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ON THE INFLUENCE OF NONUNIFORM MAGNETIC FIELDS ON FERROMAGNETIC COLLOIDAL SOLS

by S. Stephen Papell and Otto C. Faber, Jr.

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Cleveland, Ohio

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ABSTRACT

Description of a laboratory method for preparing magnetic sols, composed of ferromagnetic submicrometer particles as a stable colloidal dispersion in n-heptane, is presented herein. A fluid of this nature, originally synthesized at the NASA Lewis Research Center (U. S. Patent No. 3 215 572, S. S. Papell), was subjected to nonuniform magnetic fields in a vertically mounted solenoid-type electromagnet. Fluid accelerations of up to 7.2 g's were experienced by the magnetic sol without affecting the stability of the colloid system. Body force measurements were made on fluids containing a range of particle concentrations by weight from 1.58 to 13.41 percent that were subjected to magnetic inductions of from 0 to 2.75 teslas in magnetic gradients up to 22.3 teslas per meter. The relations between magnetic induction, magnetic gradient, particle concentration, and body force were examined, and the data are presented herein. An analysis was made from basic considerations that resulted in a general equation that effectively describes the trends in the data.

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ON THE INFLUENCE OF NONUNIFORM MAGNETIC FIELDS ON FERROMAGNETIC COLLOIDAL SOLS

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SUMMARY

Description of a laboratory method for preparing magnetic sols, composed of ferromagnetic submicrometer particles as a stable colloidal dispersion in n-heptane, is presented herein. A fluid of this nature, originally synthesized at the NASA Lewis Research Center (U. S. Patent No. 3 215 572, S. S. Papell), was subjected to nonuniform magnetic fields in a vertically mounted solenoid-type electromagnet. Uniform magnetic gradients, in addition to the Earth gravity vector, resulted in fluid accelerations of up to 7.2 g's. The magnetic body forces were transmitted to the fluid through the magnetic particles because of the stability of the colloid systems.

Body force measurements were made on several colloid systems covering a range of particle concentrations by weight from 1.5 to 13.41 percent. The sols were subjected to magnetic inductions of from 0 to 2.75 teslas in magnetic gradients up to 22.3 teslas per meter. The relations between magnetic induction, magnetic gradient, particle concentration, and body force were examined, and the data are presented herein.

A theoretical analysis was made that resulted in a general equation that effectively describes the trends in the experimental data. Direct correlation of the data with the equation was not possible because of an inability to calculate magnetization values independtly for the submicrometer magnetic particles.

INTRODUCTION

A magnetic colloidal sol is described in colloid science as consisting of minute magnetic particles in a liquid dispersion medium. Part of this description includes a statement on particle size, which is usually considered to lie in the range betweeen 5 nanometers and 0.2 micrometer (ref. 1). A system of this nature is usually considered stable when aggregation of the dispersed particles cannot be observed over a relatively long period of time (months to years). The published literature, for example, references 2 to 5, describes the preparation of dilute magnetic sols of nickel, magnetite (Fe_3O_4), and gamma iron (γFe_2O_3) with particle concentrations on the order of 0.1 percent by weight. These colloids were prepared by chemically forming the magnetic particles in the dispersion medium or by use of colloid mills. In both cases these methods were not used to produce more concentrated sols. The relatively low particle concentration of these colloid systems was sufficient to permit basic studies to be made on their optical and magnetic properties, for example, references 6 to 11. These studies were aimed specifically at the effect of magnetic fields on the colloid particles themselves. The dispersion medium was considered to be only a physical medium to hold and to separate effectively the individual solid particles. No considerations were given to any interactions between the dispersion medium and the particles of the sol.

The influence of magnetic body forces on the fluid dispersion medium, through the interaction between the solid magnetic particles and the liquid itself, has only recently been discussed in references 12 and 13. These studies required the use of a more concentrated magnetic colloidal sol than had previously been produced in the laboratory. In reference 12 there is a description of a 0- and reduced-gravity simulation technique that effectively reduces the weight of a magnetic colloidal sol by subjecting it to nonuniform magnetic fields whose gradients are oriented parallel but opposed to the Earth gravity vector. Reference 13 includes a phenomenological treatment for the fluid dynamics and thermodynamics of magnetic sols. In this study reference is also made to a private communication from the present authors that discussed the 0-gravity simulation technique described in reference 12 and, in addition, a method for priming rocket pumps in space. Continued work in this field describing the unique properties of ferromagnetic sols can be found in references 14 to 16.

The present experimental and analytical studies were aimed at examining the influence of nonuniform magnetic fields on relatively concentrated, optically opaque, stable, ferromagnetic colloidal sols. Various concentrations of a fluid of this nature, from 1.58 to 13.41 percent by weight, were subjected to magnetic inductions varying from 0 to 2.75 teslas and gradients up to 22.3 teslas per meter. The effective accelerations felt by the fluid were obtained from weight measurements of the fluid. A theoretical analysis was then made from basic considerations to attempt to correlate the experimental data.

A ferromagnetic fluid that could be subjected to magnetic forces was synthesized at the NASA Lewis Research Center (U.S. Patent No. 3 215 572, S.S. Papell) in the early months of 1963. Since information of this type is not readily available in books on colloid science, a description of a laboratory production technique is presented in this report.

MAGNETIC SOLS - PREPARATION AND DESCRIPTION

The magnetic colloidal sol used for this investigation was produced by suspending submicrometer particles of magnetic iron oxide (Fe_3O_4) in a dispersion medium of n-heptane. The submicrometer particles were made by using ball milling techniques similar to those described in reference 17. Although this reference deals with only the grinding of pure metallic powders, it was expected that oxide powders could be treated in a similar manner.

The milling procedure was to charge a ball mill with n-heptane, magnetite powder, stainless steel balls, and a surfactant such as oleic acid as a grinding aid. The mixture was allowed to grind for extended periods of time. At 24-hour time intervals the milling was stopped for fluid sampling. The samples were decanted after sufficient time had elapsed for the powder to settle out. During the initial part of the grinding, the samples drawn off were clear, colorless, and nonmagnetic. As the grinding proceeded, the fluid became opaque and brownish-black in color. The color change was caused by the solid particles suspended in the liquid. Electron-microscope photographs were then obtained to determine the shape and size distribution of these particles. A typical electron micrograph (fig. 1) shows both individual particles and agglomerations of particles. The ag-

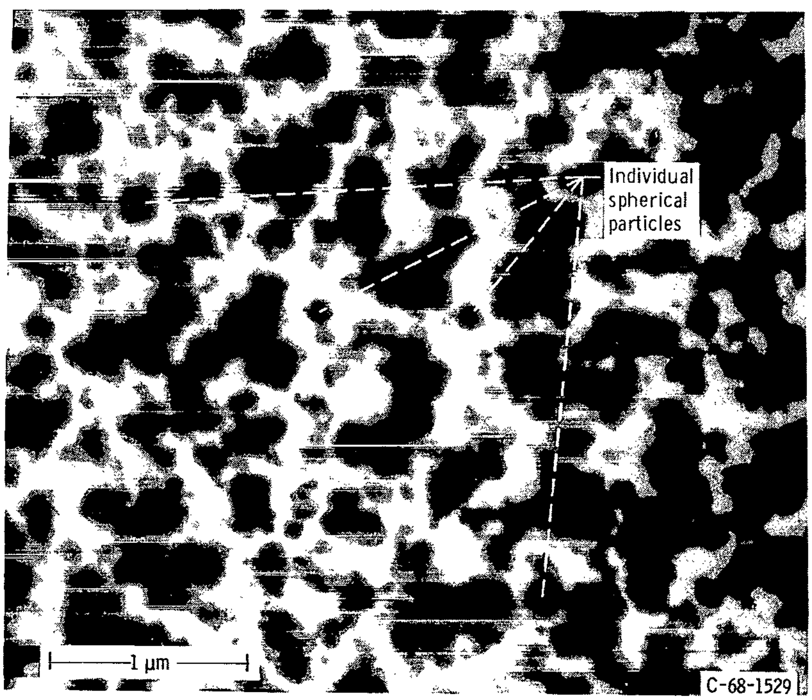

Figure 1. - Electromicrograph showing individual and agglomerated magnetite particles after evaporation of dispersion medium.

glomerations are primarily caused by the evaporation of the dispersion medium during the preparation of the slides. It was assumed that very little, if any, particle agglomeration exists in the colloid system itself since increased particle mass should result in visible precipitation. Samples of this magnetic sol have been stored for over 4 years without any apparent tendency for precipitation to occur.

The micrograph (fig. 1) also shows that the individual solid particles are nearly spherical in shape, and they were therefore assumed to be so for this study. Particle diameter measurements from these micrographs revealed a nearly normal number-size distribution that is a result of the ball milling process. Figure 2 is a number-size plot

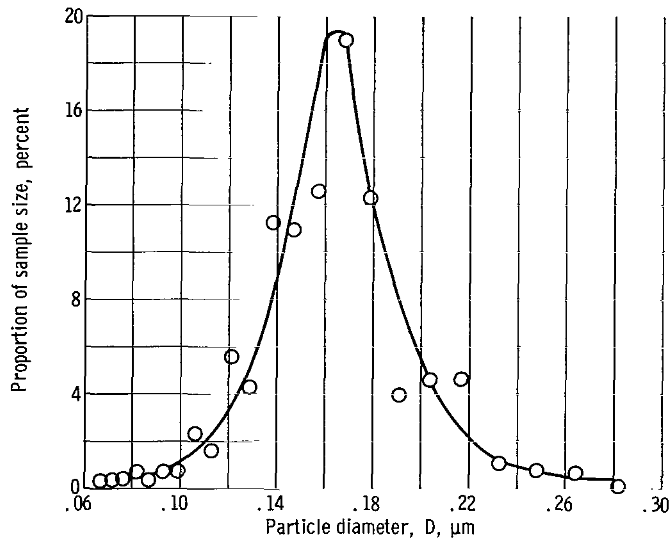

Figure 2. - Typical size distribution curve obtained from measurements of magnetic particles on electron micrographs. Sample size, 298.

showing proportion of sample size based on a sample of 298 measurements plotted against particle diameter in micrometers. For this particular sample, over 90 percent of the data lies between 0.10 and 0.22 micrometer in diameter. These dimensions are quite representative of particle diameters as produced by the grinding process described above.

The magnetic sol produced for this study had a particle concentration by weight of 13.41 percent calculated from mass density measurements by the following equation:

$$C_W = \frac{\rho_S}{\rho_C}(C_V) = \frac{\rho_S}{\rho_C}\frac{(\rho_C - \rho_L)}{(\rho_S - \rho_L)} \tag{1}$$

TABLE I. - MAGNETIC COLLOID PROPERTIES

OF MAGNETITE - n-HEPTANE

Sample	Median particle size by number, μm	Density of colloid system, ρ_c, kg/m^3	Particle concentration by volume, C_v, percent	Particle concentration by weight, C_w, percent
1	0.143	688.0	0.209	1.58
2	.163	699.0	.453	3.36
3	.083	716.9	.851	6.16
4	.163	728.3	1.105	7.86
5	.134	767.9	1.984	13.41

This relatively concentrated magnetic sol was diluted with n-heptane to achieve additional concentrations by weight of 1.58, 3.36, 6.16, and 7.86 percent. The mass density measurements used to obtain the values of these concentrations are listed in table I.

The ball-mill grinding process required several weeks of continuous grinding to produce the 13.41 percent by weight colloid system. It was later found that the grinding time could be reduced to a matter of hours by making use of an attritor grinding device. This device is essentially a modified ball-mill with a mechanical agitator submerged in the grinding media that is driven by an electric motor.

BODY FORCE MEASUREMENTS

Electromagnet Facility

The magnetic force field requirements for the experimental study were supplied by a 10-tesla (100-kG) water-cooled, solenoid-type electromagnet located at the Lewis Research Center. Figure 3 is a schematic sectional view of the electromagnet showing the orientation of its longitudinal axis parallel to the Earth gravity vector. The outside diameter of the coil and its height were both 5 decimeters (≈20 in.) in length. A 1-decimeter (≈4-in.) diameter bore provided sufficient room for positioning the experimental apparatus within the magnetic field.

The magnetic body force field within the bore of the magnet is indicated by the arrows on the drawing. A magnetic particle positioned at the top of the coil would experience a magnetic body force in the direction of the Earth gravity vector. As the particle is moved to the midsection of the coil, this body force goes to zero because the gradient

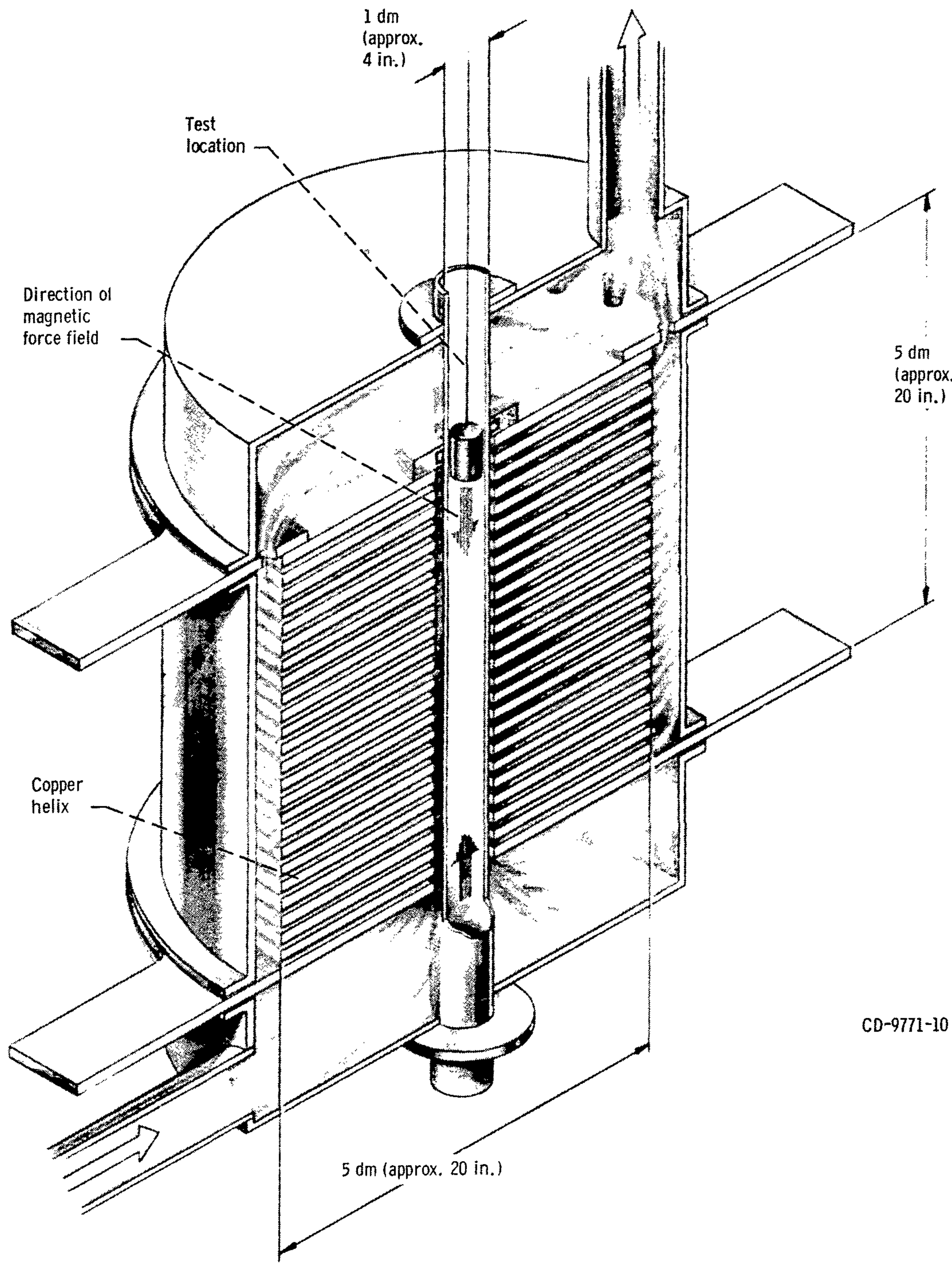

Figure 3. - Sectional view of electromagnet.

of the magnetic field goes to zero. At the bottom of the coil the direction of the magnetic body force is directly opposed to the Earth gravity vector. The experiments reported herein were run in the upper portion of the electromagnet, making use of the direction of the magnetic force field to enhance the Earth gravity force.

The actual position used for the study was required to be in a region of uniform magnetic gradient. Determination of this position was made by calculating the magnetic induction characteristics for the particular solenoid used by a method reported by Brown, et al. (ref. 18). Figure 4 is a plot of the results of the calculation showing the distribu-

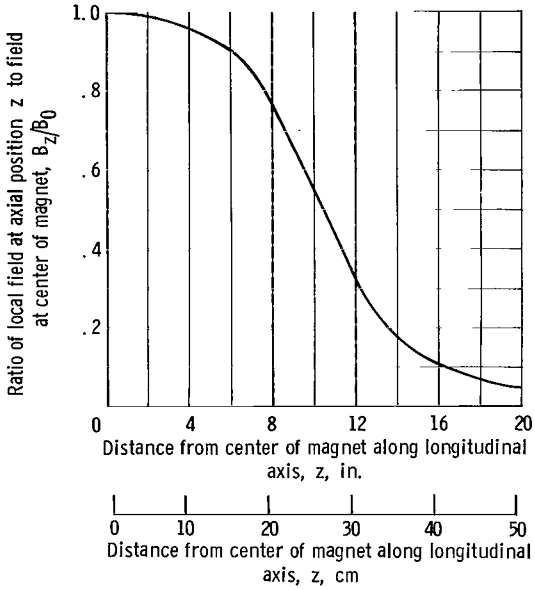

Figure 4. - Calculated distribution of axial component of magnetic field.

tion of the axial component of the magnetic induction. The ratio of the local axial magnetic induction at axial position Z to the magnetic induction at the center of the magnet B_Z/B_0 is plotted against the axial position Z measured from the center of the magnet. The slope of this curve is actually the magnetic gradient. The position Z = 25 centimeters at the inflection point is in the center of a region of nearly uniform magnetic gradient. The axial distance between Z = 20 to 30 centimeters represents 10 centimeters (≈4 in.) of essentially uniform magnetic gradient, which was used as the position in the magnet for the experimental study.

Calculations were also made to determine the magnitude of the radial component of the magnetic field at this predetermined test position. The results showed a radial magnetic gradient varying from zero along the longitudinal axis of the magnet to 1 percent of the axial gradient in the region occupied by the test apparatus. The effect of this small radial magnetic force was neglected in this study.

Experimental Procedure

A testing procedure was established to subject systematically the magnetic sols prepared for this study to a range of magnetic body forces and to measure the effect of these forces on the colloid systems. Since the weight (or force) of a mass is proportional to its acceleration, an analytical balance was used to measure the change in weight of the magnetic sols when under the influence of these magnetic body forces. The analytical balance was positioned about 5 meters (≈16 ft) above the electromagnet to prevent any influence from the magnetic field. The containers holding the fluid samples were suspended from an arm of the balance along the longitudinal axis of the magnet at the predetermined test position chosen for the experiments.

The fluid containers were sealed glass bottles with base diameters of 6.5 centimeters. The bottles contained approximately 100 cubic centimeters of the fluid samples whose weights, independent of their containers, had previously been recorded. The five fluid samples were individually positioned at the test position in the air core of the magnet while the magnetic induction B_o was varied from 0 to 5 teslas. At each magnetic

TABLE II. - BASIC BODY FORCE DATA

Magnetic induction at geometric center of magnet, B_o, T	Colloid sample				
	1	2	3	4	5
	Weight in magnetic field, N				
0.00	0.676	0.686	0.706	0.716	0.755
.25	.745	.755	.853	.883	.921
.50	.774	.833	1.009	1.039	1.127
.75	.804	.912	1.166	1.205	1.332
1.00	.833	.990	1.303	1.392	1.558
1.50	.912	1.156	1.627	1.732	1.970
2.00	.960	1.312	1.940	2.095	2.430
2.50	1.029	1.490	2.265	2.450	2.890
3.00	1.088	1.655	2.590	2.860	3.370
4.00	1.224	2.020	3.255	3.600	4.330
5.00	1.332	2.400	3.960	4.400	5.420

induction setting, the change in force on the fluid samples was recorded. The basic data is listed in table II, which shows that the forces on the fluid samples increase with increased magnetic induction B_0 and/or increased particle concentration C_w.

CALCULATIONS AND PRESENTATION OF DATA

Calculations were made using the basic data to determine the influence of nonuniform magnetic fields on a ferromagnetic sol in terms of the relations between magnetic induction, magnetic gradient, particle concentration, and the magnetic body force. In order to accomplish these, it was first necessary to determine the characteristics of the magnetic induction within which the test samples were subjected.

Magnetic Induction Characteristics

The data listed in table II do not include the magnitude of the magnetic induction or the gradient that actually influenced the test samples. The magnetic induction B_0,

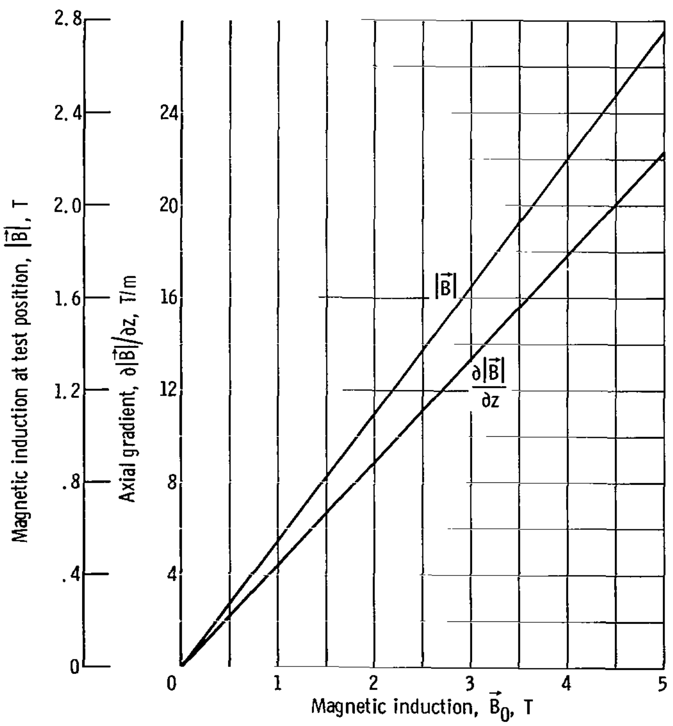

Figure 5. - Magnetic properties at test position as function of magnetic induction at geometric center of coil.

measured at the geometrical center of the electromagnet, was merely a convenient measurement to make in the test facility. The relations between B_o and the magnetic induction and gradient at the test position Z = 25 centimeters (≈10 in.) were therefore calculated by the method reported in reference 18. Prerequisites for this calculation were the geometrical properties of the electromagnet as shown in figure 3 (p. 6) and the knowledge that the current density of the magnet was inversely proportional to the coil inner radius (ref. 19).

The results of this calculation are presented in figure 5, which shows the magnetic induction and gradient at the test position Z = 25 centimeters (≈10 in.) as a function of magnetic induction B_o at the geometrical center of the electromagnet. The body force data will be presented later in this report in terms of these local magnetic induction characteristics.

Magnetic Versus Gravitational Body Force

In a nonuniform magnetic field, a magnetic particle experiences an acceleration in the direction of the gradient. If the particle is at the same time subject to the Earth gravity vector, then its effective acceleration can be described using the following relation:

$$a = \frac{F}{W} g_e \tag{2}$$

where

F weight measured under vector summation of both magnetic body force and Earth gravitational force

W weight in Earth gravity alone

g_e acceleration due to Earth gravity

The accelerations experienced by each of the five colloid test samples were calculated using equation (2) and the weight measurements listed in table II. The use of equation (2) in describing the action of magnetic and gravity forces as a vector summation is based on the assumption that a similarity exists in the action of both forces on the colloid system. The apparent stability of the solid-liquid system allows this assumption to be made.

A comparison of the nature of the action of these two forces can best be examined in terms of the manner in which they act on a basic volume of the colloid system consisting of a magnetic particle surrounded by a volume of liquid. The influence of the magnetic body force on the colloid system is essentially limited to the magnetic particle itself. In a nonuniform magnetic field, the particle experiences an acceleration in the direction of the magnetic gradient. Each particle influences a specific volume of fluid by some adsorption mechanism aided perhaps by the surfactant so that the magnetic body force is transmitted to the liquid through the particle itself.

The stability of the colloid system in a strong magnetic gradient was ascertained by subjecting the surface of the fluid to both magnetic and gravitational body forces whose lines of action were 90° apart. The liquid interface remained stationary for extended periods of time, attesting to the fact that there was no apparent migration of the magnetic particles in the direction of the magnetic gradient. The apparent stability of the system suggests that the applied magnetic body forces are smaller than the forces that hold the solid-liquid system together. On a macroscopic scale the colloid volume acts as a continuum with the magnetic body force acting through its center of mass.

Gravitational forces are considered to act equally on every elemental particle of the system, on a microscopic scale, which includes the solid and liquid components of the system. The weight of this colloid volume is the summation of all the parallel gravity forces acting as a resultant force through the center of gravity of the system. Therefore, on a macroscopic scale the acceleration experienced by the colloid system is the result of the net summation of both gravitational and magnetic body forces.

Presentation of Data

The influence of nonuniform magnetic fields on a (Fe_3O_4 - n-heptane) colloid system is presented in figure 6. Effective acceleration of the colloid system is plotted against magnetic induction as a function of colloid particle concentration by weight. Equation (2) was used along with the basic data to obtain the acceleration values while magnetic induction at the test position was supplied by figure 5. The magnetic gradient experienced by the colloid systems could also be obtained from the same figure, but it was not included in figure 6 for the sake of simplicity.

The curves in figure 6 are drawn through the data points for each of the five colloid samples that ranged in particle concentration by weight from 1.58 to 13.41 percent. All the curves show an increase in effective acceleration with increased magnetic induction. The slope of the curves also increases as a function of particle concentration. Although the data presented herein is unique for this particular colloid system, it may be significant to note that the largest value shown of a/g_e equal to 7.2 for the 13.41 percent by weight sol, does not represent an upper limit. Increases in acceleration may be obtained by changing the magnetization values of the colloid system. This can be accomplished with refined production techniques that reduce the colloid particles to single mag-

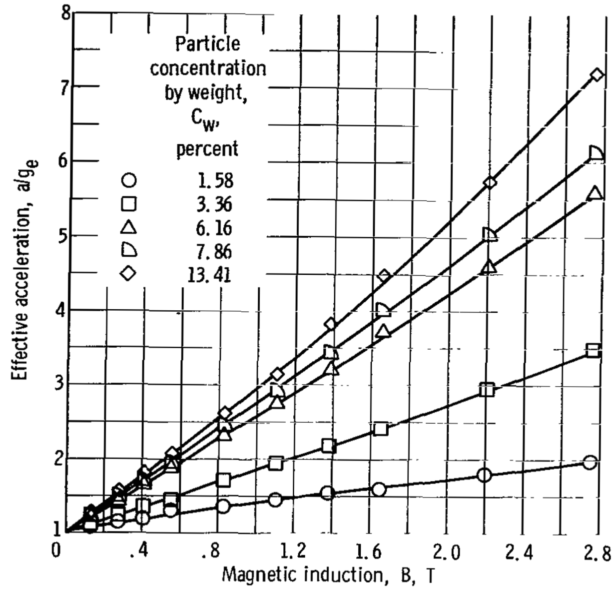

Figure 6. - Effective acceleration of magnetite - n-heptane colloid system plotted against magnetic induction as function of particle concentration by weight.

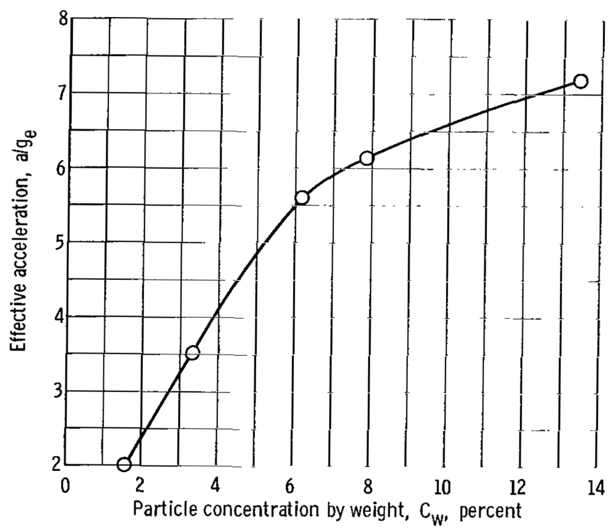

Figure 7. - Relation between effective acceleration and particle concentration at constant values of magnetic induction of 2.75 teslas and magnetic gradient of 22.3 teslas per meter.

netic domain size and/or by removing very small superparamagnetic particles whose thermal energy is much greater than their magnetostatic energy.

The relation between effective acceleration and particle concentration at constant values of magnetic induction and gradient of B_z equal to 2.75 teslas and dB/dZ equal to 22.3 teslas per meter, respectively, is illustrated in figure 7. A linear relation appears to exist up to a concentration by weight of about 7 percent. Above this value the slope of the curve decreases. It is significant to note that the departure point of the data curve from linearity corresponds to a particle concentration by volume (C_V) of approximately 1 percent. It is suggested that this apparent change in the data curve may be attributed to magnetic interaction (dipole-dipole) between the colloid particles. Reference 20 reports that such interactions should occur in magnetic colloid systems containing concentrations by volume greater than 1 percent.

ANALYSIS

An analysis was made in order to derive an expression that would predict the influence of nonuniform magnetic fields on ferromagnetic colloidal sols. Basic equations on magnetics were written explicitly in terms of properties of this solid-liquid system. Included were physical properties such as system density, particle size in terms of volume, and particle concentration. Also included were the permanent magnetic dipole moment and the induced magnetic dipole polarizability of the ferrite particles. The complete derivation can be found in appendix B with the final correlating equation presented herein for convenience.

$$\vec{a}_m = \left[\left(\int_0^\infty \left(\frac{C_V}{V}\right) \frac{|\vec{j}(V)|}{\rho_c} \left\{\coth\left[\frac{|\vec{j}(V)|\,|\vec{B}_{ext}|}{kT}\right] - \left[\frac{kT}{|\vec{j}(V)|\,|\vec{B}_{ext}|}\right]\right\} \Delta(V)\, dV\right) \vec{\nabla}|\vec{B}_{ext}|\right.$$

$$\left. + \left[\int_0^\infty \left(\frac{C_V}{V}\right) \frac{\eta(V)}{\rho_c \mu_0} \Delta(V)\, dV \pm \frac{(1 - C_V)|\chi_L^*|}{\rho_c}\right] \frac{\vec{\nabla}|\vec{B}_{ext}|^2}{2}\right] \tag{B19}$$

The above integral equation describes the acceleration felt by a ferromagnetic sol when subjected to a nonuniform magnetic field. An examination of the components of the equation shows that an increase in particle concentration (C_V), magnetic induction $|\overrightarrow{B_{ext}}|$, and/or magnetic gradients $\vec{\nabla}|\overrightarrow{B_{ext}}|$ and $\vec{\nabla}|\overrightarrow{B_{ext}}|^2$ will increase the net magnetic acceleration experienced by the fluid. These general trends can be observed in the experimental data presented in figure 6.

Equation (B19) can be solved if the magnetic properties of the ferrite particles could be calculated in some manner and if the volume distribution of the ferrite particles $\Delta(V)$ were known. Measurements made to determine the particle volume distribution showed that they did not follow a simple, well-known type of distribution law such as normal or log-normal. Although it would have been possible to curve fit some distribution function to the experimental data, this was not attempted because the more serious problem of determining the magnetic properties of the ferrite particles could not be overcome.

The particle size distribution measurements that were made showed that more than 99.9 percent of the ferrite particles are in the size range between 0.05 and 0.3 micrometer. The bulk of the particles is therefore above the single magnetic domain size of about 0.03 micrometer and below the true multidomain size of about 5 to 10 micrometers which, unfortunately, is a size range region of solid state physics in which very little theoretical or experimental work has been conducted (refs. 21 to 25). Consequently, there are no good relations for predicting the permanent magnetic dipole moment $|\vec{j}(V)|$ and the induced magnetic dipole polarizability $\eta(V)$ of magnetite in the size range used in this study. Correlation of the experimental data was therefore not attempted.

It may be significant to note that equation (B19) may be used only if the particle concentration by volume of the sol (C_V) is less than about 1 percent. At concentrations greater than 1 percent, the particles could be expected to undergo magnetic interactions with each other, which is a phenomenon not included in the analysis.

CONCLUDING REMARKS

Ferromagnetic colloidal sols, as reported in this study, can be subjected to external magnetic fields that produce magnetic body forces without affecting the colloid system stability. The liquid components of the colloid system can therefore be indirectly subjected to these forces. This unique property of a solid-liquid system of low viscosity could possibly have diverse technical application in the realms of fluid dynamics and heat transfer. Consequently, an investigation was undertaken to obtain some idea of the magnitude of the magnetic forces required to influence a fluid of this nature. The results of this study are actually threefold:

1. A description of a laboratory method for synthesizing a ferromagnetic colloidal sol was presented since this information is not readily available in books on colloid science. The particular sol produced for this study was composed of submicrometer-size particles of magnetite (Fe_3O_4) suspended in a dispersion medium of n-heptane at a 13.41-percent concentration by weight.

2. An experimental investigation was conducted to obtain body force measurements on several magnetite colloid systems by subjecting them to various magnetic fields. The relations between magnetic induction, magnetic gradient, particle concentration, and body forces were obtained, and the data are presented graphically.

3. An analysis was made from basic considerations to derive an equation that would predict the influence of nonuniform magnetic fields on ferromagnetic sols. The derived equation was found to predict the trends in the experimental data, but, unfortunately, could not be solved because of an inability to calculate independently the magnetization values of the submicrometer magnetite particles.

Lewis Research Center,
National Aeronautics and Space Administration,
Cleveland, Ohio, April 24, 1968,
129-01-11-03-22.

APPENDIX A

SYMBOLS

$\vec{a}$ acceleration, m/sec^2

$\vec{B}$ magnetic induction, T

$\vec{B}_o$ magnetic induction at geometrical center of electromagnet coil, T

C concentration, percent or decimal

$\vec{F}$ force, N

$\vec{f}$ force per unit volume, N/m^3

$\vec{g}_e$ gravitational acceleration at the local surface of the Earth, m/sec^2

$\vec{H}$ magnetic field, A-turns/m

$\vec{j}$ permanent magnetic dipole moment, J/T

k Boltzmann constant, J/K

$\vec{M}$ magnetization, A-turns/m

n number of solid particles per unit volume, m^{-3}

T temperature, K

V volume, m^3

$\vec{W}$ weight, N

Δ number-volume distribution function

δ Dirac delta function

η induced magnetic dipole polarizability (the magnetic analog to the induced electrical polarizability of a molecule in an external electric field), m^3

μ magnetic permeability, H/m

μ_o magnetic permeability of free space, H/m

ρ mass density, kg/m^3

χ volumetric magnetic susceptibility

χ^* volumetric magnetic susceptibility per magnetic permeability, m/H

Subscripts:

c colloid system

ext external

L liquid

m magnetic

s solid

v volume

w weight

APPENDIX B

MAGNETIC BODY FORCE ANALYSIS

When a small, isolated volume of ferromagnetic sol is subjected to a nonuniform magnetic field, the translational magnetic force per unit volume can be given as

$$\vec{f}_m = (\vec{M}_c \cdot \vec{\nabla})\vec{B}_{ext} \tag{B1}$$

where $\vec{B}_{ext}$ is the externally applied magnetic induction (by the law of conservation of momentum, the magnetic induction produced by the magnetized body itself cannot contribute to the translational force acting on the magnetized body (ref. 26)), $\vec{\nabla}$ is the vector differential operator del, and $\vec{M}_c$ is the net magnetization of the solid and liquid components of the colloidal system.

For an isotropic system of this nature, the net magnetization is in the direction of the external magnetic induction:

$$(\vec{M}_c \cdot \vec{\nabla})\vec{B}_{ext} = \frac{|\vec{M}_c|}{|\vec{B}_{ext}|}(\vec{B}_{ext} \cdot \vec{\nabla})\vec{B}_{ext} \tag{B2}$$

Using the vector identity

$$\vec{\nabla}(\vec{B}_{ext} \cdot \vec{B}_{ext}) = 2(\vec{B}_{ext} \cdot \vec{\nabla})\vec{B}_{ext} + 2\vec{B}_{ext} \times (\vec{\nabla} \times \vec{B}_{ext}) \tag{B3}$$

equation (B2) becomes

$$(\vec{M}_c \cdot \vec{\nabla})\vec{B}_{ext} = \frac{|\vec{M}_c|}{|\vec{B}_{ext}|}\left[\frac{\vec{\nabla}(\vec{B}_{ext} \cdot \vec{B}_{ext})}{2} - \vec{B}_{ext} \times (\vec{\nabla} \times \vec{B}_{ext})\right] \tag{B4}$$

The last term in the brackets vanishes (because the curl of $\vec{B}_{ext}$ vanishes), and therefore the translational magnetic body force, equation (B1), reduces to:

$$\vec{f}_m = (\vec{M}_c \cdot \vec{\nabla})\vec{B}_{ext} = |\vec{M}_c|\vec{\nabla}|\vec{B}_{ext}| \tag{B5}$$

The net magnetization of the colloidal sol system $\vec{M}_c$ is assumed to be the vector resultant of the magnetization of the solid dispersed phase $\vec{M}_s$ and the liquid dispersing phase $\vec{M}_L$:

$$|\vec{M}_c| = |\vec{M}_s + \vec{M}_L| \tag{B6}$$

Since these latter two magnetizations are collinear with the external magnetic induction, equation (B5) may be written as

$$\vec{f}_m = |\vec{M}_s| \vec{\nabla} |\vec{B}_{ext}| \pm |\vec{M}_L| \vec{\nabla} |\vec{B}_{ext}| \tag{B7}$$

where the positive or negative sign depends upon whether the magnetization of the solid dispersed phase is parallel or antiparallel to the magnetization of the liquid dispersing phase, respectively. Since for the colloids considered in this report, the solid dispersed phase would be either ferromagentic or ferrimagnetic, the choice of sign in reality depends upon whether a paramagnetic or dimagnetic dispersing phase is used, respectively.

An inspection of equation (B7) shows that the net magnetic force per unit volume on the colloid system has been resolved into two parts: one which depends only on the magnetization of the solid dispersed phase, and the other which depends only on the magnetization of the liquid dispersing phase. In the following, the contributions of the two factors will be treated separately and then combined at the end.

In the case of the solid dispersed phase, the magnetization is assumed to arise as a result of the permanent magnetic dipole moments and the induced magnetic dipole moments of the colloidal-size particles. If the colloidal particles were all of the same size (if they all had the same volume V_1) and did not interact with one another, this magnetization would be given by

$$|\vec{M}_s| = n|\vec{j}| \left\{ \coth \left[\frac{|\vec{j}|\,|\vec{B}_{ext}|}{kT} \right] - \left[\frac{kT}{|\vec{j}|\,|\vec{B}_{ext}|} \right] \right\} + \frac{n\eta}{\mu_o} |\vec{B}_{ext}| \tag{B8}$$

where

$n = \dfrac{C_v}{V_1}$ number of colloidal particles per unit volume (C_v = concentration by volume)

$|\vec{j}|$ magnitude of permanent magnetic dipole moment of a single particle

k Boltzmann constant

T absolute temperature

η induced magnetic dipole polarizability of a single particle

Equation (B8) is equivalent in form to the corresponding equation for the polarization (electric dipole moment per unit volume) of an assembly of noninteracting identical molecules in an external electric field if the appropriate correspondences are made (ref. 27). Theoretical and experimental justification of equation (B8) has been reported in the literature for magnetic colloidal sols and suspensions (refs. 2, 5, and 6).

A significant point mentioned in the literature warrants emphasis here: before quantitative agreement of the experimental results with the first term of equation (B8), the so-called Langevin equation, could be accomplished in reference 5, it was necessary to modify the Langevin equation by assuming a distribution in the size of the particle permanent magnetic dipole moments. And, although there is no explicit mention of the adoption of a similar procedure in the case of the last term in equation (B8), it is nonetheless expected that similar considerations would apply. One of the easiest ways, though not necessarily the most rigorous, to accomplish this alteration is as follows:

In the case when all the particles were of the same volume V_1, write n, $|\vec{j}|$, and η as functions of the volume V. (In practice, both $|\vec{j}|$ and η are also functions of the temperature T (refs. 28, 29, and 30), but for an isothermal environment this implicit temperature dependence will be suppressed.) Thus for the case considered:

$$\begin{aligned} n(V) &= n = \frac{C_v}{V_1} \text{ at } V = V_1 \\ &= 0 \text{ at } V \neq V_1 \\ |\vec{j}(V)| &= |\vec{j}| \text{ at } V = V_1 \\ &= 0 \text{ at } V \neq V_1 \\ \eta(V) &= \eta \text{ at } V = V_1 \\ &= 0 \text{ at } V \neq V_1 \end{aligned} \tag{B9}$$

The singular nature of these functions allows equation (B8) to be written in the form of an integral equation by making explicit use of the well-known Dirac delta function for functions which vanish everywhere except at a single point. The delta function is considered to be a one-dimensional delta function in a volume space and thus has units of a reciprocal volume (ref. 30).

$$|\vec{M}_s| = \int_0^\infty \left(\frac{C_v}{V}\right)|\vec{j}(V)|\left\{\coth\left[\frac{|\vec{j}(V)||\vec{B}_{ext}|}{kT}\right] - \left[\frac{kT}{|\vec{j}(V)||\vec{B}_{ext}|}\right]\right\}$$

$$\times\ \delta(V - V_1)\ dV + \int_0^\infty \left(\frac{C_v}{V}\right)\frac{\eta(V)}{\mu_o}|\vec{B}_{ext}|\delta(V - V_1)\ dV \qquad \text{(B10)}$$

In equation (B10), $\delta(V - V_1)$ is the one-dimensional Dirac delta function which vanishes everywhere except at the point $V = V_1$. If the condition that all the particles be of the same volume V_1 is now relaxed to include a distribution in volumes, then the Dirac delta-function $\delta(V - V_1)$ must be replaced by a generalized distribution function $\Delta(V)$ to include the effect of varying particle volumes on n $(= C_v/V)$, $|\vec{j}|$, and η in equation (B10). The generalization of equation (B8) to include a distribution of particle volumes and the relation between these respective volumes and the number of particles per unit volume of volume V, $(C_v/V)\ \Delta(V)$ (ref. 31); the magnitude of the permanent magnetic dipole moment of a particle of volume V, $|\vec{j}(V)|$; and the magnitude of the induced magnetic dipole polarizability of a particle of volume V, $\eta(V)$, can thus be expressed as

$$|\vec{M}_s| = \int_0^\infty \left(\frac{C_v}{V}\right)|\vec{j}(V)|\left\{\coth\left[\frac{|\vec{j}(V)||\vec{B}_{ext}|}{kT}\right] - \left[\frac{kT}{|\vec{j}(V)||\vec{B}_{ext}|}\right]\right\}\Delta(V)\ dV$$

$$+ \int_0^\infty \left(\frac{C_v}{V}\right)\frac{\eta(V)}{\mu_o}|B_{ext}|\ \Delta(V)\ dV \qquad \text{(B11)}$$

for a noninteracting colloidal sol system.

The magnetization of the liquid dispersing phase is defined by the relation (ref. 27)

$$|\vec{M}_L| = |\chi_L||\vec{H}_{ext}| \qquad \text{(B12)}$$

where

$|\chi_L|$ absolute value of relative volume magnetic susceptibility of the liquid

$|\vec{H}_{ext}|$ absolute value of external magnetic field acting on the liquid

Equation (B12) may be rewritten

$$|\vec{M}_L| = \frac{\mu_L}{\mu_L} |\chi_L| |\vec{H}_{ext}| = |\chi_L^*| |\vec{B}_{ext}| \tag{B13}$$

on defining the absolute value of the modified volume magnetic susceptibility of the liquid as

$$|\chi_L^*| \equiv \frac{|\chi_L|}{\mu_L} \tag{B14}$$

where μ_L is, of course, the magnetic permeability of the liquid dispersing phase.

As it stands, the right-hand side of equation (B13) must still be multiplied by a factor $(1 - C_v)$, where C_v is the concentration by volume of the solid phase in the colloid system, to correct for the volume of the liquid displaced by the solid phase:

$$|\vec{M}_L| = (1 - C_v)|\chi_L^*| |\vec{B}_{ext}| \tag{B15}$$

The net magnetic force per unit volume on a dilute magnetized colloidal sol system is then found by combining equations (B7), (B11), and (B15):

$$\vec{f}_m = \left(\int_0^\infty \left(\frac{C_v}{V}\right) |\vec{j}(V)| \left\{ \coth\left[\frac{|\vec{j}(V)| |\vec{B}_{ext}|}{kT}\right] - \left[\frac{kT}{|\vec{j}(V)| |\vec{B}_{ext}|}\right]\right\} \Delta(V)\, dV \right.$$
$$\left. + \int_0^\infty \left(\frac{C_v}{V}\right) \frac{\eta(V)}{\mu_o} |\vec{B}_{ext}| \,\Delta(V)\, dV \pm (1 - C_v)|\chi_L^*| |\vec{B}_{ext}| \right) \vec{\nabla}|\vec{B}_{ext}| \tag{B16a}$$

Equation (B16a) may also be written as

$$\vec{f}_m = \left(\int_0^\infty \left(\frac{C_v}{V}\right) |\vec{j}(V)| \left\{ \coth\left[\frac{|\vec{j}(V)| |\vec{B}_{ext}|}{kT}\right] - \left[\frac{kT}{|\vec{j}(V)| |\vec{B}_{ext}|}\right]\right\} \Delta(V)\, dV \right) \vec{\nabla}|\vec{B}_{ext}|$$
$$+ \left[\int_0^\infty \left(\frac{C_v}{V}\right) \frac{\eta(V)}{\mu_o} \Delta(V)\, dV \pm (1 - C_v)|\chi_L^*| \right] \frac{\vec{\nabla}|\vec{B}_{ext}|^2}{2} \tag{B16b}$$

by rearranging slightly the terms appearing in equation (B16a) and then applying the rule of vector analysis

$$\vec{\nabla}|\vec{B}_{ext}|^2 = 2|\vec{B}_{ext}|\vec{\nabla}|\vec{B}_{ext}| \tag{B17}$$

to equation (B16a).

From the Newtonian equation (ref. 33)

$$\vec{f}_m = \rho_c \vec{a}_m \tag{B18}$$

the translational magnetic acceleration $\vec{a}_m$ on a dilute magnetized colloidal sol system, the colloidal sol system remaining a continuum, may be computed from the following equation:

$$\vec{a}_m = \left(\int_0^\infty \left(\frac{C_V}{V}\right) \frac{|\vec{j}(V)|}{\rho_c} \left\{\coth\left[\frac{|\vec{j}(V)|\,|\vec{B}_{ext}|}{kT}\right] - \left[\frac{kT}{|\vec{j}(V)|\,|\vec{B}_{ext}|}\right]\right\} \Delta(V)\, dV\right)$$

$$\vec{\nabla}|\vec{B}_{ext}| + \left[\int_0^\infty \left(\frac{C_V}{V}\right) \frac{\eta(V)}{\rho_c \mu_o} \Delta(V)\, dV \pm \frac{(1 - C_V)|\chi_L^*|}{\rho_c}\right] \frac{\vec{\nabla}|\vec{B}_{ext}|^2}{2} \tag{B19}$$

where ρ_c is the mass density of the colloidal sol system.

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