SUPERPARAMAGNETIC BEHAVIOR IN MAGNETIC FLUIDS CONTAINING COBALT PARTICLES DISPERSED IN MERCURY

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The magnetization curves of magnetic fluids consisting of cobalt particles dispersed in mercury have been measured at 300 K and the effect of particle interactions on the magnetization curve studied. An estimate of particle diameter, D_V = 100 Å, has been obtained and some assessment of the effect of particle interactions made. The effect of adding tin to the magnetic fluid to reduce particle interactions is also considered.

INTRODUCTION

Magnetic fluids consisting of iron or cobalt particles dispersed in a mercury carrier have been studied by Windle et al. [1], Popplewell et al. [2], and Keeling et al. [3] with particular emphasis on determining conditions for improving the stability of these systems in magnetic fields. In this paper, the superparamagnetic behavior as characterized by the absence of remanence and coercivity in the magnetization curve is discussed in terms of particle interactions. It should be noted that the metallic particle content in metallic fluid systems is limited to only a few percent by volume of the iron or cobalt. No effective surfactants can be added to produce repulsive forces to counter the attractive van der Waals forces, and aggregation from both long- and short-range forces is, therefore, extensive even in zero magnetic field. Nonmagnetic particle coatings such as those obtained by adding tin to the magnetic fluid can, however, be effective in reducing magnetostatic interactions by preventing the magnetic cores coming into contact. Coatings also inhibit particle growth and, as a consequence, improve fluid stability.

EXPERIMENTAL

Magnetic fluids containing cobalt particles dispersed in mercury have been prepared electrolytically using a cell which contains a platinum anode and an agitated mercury surface as cathode. A solution of cobalt chloride was used as the electrolyte. Depositions made at a current density of 25 mA cm^{-2} could be continued until the mercury became a thixotropic slurry, at which stage the cobalt content was a few percent by volume. In some instances tin was added to the final preparation in excess of that required to form a nonmagnetic monolayer coating on the particles.

Magnetization measurements in fields up to 20,000 Oe were made at 300 K on the liquid samples using an Oxford Instruments vibrating sample magnetometer.

RESULTS AND DISCUSSION

The magnetization curve for a magnetic fluid containing 0.3% by volume of uncoated cobalt particles in mercury is shown in Fig. 1. Superparamagnetic type behavior is evident, as is characteristic of samples prepared with cobalt concentrations of less than about 1% by volume.

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An analysis of the magnetic data of Fig. 1 using the technique of Chantrell et al. [4] shows that the experimental values for the variation of the magnetization with field fall well below the theoretical values for a system of noninteracting particles. The theoretical curve shown in Fig. 1 is derived on the assumption that the magnetic fluid contains a lon-normal distribution of noninteracting cobalt particles with mean diameter of 100 Å and reduced standard deviation of $\sigma = 0.4$. The mean diameter of 100 Å has been chosen to give magnetic saturation in a field of about 3 kOe as observed experimentally. A smaller mean diameter would give saturation at much higher fields and a larger mean diameter at too low a field. It has always been a problem to measure the particle size in metallic systems since methods for removing particles from the carrier are unreliable and measurements may, therefore, be unrepresentative. The approach outlined here could be a more reliable method of estimating particle size in metallic systems.

Chantrell et al. [5] have derived a theoretical magnetization curve for a strongly interacting system of cobalt particles 150 Å diameter using a two dimensional Monte Carlo model. This shows many features of the magnetization curve of Fig. 1 with magnetization values depressed well below those for the equivalent noninteracting system. The experimental magnetization curve shown in Fig. 1 is a typical of a system containing strongly interacting particles.

The magnetization curve for a magnetic fluid containing cobalt particles coated with tin is shown in Fig. 2. The curve is similar to that observed for uncoated particles in Fig. 1, but in this case the sample is more concentrated, containing 0.9% of cobalt by volume. As might be expected, therefore, particle interactions are more extensive and this is reflected by a greater deviation from the noninteracting particle curve. The effect of the tin coating can be seen by comparing the magnetization curve for a magnetic fluid containing 0.9% by volume of uncoated cobalt particles with that shown in Fig. 2. The magnetic fluid containing uncoated particles is in the form of a high viscosity slurry and has both remanence and coercivity. The coated particles, however, are superparamagnetic and the viscosity of the magnetic fluid is much lower. It may be inferred, therefore, that tin coatings are able to inhibit not only diffusional growth [6] but also the long-range structural aggregation that is evident in mercury-based magnetic fluids containing uncoated particles. The tin coating reduces the magnetic forces by increasing the average distance between the magnetic cores.

A further assessment of the effect on particle growth and particle interactions of coating particles with tin can be made by comparing the high-field regions of the magnetization curves of Figs. 1 and 2. It will be observed that the approach of the magnetization to saturation for the sample contining tin coated particles takes place over a greater field range than for the sample containing uncoated particles. This suggests that the particle size distribution for the coated particles contains a greater proportion of small particles. Particle growth, therefore, has been inhibited and corresponding reduction in particle interactions would also be expected.

The approach of the magnetization to saturation at high fields is given by [7]

$$\bar{I}/\bar{I}_S = 1 - 3kT/4\pi I_S r^3 H \qquad (1)$$

where $\bar{I}$, $\bar{I}_S$ refer to the magnetization of the magnetic fluid and I_S that of the particle, r is the particle radius and H the field, k is Boltzmann's constant, and T is the temperature. For a lognormal distribution of particle sizes, Chantrell et al. [4] gives

$$\bar{I}/\bar{I}_S = 1 - 3kT[4\pi I_S r^3 \int_0^\infty y^{-3} f(y)dy] \qquad (2)$$

where f(y) represents a lognormal distribution of reduced diameter $y = D/D_V$, where D_V is the median diameter and σ standard deviation. For the lognormal distribution

$$\int_0^\infty y^{-3} f(y)dy = \exp(9\sigma^2/2)$$

and Eq. (1) and (2) become identical for $\sigma = 0$

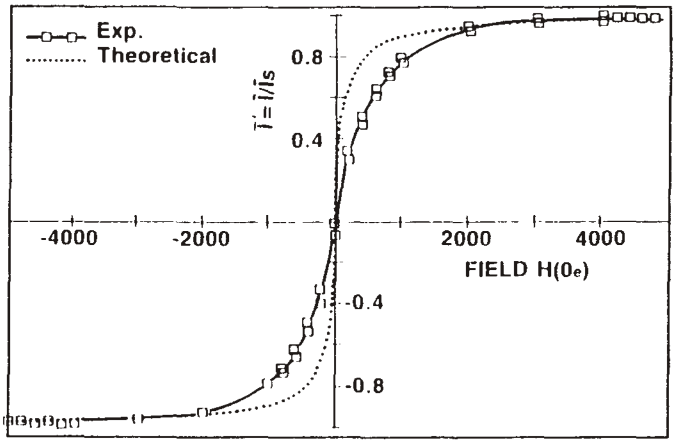

Fig. 1. The magnetization curve at 300 K for a magnetic fluid containing 0.3% by volume of cobalt particles in mercury. The dotted line is the best to fit to the experimental results for a noninteracting particle system (Chantrell et al. [4]).

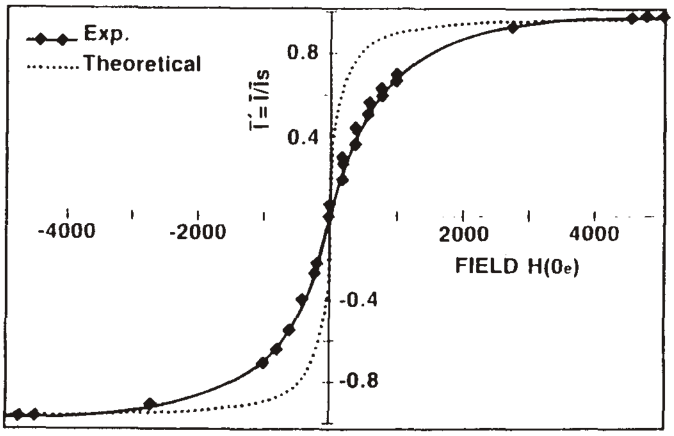

Fig. 2. The experimental magnetization curve at 300 K for a magnetic fluid containing 0.9% by volume of cobalt particles which have been coated with tin to inhibit diffusional growth and improve stability. The noninteracting particle theoretical curve is also shown for comparison.

At high fields Eq. (2) can be used to gauge the effect of tin coatings on controlling particle size. Thus with $\bar{I}/\bar{I}_s = 0.9$ at H = 2000 Oe for uncoated particles and H = 2750 Oe for coated particles, Eq. (2) indicates that coating with tin can lead to a reduction of about 25% in the volume of small particles (or 10% in the particle radius). The approach to saturation is therefore, a sensitive measure of the volume of the small particles in the distribution and consequently is valuable for assessing the effects of tin coatings on controlling particle size and the extent of particle interactions.

In conclusion it must be emphasized that a quantitative analysis of the magnetization curve to include the effect of particle interactions is not yet possible. This general conclusion is especially true for magnetic fluids based on liquid metal carriers.

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