticles, produced in Trinity College, Dublin. Nanoparticles have been synthesized by co-precipitation method and stabilized in water-soluble polymer.

Theory part 2. To understand, what exactly we measure with the setup, we've developed a model object. It was a copper ring. There are 3 mechanisms of heat transfer: conductivity, irradiation, convection [8]. In our system the main mechanism is convection, described with Eq. 4:

$$dQ/dt = \bar{h}A(T - T_\infty), \tag{4}$$

where Q –heat amount, $\bar{h}$ is a coefficient depending of material and the environment in which convection is produced, A is the surface of the object, T and T_∞ are current and initial temperatures respectively.

According to the 1st law of thermodynamics, common equation of heat transfer looks like Eq. 5:

$$\partial Q_{con} = dU + \partial A_{magn}, \tag{5}$$

where ∂Q is heat lost for convection, dU is internal energy differential, ∂A_{magn} is magnetic field work for the heating of the ring. Now we assume that this capacity is constant and depends on the parameters that are not changing in the experiment.

According to the capacities balance: the heat taken from the magnetic field used on internal energy increasing and to the convection (Eq. 6).

$$\bar{h}A(T - T_\infty) = cm\frac{dT}{dt} + dA_{magn}/dt, \tag{6}$$

where c is the specific heat capacity.

This is a simple differential equation and solves like shown in Eq. 7:

$$T = \frac{\bar{h}AT_\infty + dA_{magn}/dt}{\bar{h}A} + \left(T_\infty + \frac{\bar{h}AT_\infty - dA_{magn}/dt}{\bar{h}A}\right)e^{-\frac{\bar{h}A}{cm}t}. \tag{7}$$

In the area around t=0 it will look like linear dependence (Eq. 8).

$$T = T_\infty - \frac{dA_{magn}/dt}{cm}t \tag{8}$$

Then we need to determine the initial slope. We decided to take it from the real graph's derivate in $t = 0$ (Fig. 2).Derivate is 0.63. In that case low temperature approximation will look like shown on the graph. And $dA_{magn}/dt = 0.061\ W$.

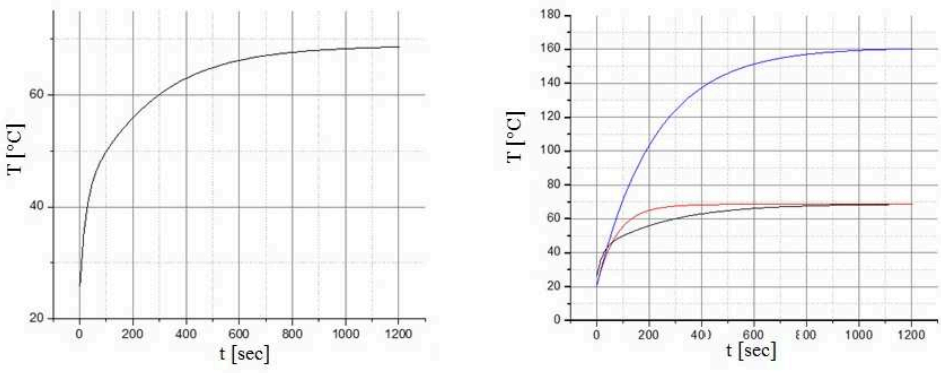


Figure 2. Left: Real dependence, taken with help of the setup. Right: On the picture you can see the real graph (black) and two approximations for different convection coefficients. Blue graph corresponds to the h=4.3 for plait sample, red graph – is for h=11.5 which is much better shows she toroidal shape of the sample.

The same idea of constant heating power may be used to calculate thermal effect for magnetic nanoparticles. According to R.E.Rosenveig [9] the power capacity for nanoparticles can be described by the Eq. 9:

$$P = \mu_0 \pi \chi'' f H_0^2, \tag{9}$$

where μ_0 is magnetic vacuum permeability, χ'' is out-of-phase component of magnetic susceptibility, f – is magnetic field frequency, H_0 is magnetic field amplitude.

So, this object will comply to the same Eq.5- Eq.9 but here we see the flection (at the beginning) that we can't explain for the moment. So to determine the initial heating power we have taken the meaning if derivative after this flection when function starts to behave like damped exponent. It is 0.030 which means the power density is about 0.0029W. The convective constant has been determined according the fact that powder intercommunicates with the environment with one flat side (like a surface).

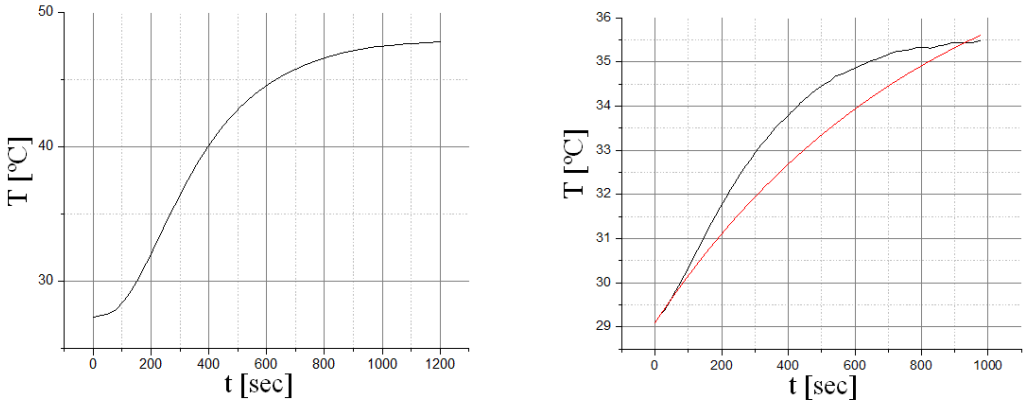


Figure 3.Left: Real graph measured for cobalt ferrite nanoparticles powder. Right: The graph there is a experimental curve (black) and simulation (red) for convective cooling mechanism.

Taking into account received data we can calculate SAR for the cobalt ferrite powder (Eq. 10):

$$SAR = \frac{\Delta T * C}{t} = \frac{7*140.4}{987} = 1\frac{W}{g} \quad (10)$$

Conclusions. The automated experimental setup for measuring of heating effect of magnetic fluid in ac magnetic field has been developed and constructed. This apparatus is made for examining of magnetic fluids thermal properties, namely SAR, in order to determine their applicability for the magnetic hyperthermia. The setup consists of magnetic field generation system, temperature measuring system, magnetic field measuring system and the control computer with control software. The setup has been tested on the model objects: copper ring and cobalt ferrite nanoparticles. For both of model objects there has been calculated an equation describing its heating process. The discrepancy between the experimental and theoretical graphs can be explained as another contribution into cooling mechanism (irradiation). It has not been taken into account because of the complexity of the relevant equations (Eq. 5 – Eq. 6).

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