Synthesis and complex magnetic susceptibility characterization of magnetic fluids in different liquid carriers

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Abstract

Kerosene, isopar M, and mineral oil based magnetic fluids were prepared. The magnetite nanoparticles have spherical shapes and a mean diameter of 10.2±3.2 nm. The complex magnetic susceptibility $X(\omega) = X'(\omega) - iX''(\omega)$ of the magnetic fluids was measured using the transmission line technique applying dc magnetic fields over the range 0–1212 Oe over the frequency range 70 MHz–6 GHz, where resonance and relaxation are present. The resonance frequency f_{res} defined as the frecuency at which $X'(\omega)$ goes from positive to negative values ranges from 2.1 to 5.4 GHz. The maximum frecuency f_{max} defined as the frecuency at which maximum absorption occurs in $X'(\omega)$ ranges from 1.2 to 4.8 GHz. Anisotropy fields values were also determined.

Introduction

Magnetic fluids are stable colloidal dispersions of nanosized magnetic particles in a liquid carrier.[1] The particles used in most commercial applications are iron oxides smagnetite, maguemited because of their resistance to oxidation, whereas the transition metals have the benefit of higher saturation magnetization. Magnetic fluids can be used in rotating shaft seals, exclusion seals, loudspeakers, dampers, inclinometers, medical applications, etc.[2]

In this work we studied magnetite nanoparticles based magnetic fluids in three different liquid carriers: kerosene, mineral oil, and isopar M, using oleic acid as

surfactant. X-ray diffraction sXRDd, transmission electron microscopy sTEMd, vibrating sample magnetometry sVSMd, thermal analysis, and short circuited transmission line were used as characterizing techniques.

Experiment

Fe$_3$O$_4$ nanopowders were synthesized by a chemical coprecipitation method from JT Baker reagent grade chemicals, FeCl$_3$·6H$_2$O, FeCl$_2$·4H$_2$O, and NH$_4$OH. An amonium hydroxide solution at 10% was poured into a solution with a Fe^{+2}:Fe^{+3} ratio of 1:2 at 70°C with continuous stirring at 5000 rpm. A black precipitate was obtained. The washed precipitate was oven dried at 60 °C. From XRD patterns the phase composition of the obtained powders was determined using a D-5000, Siemens diffractometer. Powder magnetizations were carried out by a LDJ-9600 VSM.

Afterwards, to obtain the magnetic fluids a peptization process was used at 80 °C and 1000 rpm. Oleic acid (JT Baker) was used as surfactant and Isopar M (Exxon Mobil Chemical), kerosene (Fluka), and mineral oil (industrial grade) as liquid carrier. Three different magnetic fluids were obtained.

The particle shape and size of the magnetic fluids were observed by TEM and their magnetization curves were measured by VSM. Measurements of the complex magnetic susceptibility over the frequency range from 60 MHz to 6 GHz were carried out by the short-circuited coaxial transmission line technique.[3] Measurements were made using an Agilent Vector Network Analyzer 8753ES and a 50 Ω coaxial line incorporating a coaxial cell. The coaxial cell containing the magnetic fluid sample was terminated in a standard short circuit load and placed between the pole faces of an electromagnet, the axis of the cell being perpendicular to the bias field.[4] The bias field

H_s was varied between 0 and 1212 Oe. Automatic swept measurements over the entire frequency range were made and from these measurements, the real and imaginary components of the complex magnetic susceptibility were determined.

Results and discussion

The Fe_3O_4 powder was identified by XRD as a cubic spinel crystal structure. From TEM observations the nanoparticles were found to have a spherical shape with diameters between 5 and 13 nm, and a mean diameter of 10.2±3.2 mn.

Figure 1 shows the magnetization curves of Fe_3O_4 powder and magnetic fluids for different carriers. They all show reversible magnetization curves due to the superparamagnetic behavior of the magnetic nanoparticles in the carrier fluid. All these curves have experimentally negligible coercivities and remanent magnetizations. The different values of the maximum magnetization reached in these fluids are related to the different concentrations of the magnetic phase in the fluids. The magnetization curves show high field susceptibility, which can be attributed to a superficial spin disorder when the nanoparticle surface has significant roughness and a high broken bond density.[5] The magnetic fluid with Isopar M has the higher maximum specific magnetization of 21.3 emu/g because of the higher residual solids content of 9.64%; this is consistent for the three different magnetic fluids, kerosene based fluid 18.5 emu/g with 9.5 wt % in solids while for mineral oil based magnetic fluid with 16.2 emu/g has 8.18 wt % of residual solid phase which represents less content of magnetic material in the fluid.

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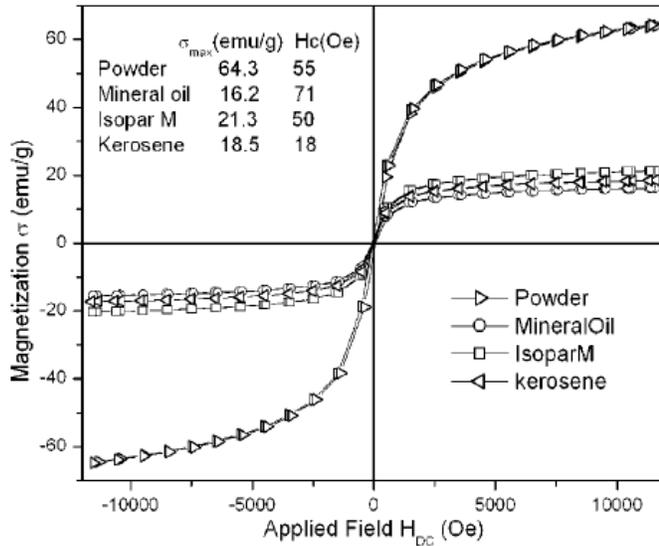
FIG. 1. Magnetization curves of magnetite powder and magnetic fluids using different liquid carriers.

Figure 2 shows the real component X'(ω) and the imaginary component X''(ω) of the complex magnetic susceptibility for magnetite in isopar M magnetic fluid, measured for bias fields H_s from 0 to 1212 Oe in the frequency range 70 MHz to 6 GHz in which the relaxation due to Brownian[6] rotational diffusion of particles can be ignored.
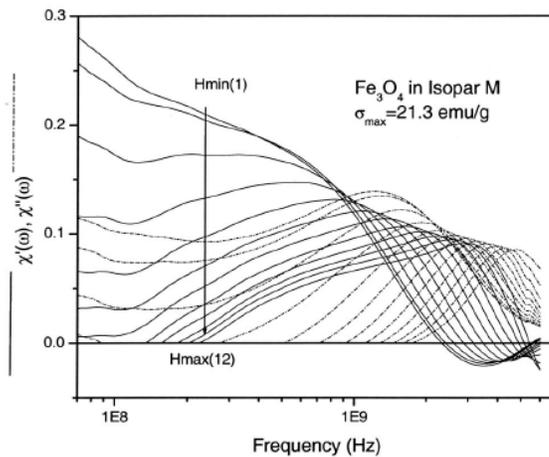
FIG. 2. Plot of real (solid line) part and imaginary (dash-dot line) part of susceptibility against frequency in Hertz, over the frequency range 70 MHz–6 GHz, for 12 increasing values of polarizing fields, of ≈0, 56, 156, 266, 378, 495, 613, 732, 852, 972, 1091, and 1212 Oe for isopar M based magnetic fluid of Fe_3O_4 nanoparticles.

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The X''(ω) component is constant up to a frequency of ≈400 MHz while at the same time the corresponding X'(ω) component decreases. These effects are a manifestation of the contribution of relaxational Néel[7] components to the susceptibility.[7] Beyond 400 MHz a relaxation to resonance transition occurs, and the resonance frequency f_{res} is determined when X'(ω) goes from positive to negative values (crossing through the zero valued). Over the range of biasing field, resonance frequencies range from 2.32 to 5.41 GHz for isopar M, from 2.09 to 5.06 GHz for kerosene and from 2.94 to 5.28 GHz for mineral oil magnetic fluids and corresponding values of f_{max} range from 1.19 to 4.77 GHz, 1.19–4.68 GHz, and 1.56–4.36 GHz. Increasing Hs produced a shift of f_{res} and also of f_{max} to higher values which is in accordance with the ferroresonance formula $f_{res} = \gamma / 2\pi(\overline{H_A} + H_s),$ where Y is the gyromagnetic ratio, and H_A is the anisotropy field. Also the higher the polarizing field, the lower the difference in the corresponding values of f_{res} and f_{max}; this arises because the bias polarizing field effectively results in an increase in the energy barrier, which the magnetic moments must overcome, and therefore in less spontaneous flipping of the magnetic moments (Néel relaxation) and increasing dominance to X(ω) from resonance.[8]

Also anisotropy fields of about 830, 767, and 811 Oe for isopar M, kerosene, and mineral oil magnetic fluids, respectively, were obtained by fitting the resonance frequency and bias polarizing field data to the resonance frequency equation

$$f_{res} = \gamma / 2\pi(\overline{H_A} + H_s).$$

Conclusions

Isopar M, kerosene, and mineral oil based magnetic fluids were prepared with superparamagnetic nanoparticles of magnetite spinel ferrite synthesized by a chemical coprecipitation method using metal salts. The nanoparticles of the magnetic fluids have spherical shapes with a mean diameter of 10.2±3.2 nm. Solids residual contents were 9.64, 9.5, and 8.18 wt % for isopar M, kerosene, and mineral oil, respectively, and corresponding specific maximum magnetizations were 21.3, 18.5, and 16.2 emu/g. The observed effects in the frequency range from 70 up to 400 MHz are a manifestation of the contribution of Néel relaxation to the susceptibility. Beyond 400 MHz a relaxation to resonance transition occurs. For the magnetite fluids, polarized over the range 0–1212 Oe, resonance frequencies ranged from 2.32 to 5.41 GHz, from 2.09 to 5.06 GHz, and from 2.94 to 5.28 GHz with corresponding frequencies of maximum absorption in the range from 1.19 to 4.77 GHz, 1.19 to 4.68 GHz, and 1.56 to 4.36 GHz. Anisotropy field values of about ≈830, 767, and 818 Oe for isopar M, kerosene, and mineral oil magnetic fluids, respectively, were also determined.

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