Investigation of the Magnetic Properties of Ni Microparticles for the Utilisation in Magnetopneumography

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Abstract *Magnetopneumography is the method of investigation of the contamination and of the clearing mechanism of the human lungs based on the measurement of the remanent magnetic moment of the magnetised contaminating ferromagnetic particles in the lung. The aim of this work was to find the optimal conditions which enables to provide the most precise quantification of contaminating ferromagnetic particles in the human lungs. The magnetisation and the relaxation process of spherical Ni particles was investigated with special attention to the influence of the magnetisation time, the size and the concentration of the particles and to the viscosity of carrying medium. From the gained results the optimal conditions were concluded and into the praxis implemented.*

Keywords: SQUID, ferromagnetic particles, magnetopneumography, relaxation process

1. Introduction

In the past years the utilisation of the single domain magnetic nanoparticles in the field of biology and medicine became evidently wide spread. The measurement of the magnetic relaxation of these nanoparticles is used for the detection of the amount and location of immobilised MNP in vivo [1]. The occupational medicine faces the problem of the diagnostic of contaminating ferromagnetic particles in the human organs for example in the liver and in the human lungs.

The living organism is a compact system, where competing mechanisms try to influence the measured signal, hence giving higher uncertainties to the measured values. That is why it is necessary to know basic properties and behaviour of ferromagnetic particles as a response to the magnetic field in a more simplified media. A measurement system for magnetisation and relaxation measurements of magnetic microparticles was tested in regard to its applicability for *in vivo* diagnostics. The system is based on SQUID sensor enabling to detect the magnetic fields of order of 10^{-14}T. The aim of this work is to find the optimal conditions (time of magnetization, intensity of the applied field) for the measurement of the remanent magnetisation of ferromagnetic microparticles in the human lungs.

2. Theory

The intensity, duration and spatial homogeneity of the magnetising field are important determinants of the remanent magnetic induction. A ferromagnetic particle can become permanently magnetised through application of a strong magnetic field, which causes favourably oriented domains to grow in size and other domains to rotate into alignment with the applied field lines. Domain walls adjust rapidly and remain after removal of the external field, and the material generates a substantial remanent magnetic field. For moveable particles magnetisation duration will determine if particles rotate up with the applied field or become magnetised in place. It gives additional increase of the remanent magnetic induction signal although the remanent magnetic moment of the particle should increase no more. For particles of constant remanent magnetisation in low applied fields H_a the angle θ between H_a and the particles magnetisation direction decreases as a function of time [2]:

$$\tan(\theta/2) = \tan(\theta_o/2)\exp(e^{-t/\tau_0}) \qquad (1)$$

where $\tau_o = (\frac{\kappa\eta}{\mu_o M_r H_a})$, where M_r is the remanent magnetisation of a particle, V is the volume of the particle, κ is the particle shape factor, η is the viscosity of the carrying media and it holds that $\theta = \theta_o$ at t=0. After switching off the magnetic field, the magnetic moment return to equilibrium by two different mechanisms [5]:

1) Brownian relaxation is either due to the rotational diffusion of the whole particle in the

carrier liquid. The time constant of this process is given by:

$$\tau_B = \kappa \frac{V\eta}{2kT} \qquad (2)$$

where k is the Boltzmann's constant and T is the temperature of the liquid. This mechanism dominates for the particles with strong shape anisotropy.

2) Néel relaxation is due to the rotation of the internal magnetisation vector inside the particle with the time constant of

$$\tau_N = \tau_o \exp(\frac{\Delta E}{kT}) \qquad (3)$$

where $\tau_o = \frac{\pi^{1/2}}{2} \frac{m}{\gamma_o K_t V} (\frac{kT}{K_t V})^{1/2}$ and the energy barrier $\Delta E = K_t V$ depends on the total anisotropy energy constant K_t, m is the magnetic moment of the particle and γ is the gyromagnetic ratio. The time constant of Néel relaxation depends much stronger on the particle's diameter.

Since in general both mechanisms contribute to the magnetisation an effective relaxation time of

$$\tau_{eff} = \frac{\tau_N . \tau_B}{(\tau_N + \tau_B)} \qquad (4)$$

can be used. The effective relaxation time is dominated by the faster relaxation process. The time dependent magnetisation of a fraction of identical particles follows a simple exponential law:

$$M(t) = M(t_o) \exp(-\frac{t}{\tau_{eff}}) . \qquad (5)$$

Particles with τ_{eff} longer then the measurement period are called blocked particles and exhibit remanence. Particles with τ_{eff} shorter than the measurement interval are unblocked particles and do not show remanence. In real systems there is always a distribution of particle sizes, shapes and energy barriers which leads to a wide spread of relaxation times.

3. Materials and methods

Based on one of our earlier work [4] where we had examined 3 arc welders with contaminated lungs, we concluded that the contaminating welding fumes contained Fe and Ni particles in ratio 27%:73 %. It is proved that the contaminating particles in the human lungs are tapped in alveolar macrophages or in the connective tissue among them [4]. Therefore the hydrodynamic behaviour of the surrounding fluid characterises the cytoplasm, which viscosity was estimated to be from 60-100 Pa s. Therefore it was necessary to consider a carrying medium for the ferromagnetic particles from this interval of viscosity.

Sample preparation: Ni powder was mixed with epoxy gel of viscosity 70, 80, 170 Pa s without hardening agent and filled to small plastic cylinders of volume $V = \pi R^2 h$, where R = 0.0045 m and h= 0.008 m. During the night the particles sedimented to the bottom of the cylinders. Samples with different planar mass concentration of particles c_p were prepared: 1, 5, 10 mg cm^{-2}. Microscopy studies of the prepared samples showed that the microparticles consisted predominantly of single spherical shaped particles with mean core diameters d of about 10 um and 28 um and of aggregates of small sized FMP.

The measuring system contains a single channel second order SQUID [5] gradiometer as magnetic sensor. The measurement was provided using the compensation method [6]. The principle of this method is that the measured sample is inserted into the low-frequency AC difference magnetic field produced by two concentric coils with different diameters and place below the cryostat with the SQUID. The system is compensated before inserting the sample, the gradiometer output is a zero signal. After inserting the sample the compensation is disturbed and a voltage signal proportional to the instantaneous magnetic moment of the sample occurs in the gradiometric output. A second order gradiometer was used to suppress the disturbing signals of geomagnetic field. The magnetising signal was increased for the first t_u=10 s until it reached the value of 20 mT. After that the signal was held constant for times t_e= 2.5, 15, 30, 60 s. It means $t_{mag} = t_u + t_e$. The response of the system of particles was simultaneously recorded (see Fig. 1). The only practical limit for the application of longer magnetising times was the increasing thermal instability of the system. In order to reject this contribution to the useful signal, this was recorded and subtracted from measured values. To avoid the time window in

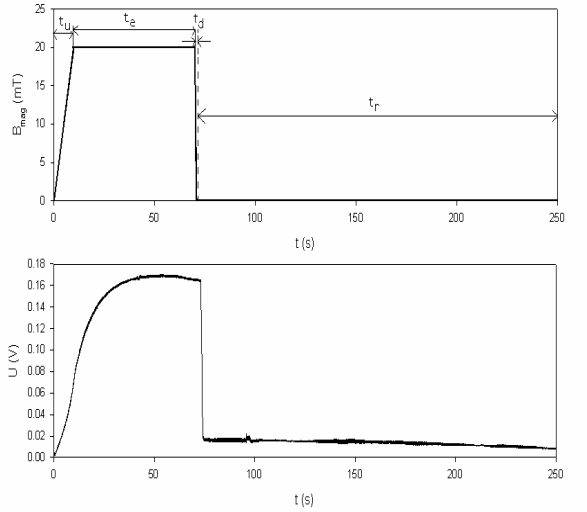

Fig. 1. The generated magnetising and the measured signal in time scale

the recording of the relaxation which is necessary for the SQUID to stabilise after switching off the magnetising field, we decreased the magnetising field gradually during 1 s from the value of 20 mT to the value 0 mT. The whole measurement was controlled by a computer by means of a LabWindows program. In order to eliminate irregularities in the signal which are caused by noise, different procedures were implemented in the data acquisition and data evaluation process. Using a digital first order low-pass recursive filter the data were smoothed, reducing frequency components below 5 Hz.

4. Results

Fig. 2 shows the time dependence of the magnetisation of samples with different c_p of Ni particles in the media of viscosity 70 Pas. The magnetisation increases with t_{mag} for low magnetising times and decreases after 50 s of magnetisation. The signal illustrated in Fig. 3 was used as a correction curve by the long-time magnetisation measurements. Fig. 4 shows a typical relaxation signal in the time window 0 - 1 s. Curve fit using a single exponential decay was unsatisfactory. However the superposition of two exponential decays fits the measured values well. The dependence of the relaxation curve on the concentration shows less the 10% difference for low c_p and d. However by increasing t_{mag}, c_p and d, the relaxation curves have diverging character (Fig. 5). One of the possible explanation should be the agglomeration tendency of highly concentrated big particles.

Fig. 6 shows the dependence of the relaxation of the $\boldsymbol{B}_r$ of samples with c_p = 10 mg cm^2 for different t_{mag} of the samples Fig. 7 shows the dependence of the relaxation of B_r on the η. Relaxation at this case was studied in the time window ranging from 500 ms till 90 s. The amplitude of the measured signal increases with the decreasing viscosity.

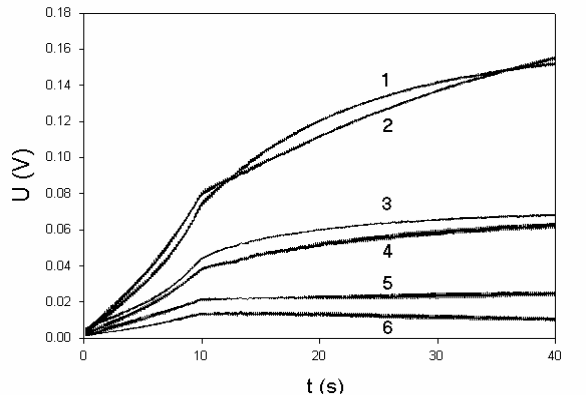

Fig. 2. The measured time dependent magnetisation signal of the samples with different c_p and d of Ni particles: 2, 4, 6 - particles with d = 10um; 1, 3, 5 – particles with d = 28 um; 1,2 – c_p = 10 mg cm^{-2}; 3, 4 – c_p = 5 mg cm^{-2}; 5, 6 – c_p = 1 mg cm^{-2}

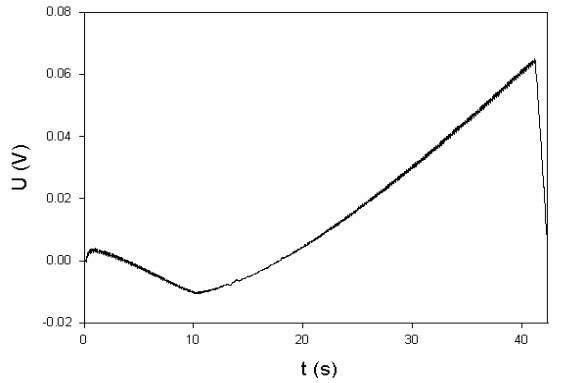

Fig. 3 The signal caused by the thermal instability of the system recorded for 40 s

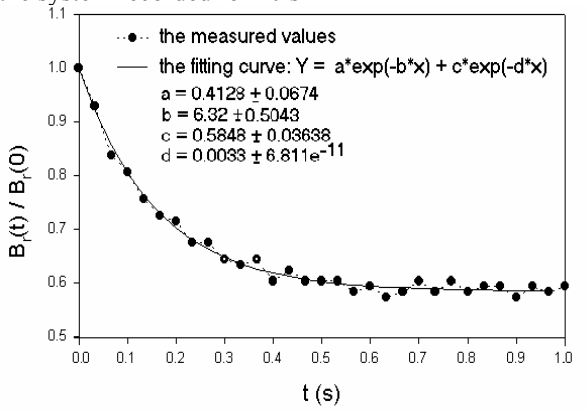

Fig. 4 The relaxation curve of 1 mg Ni particles of d = 10 um in the time window of 0 – 1 s magnetised for 60 s in the field of 20 mT.

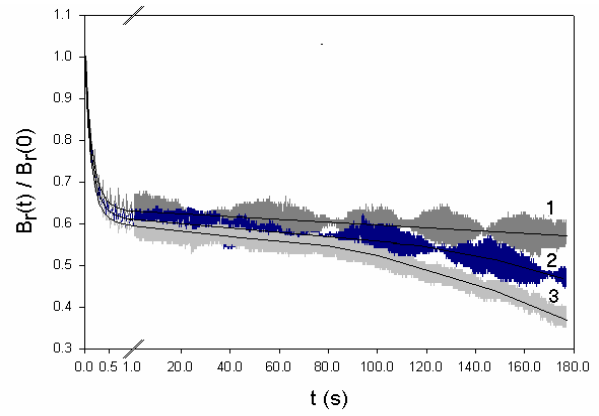

Fig. 5 The relaxation curves in the time window 0 – 180 s for Ni particles magnetised 60 s in the field of B_a = 20 mT of: 1) d = 10um, c_p =5 mg cm^{-2} 2) d = 28 um, c_p = 5 mg cm^{-2} 3) d = 28 um, c_p =10 mg cm^{-2}

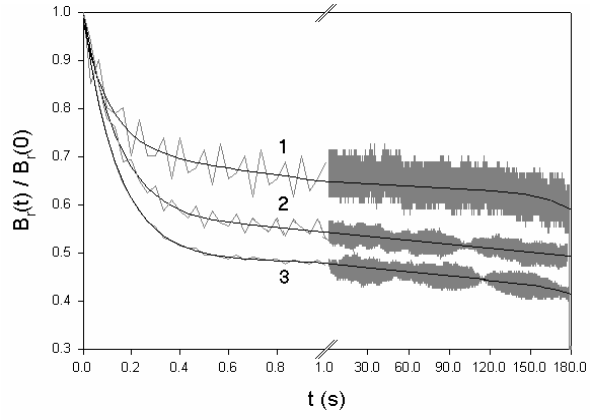

Fig. 6. Relaxation signals of samples of Ni particles of d = 10 um , c_p = 5 mg cm^{-2} magnetised in B_a = 20 mT for different t_{mag} : 1) 15 s, 2) 60s, 3) 30 s

5. Conclusions

A method for the measurement of the time dependent magnetisation of microparticles at room temperatures was presented, which also enables the investigation of the influence of the viscosity of colloidal materials. The measured results allow to make conclusions about the optimal conditions for magnetopneumographic investigation of lung dust loads.

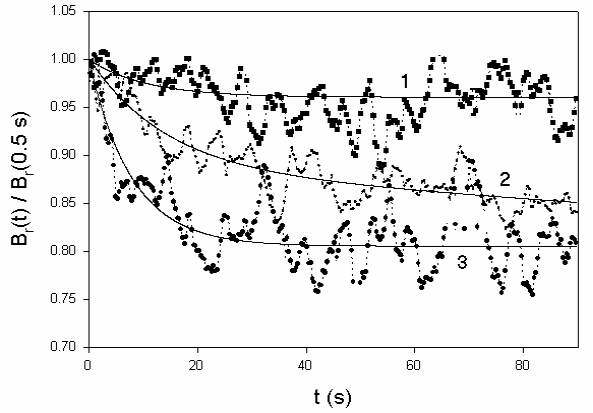

Fig. 7. The relaxation signal of 10 mg Ni particles of d =10 um (1, 3) and 48 um (2) magnetised in the B_a = 20 mT for 2.5 s in the carrying media of viscosity: 170 Pa s (1, 2) and 80 Pa s (3)

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