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AN EXPERIMENTAL AND THEORETICAL INVESTIGATION OF THE MAGNETIZATION PROPERTIES AND BASIC ELECTROMECHANICS OF FERROFLUIDS

FINAL REPORT

September, 1977

Contracts #N00014-67-A-0299-0020
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Power Program, Office of Naval Research

Prepared by

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I. Introduction

Ferrofluids are stable colloidal suspensions of sub-domain sized ferrite particles dispersed in a liquid medium by a suitable surfactant agent. Ferrofluids have been successfully prepared using water, hydrocarbons, esters, diesters, fluorocarbons, and even liquid mercury. Since their recent development, considerable interest in the possible application of ferrofluids has emerged. A great diversity of applications has been proposed, including propulsion systems, accelerometers, fluidic elements, fluid damping components, energy converters, and magnetic printers. Two applications showing considerable promise are ferrofluid rotary shaft seals and scrap metal separaters. Rotary shaft seals have been commercially available for several years.

Basic research on the magnetization properties and the bulk response of ferrofluids to magnetic fields has been conducted to meet the needs of the developing technology of ferrofluids. To this date, research on ferrofluids has been fragmented and hampered by apparent efforts to protect proprietary knowledge concerning the constituents of these fluids and their application. Recently, Dr. B. Berkovsky has organized the first meeting devoted specifically to ferrofluids and ferrofluid applications.* Many things about ferrofluids and their behavior in a magnetic field are not yet understood, and this lack of knowledge presently hampers development of the technology.

From the fluid mechanical point of view, ferrofluids are novel because they can interact with a magnetic field to produce a controllable body force

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*International Advanced Course and Workshop on the Thermomechanics of Magnetic Fluids, sponsored by UNESCO, Oct. 3-7, 1977, Udine, Italy.

on the fluid, a body force significant with respect to terrestrial gravity. From the basic physical point of view, ferrofluids are interesting because of the mechanisms which are involved in the transformation of the forces on individual ferrite particles to the bulk of the liquid carrier. Historically, two areas relevant to ferrofluids have received considerable attention. These are the study of fluids with weak magnetic properties, and the study of solids with strong magnetic properties. The wedding of these two fields has become possible and necessary through the development of ferrofluids. This final report summarizes three years of research funded by the Office of Naval Research under contracts #N00014-67-A-0299-0020 and N00014-76-C-0250. The research program was divided into studies of the magnetization properties, principally investigated by D. A. Krueger, and the electromechanics and applications, principally investigated by T. B. Jones, both of Colorado State University. An outline of the essential features and sigificant results of the research is provided below for convenience in reviewing the program. A more detailed summary follows in the next several sections of this report.

Magnetization Studies

The principal result of this effort has been the discovery of a reversible, magnetic field induced agglomeration effect which occurs in water-base, ester-base, and hydrocarbon-base ferrofluids.* This agglomeration is observed in constant and time-varying magnetic fields as low as 5 Oe. It occurs whether the field is uniform or non-uniform. Measurements indicate that the agglomerates increase to sizes involving up to 10^5

*Other ferrofluids were not examined due to their high cost.

individual ferrite particles, then settle out due to gravitational forces. The agglomerates break up if the magnetic field is removed and gentle mixing restores the ferrofluid to its original condition. The effect is much more pronounced in water-base ferrofluid than in hydrocarbons. Substantial modeling efforts have been made, and some of this effort is reviewed in section II.

Electromechanics and Applications

Simple experiments with ferrofluids to determine their electromechanical response to magnetic fields have been conducted. Principal among these is the Quincke experiment, in which the height of rise of a column of ferrofluid in a magnetic field is measured. These measurements are found to be consistent with magnetization measurements obtained using a vibrating sample magnetometer. Some evidence for agglomeration is noted if the ferrofluid is not kept mixed throughout the measurements. A principal conclusion of these experiments is that modeling the electromechanics of ferrofluids is successful only if the magnetic non-linearity is properly accounted.

A static ferrofluid plug has been tested. Measurements made with these seals are consistent with a simple ferrofluid seal model. The mechanism of failure for a fluid/fluid seal has been observed. An extended series of experiments with a rotary ferrofluid seal has provided the chance to gain insight concerning the important seal design parameters, including ferrofluid seal inventory, magnetic flux, gap spacing, and rotational speed. These results are detailed in section III.

II. Magnetization Studies

The utility of and intrinsic interest in ferrofluids arises because these fluids have magnetic properties as well as fluid properties. We have made measurements of basic magnetic properties for several fluids subjected to a variety of conditions, and we have used magnetization measurements as a tool to investigate agglomeration phenomena. Using our newly developed experimental technique, we have observed agglomeration in concentrated ferrofluids like those used in devices. Previous observations of agglomeration were made in very dilute ferrofluids which are poorly suited for devices. In addition our observations were on fluids with measured intrinsic magnetic properties. This is in contrast to earlier experiments and aids greatly in our theoretical understanding.

A. General Properties

Using a PAR vibrating sample magnetometer, we measured the magnetization, M, as a function of applied magnetic field, H, for various combinations of fluid base (water, hydrocarbon, and ester), storage temperature (1°C, 25°C, and 50°C), and magnetic field while stored (zero field, 380 Oe uniform field and a 500 Oe/cm gradient field). After about 6 months storage the magnetizations, in almost all cases, had dropped by about 5%. This is possibly due to the formation of agglomerates which have some flux closure and thus do not respond to the externally applied field.

Osmotic pressure measurements should give information on the number density of the magnetite particles in the colloid. We found an irreproducibility which might also reflect an agglomeration of the particles.

An electron microscope was used to measure the size of the magnetite particles. We found that the water-base fluid had significantly larger magnetite particles than did the ester or hydrocarbon base fluids. The size distributions for these two fluids were indistinguishable. Using the Langevin theory to calculate the magnetization as a function of applied magnetic field we found fairly close agreement with our measurements discussed above. However, the average of the volume, <v>, as calculated from the electron micrograph measurements are roughly a factor of two larger than those deduced from the M vs. H measurements. This is not surprising because a few very large particles observed in the electron micrographs contribute very heavily to the average volume. In addition the extrapolations of the M vs. H are uncertain. These measurements are reported at length in earlier annual reports of this project [1] and in Peterson [2]. These measurements indicate that agglomeration of the magnetite particles is occurring. We next briefly review a long series of experiments which unequivocally demonstrate this agglomeration.

B. Agglomeration - Experiments

The basic experiment which we devised to study agglomeration is basically to measure the gravitational settling of the agglomerates in a vertical tube 11 cm in length and 0.6 cm in diameter. A schematic of the experiment is shown in Figure 2.1. The shift in the resonant frequency of the Colpitts oscillator circuit, Δf, upon insertion of the tube into the coil was found to be proportional to the saturation magnetization of the fluid in the column within 0.37 cm of the center of the coil. This in turn is proportional to the amount of magnetite per unit volume. A typical result for Δf as a function of position along the tube is shown in Figure 2.2. It is clear that magnetite particles are moving from

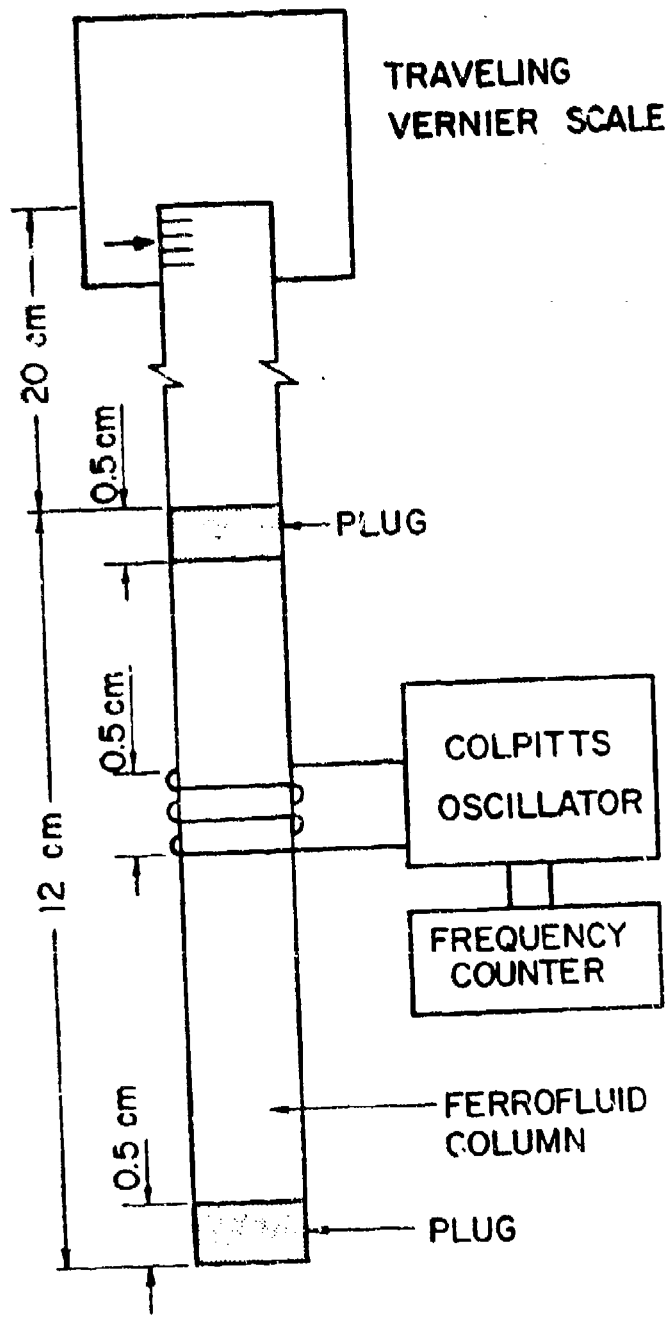

Fig. 2.1 Experimental arrangement used for *in situ* magnetization measurements using oscillator circuit.

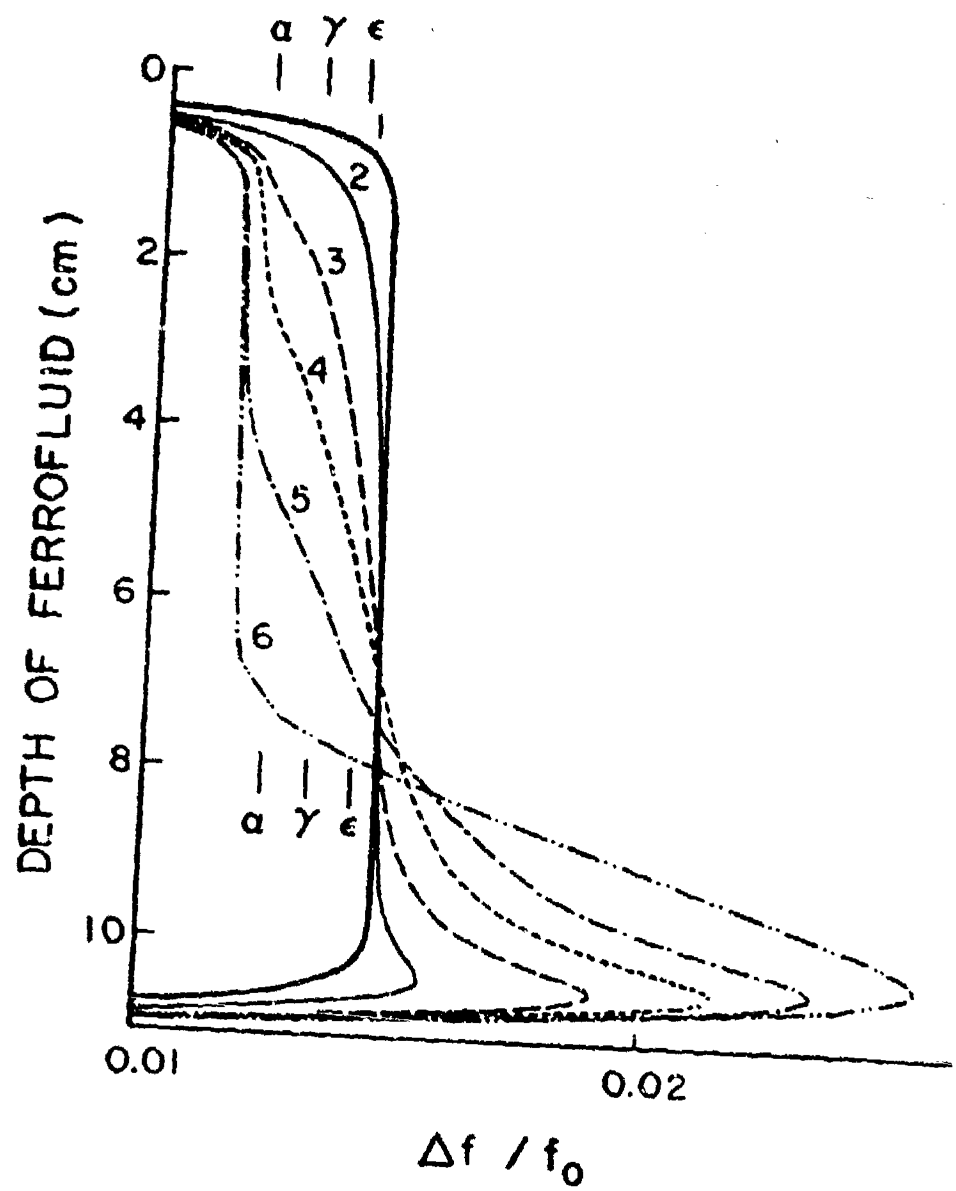

Fig. 2.2 Concentration profiles after exposure to a 230 - Oe uniform vertical field for (1) 0 min, (2) 5 min, (3) 10 min, (4) 15 min, (5) 25 min, and (6) 30 min.

the top of the tube to the bottom. Analyzing this as a function of time shows that the particles are traveling at a constant velocity which we identify as the terminal velocity. This terminal velocity depends upon the uniform magnetic field which is applied to the fluid as shown by the crosses in Figure 2.3. The solid line is the result of our theoretical model which we discuss in detail in Section C.

From our data, several features of the agglomeration are evident. The agglomerates are made up of on the order of 10^9 magnetite particles. In fact, our technique is insensitive to agglomerates of less than about 10^7 particles. The time for an agglomerate to form is about 40 sec. The time for an agglomerate to break up upon removal of the magnetic field is about 10 sec. The agglomeration is reversible. From electron micrographs of the fluid taken from the top of the tube and from the bottom of the tube after agglomeration has taken place, we find that all sizes of magnetite particles in the fluid participate in the agglomerates. Agglomeration and gravitational settling can change the concentration of magnetite particles by factors of two within an hour. Water-base fluid exhibits much more agglomeration than does an ester or hydrocarbon-base fluid. The fraction of the magnetite which participates in agglomeration increases as $H^{0.5}$. If there is a threshold for agglomeration, it is less than 5 Oe. Agglomeration occurs for both D.C. and 60 hz A.C. magnetic fields and for $\underset{\sim}{H}$ horizontal and vertical. These experiments are discussed more extensively in Peterson [2] and Peterson and Krueger [3]. Agglomeration also occurs in nonuniform magnetic fields.

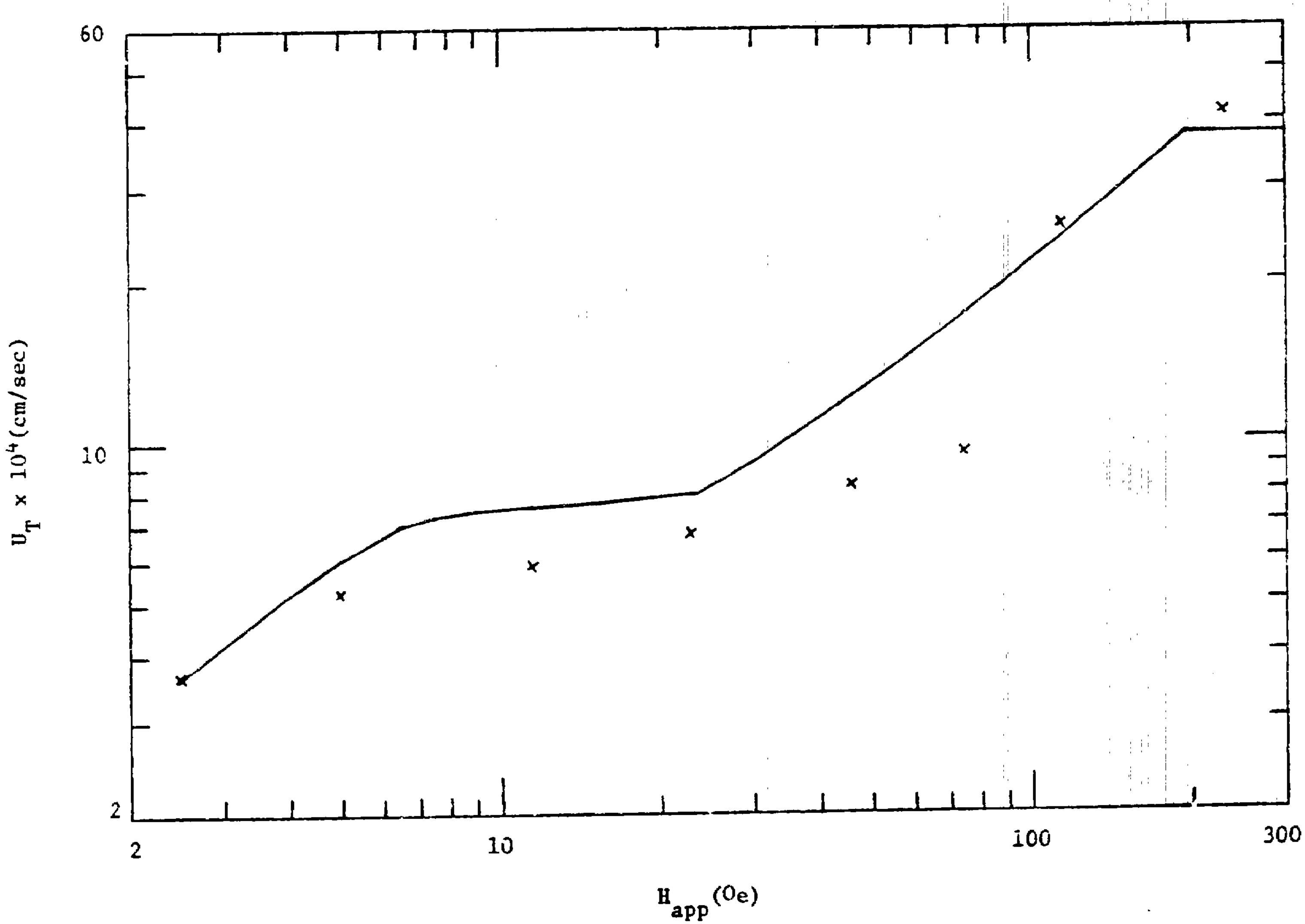

Fig. 2.3 Comparison of theoretical (—) and experimental(x) terminal velocities of the agglomerates vs. applied magnetic field.

C. Agglomeration - Theory

We have calculated the terminal velocity of the agglomerates as a function of applied magnetic field. The results were shown in Figure 2.3. The terminal velocity can be calculated easily once the shape and volume, v, of the agglomerate is known because the relevant Reynolds number is much less than unity so the flow is viscous dominated. We assume the shape is a prolate spheroid (i.e. cigar-shaped) aligned along the vertical magnetic field. The aspect ratio, k, is defined as the ratio of the semi-minor axis to semi-major axis and is thus less than or equal to one (equal for a sphere). To determine k and v we need two equations. Firstly we minimize the total energy of an agglomerate in a magnetic field. The energy has two contributions. The magnetic dipole-dipole energy tends to align the particles into long chains (i.e. makes k very small). On the other hand, the surface tension energy tends to favor a spherical agglomerate shape (k = 1). The second equation is a force balance on the particle on the trailing tip of the agglomerate. The viscous drag, F_D, as the agglomerate falls through the medium tends to pull the last particle from the agglomerate. On the other hand, the magnetic dipole-dipole force, F_M, attracts the particle to the agglomerate. For a marginally stable agglomerate we will have $F_M = F_D$.

In principle these two equations suffice to fix the volume, v, and aspect ratio, k, for an agglomerate. Before this can be done, there are several auxillary problems to be solved. The calculation of F_D is non-trivial because of the large shielding by the large agglomerate. If the agglomerate were a sphere of radius R_L and the particle has a radius R_S, then Goren [4] showed that the drag force F_D is proportional to R_S^2/R_L^2.

For our agglomerates this factor is roughly 10^{-6} so the drag is roughly 10^{-6} times the Stokes drag on an isolated particle. For non-spherical agglomerates which we almost certainly have, no calculations of F_D were available. Extending the multipole-expansion-collocation technique of Gluckman, et.al. [5], we found that for relative volumes of less than 10^4 a reasonable analytical approximation to F_D is

$$F_D = 19.4\pi \eta u^* R_S$$

where η is the viscosity of the ferrofluid and u^* is the velocity of the fluid at a distance R_S from the end of an isolated agglomerate. We have extrapolated this expression to larger size agglomerates where the collocation technique fails due to numerical problems. The present justification of this extrapolation is based on a reflection technique as discussed in Happel and Brenner [6]. Further discussion of this work is given in a paper submitted for publication by Liao and Krueger [7].

The drag force is proportional to the ferrofluid viscosity. Rosensweig [8] has shown that the viscosity increases by factors of 3 or 4 in strong magnetic fields. He also showed that the viscosity is shear dependent. We have fit his data with empirical curves and have used these in our evaluation of the drag force.

To evaluate the magnetic force, F_M, and the magnetic dipole-dipole energy, we must determine the arrangement of the magnetite particles within an agglomerate. For any applied field Luttinger and Tisza [9] have calculated the magnetic dipole-dipole energy at absolute zero of temperature for particles arranged in three lattice structures, simple cubic, face-centered cubic and body-centered cubic. We have shown that, for all applied fields, the simple cubic structure has the lowest energy. Thus we assume a simple cubic structure. Sullivan and Deutch [10] have analyzed a simple cubic structure of magnetic dipoles at zero applied field but for nonzero temperature. We have essentially interpolated from these two theories to obtain results for nonzero temperatures and nonzero fields.

The result of all this is a rather complicated set of five highly nonlinear equations which must be solved by iteration. The surface tension coefficient is fixed by fitting the theory to experiment at the lowest field point. Despite the complexity, the results as shown in Figure 2.3 are qualitatively correct and quantitatively quite good.

In Figures 2.4 and 2.5 we show the results for the aspect ratio, k, of the agglomerates and their volume as a function of applied magnetic field. The particles are very elongated ($k \approx 10^{-3}$) for quite small fields (~20 Oe). The number of particles in an agglomerate is very large ($\sim 10^{10}$) for similar fields. As expected the agglomerates become larger and more elongated as the magnetic field is increased. A more detailed discussion of the theory is given in Liao [11] and Liao and Krueger [12].

D. Areas for Further Investigation

Perhaps the most important area for further work is to investigate the effect of shear stresses on agglomeration. Agglomeration in a static fluid is important because many devices such as seals or magnetic ink jet printers will be turned off for long periods of time. In addition the effects of agglomeration in a dynamic setting are potentially

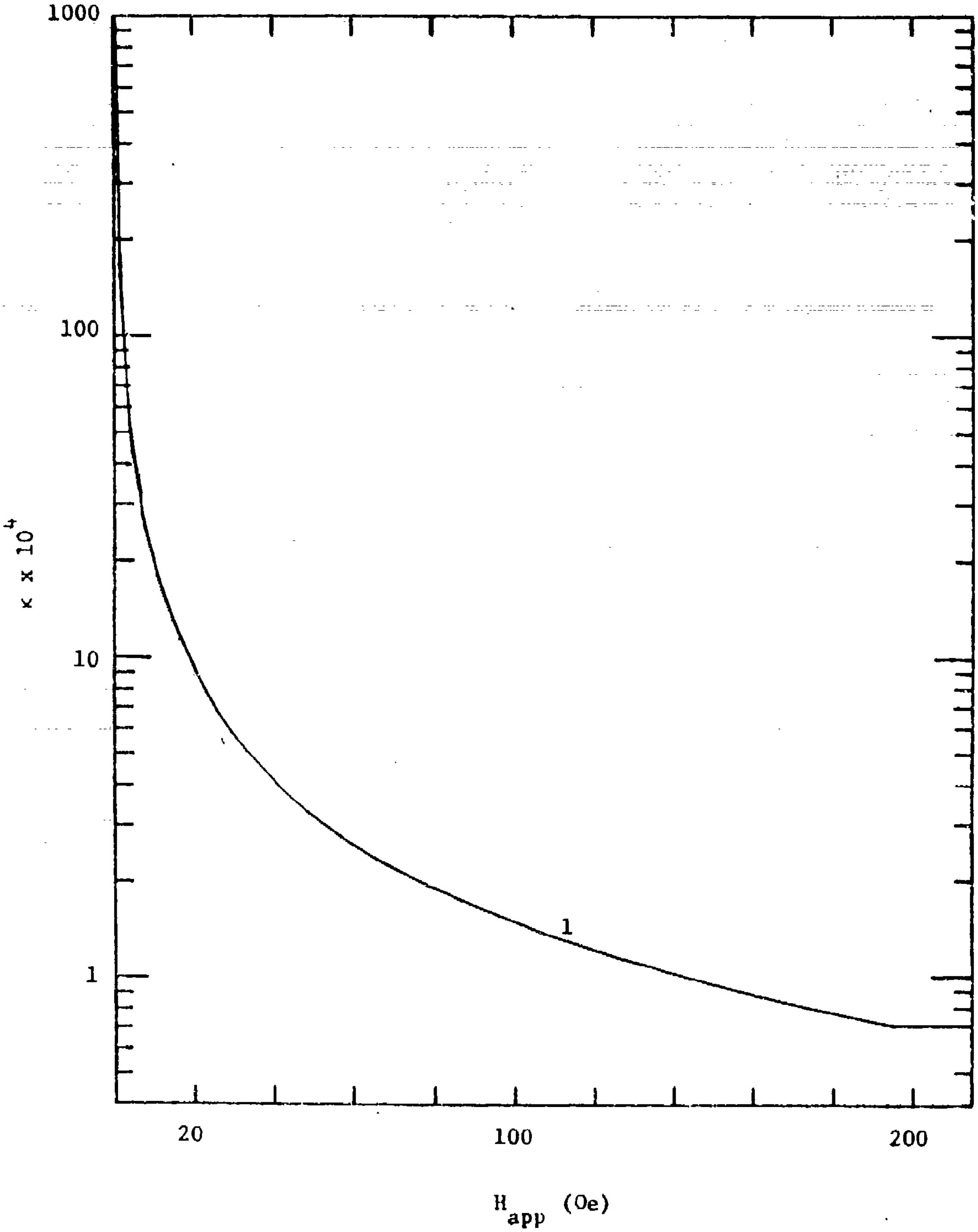

Fig. 2.4 Aspect ratio, κ, of the agglomerates vs. applied magnetic field.

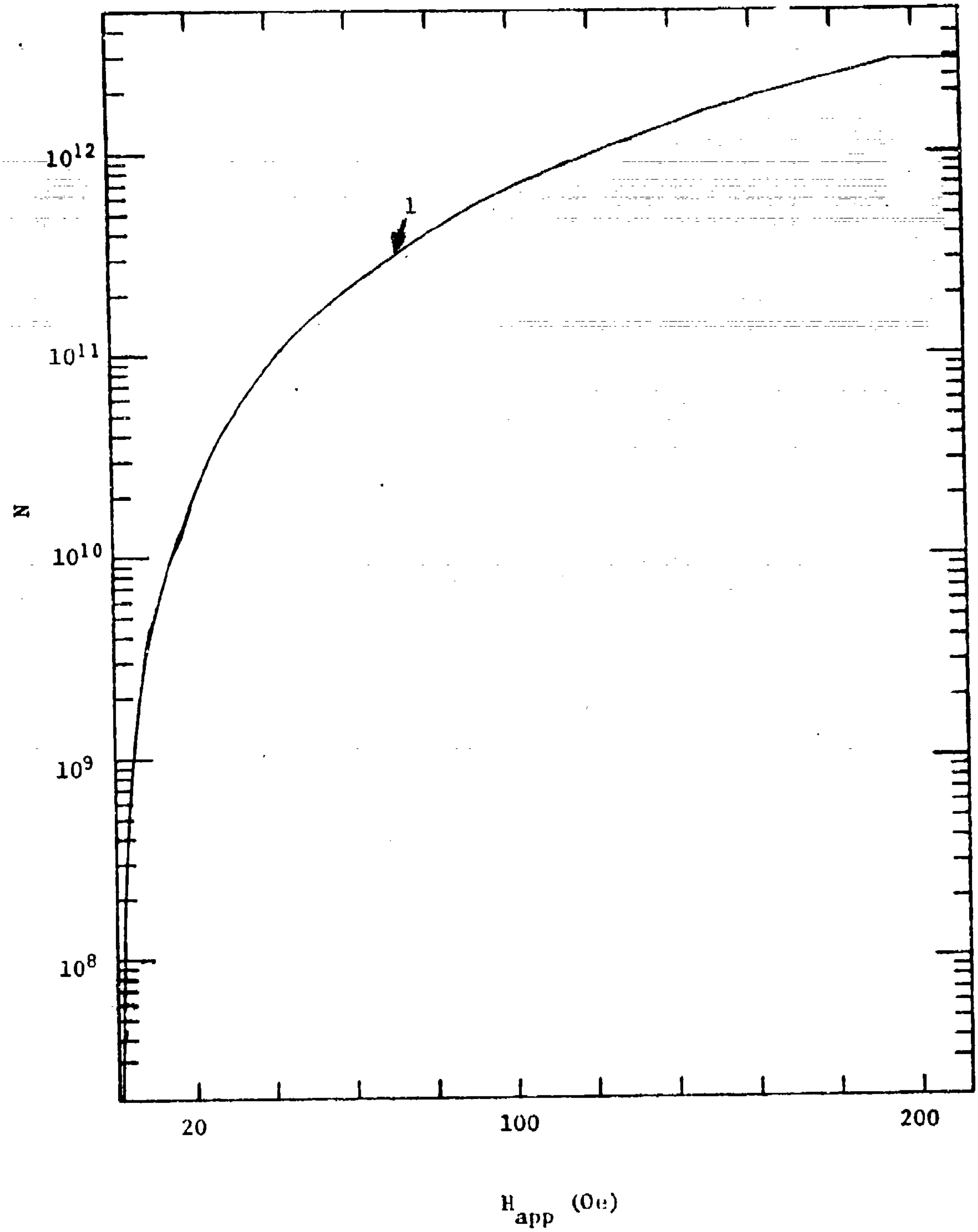

Fig. 2.5 Number of magnetite particles per agglomerate, N, vs. applied magnetic field.

important. Effort should go into deciding if there is a threshold shear stress for breaking up agglomerates. Additional effort should go into understanding the shear and magnetic field dependence of the viscosity of ferrofluids.

III. Electromechanics Studies

The essential feature of the response of ferrofluid to a magnetic field is electromechanical. It is attracted into regions of more intense magnetic field. The force is exerted on the individual permanent magnetic dipoles of the ferrite particles by an imposed non-uniform magnetic field. This force is

$$\overline{f}_i = \mu_o \overline{m}_i \cdot \nabla \overline{H} \tag{3.1}$$

where $\mu_o = 4\pi \cdot 10^{-7}$ H/m is the permeability of free space, $\overline{m}_i$ is the magnetic moment of a ferrite particle and $\overline{H}$ is the externally imposed magnetic field. The net force per unit volume may be obtained by summing the forces on all the individual particles:

$$\overline{F} = \mu_o \overline{M} \cdot \nabla \overline{H}, \tag{3.2}$$

where $\overline{M}$ is the magnetization vector. Eq. (3.2) neglects certain inter-particle (dipole-dipole) interactions which have no effect if one only wishes to calculate the *total* force on a mass of ferrofluid. Another more common force density, attributed principally to Korteweg and Helmholtz, and derivable from energy considerations [13] is:

$$\overline{F} = -\tfrac{1}{2} H^2 \nabla \mu \tag{3.3}$$

where μ is the magnetic permeability of a linear ferrofluid. Most ferrofluids are not linear, requiring a more generalized derivation. The general result, valid for non-linear ferrofluids, is [14]

$$\overline{F} = - \sum_i \frac{\partial w'}{\partial \alpha_i} \nabla \alpha_i , \tag{3.4}$$

where w' is the co-energy density

$$w' = \int_0^{H^2} \tfrac{1}{2} \mu dH^2 \tag{3.5}$$

and μ is the generalized (non-linear) permeability as defined by

$$\overline{B} = \mu(\alpha_1, \alpha_2, \ldots, \alpha_n, H^2)\overline{H}. \tag{3.6}$$

The α_i's are fluid properties such as density, ferrite particle concentration, etc. Note that in Eq. (3.3) and (3.4) rate-dependent and hysteretic magnetization effects are neglected and the continuum approximation for the colloidal suspension is assumed.

A. Height of Rise Experiment

To gain confidence in the behavior of ferrofluid, a classical experimental technique attributed to Quincke [15] was repeated using a variety of types of ferrofluids. The method, used to measure the magnetic properties of weakly susceptible liquids, involves the accurate measurement of the height to which a column of ferrofluid will rise in a glass tube when subjected to a uniform transverse magnetic field. See Fig. 3.1. The height of rise h is a function of the applied uniform magnetic

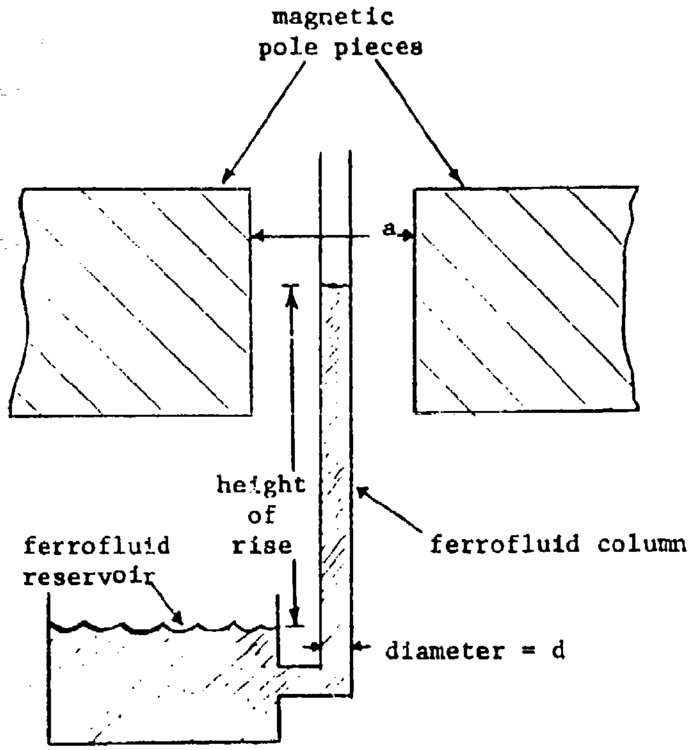

Fig. 3.1 Illustration of experimental apparatus for determining the magnetization properties of ferrofluid samples using the Quincke method.

field H

$$h = \frac{\mu_0}{\rho g} \int_0^H M(H')dH', \qquad (3.7)$$

where ρ is the density of the ferrofluid and $g = 9.81\ m/sec^2$ is the acceleration due to gravity. The density of air is neglected here. Representative experimental results for two ferrofluid samples are shown in Fig. 3.2. As a check on these data, a "theoretical" height of rise was calculated using M(H) data obtained from vibrating sample magnetometer data (see section II) in Eq. (3.7). The agreement is well within the uncertainty of the measurements.

The above measurements were carefully made using only well-mixed ferrofluid samples. Long exposure of the ferrofluid to the magnetic field affected the height of rise data. This effect depended on the length of the exposure. It is supposed that this effect is the same as that discussed in section II.B above and attributed to particle agglomeration.

B. Parametric Surface Wave Dynamics

Parametrically induced surface perturbations may be important in the free surface pumping of ferrofluids, and so parametric surface instability was investigated. A fairly considerable background of work on the interfacial dynamics of ferrofluids existed prior to the start of this research. However, parametric surface waves had not been studied. The experiment is shown in Fig. 3.3. The parametric surface instability exhibits a distinct threshold which is dependent on the frequency and the magnitude of the magnetic field. Above this threshold,

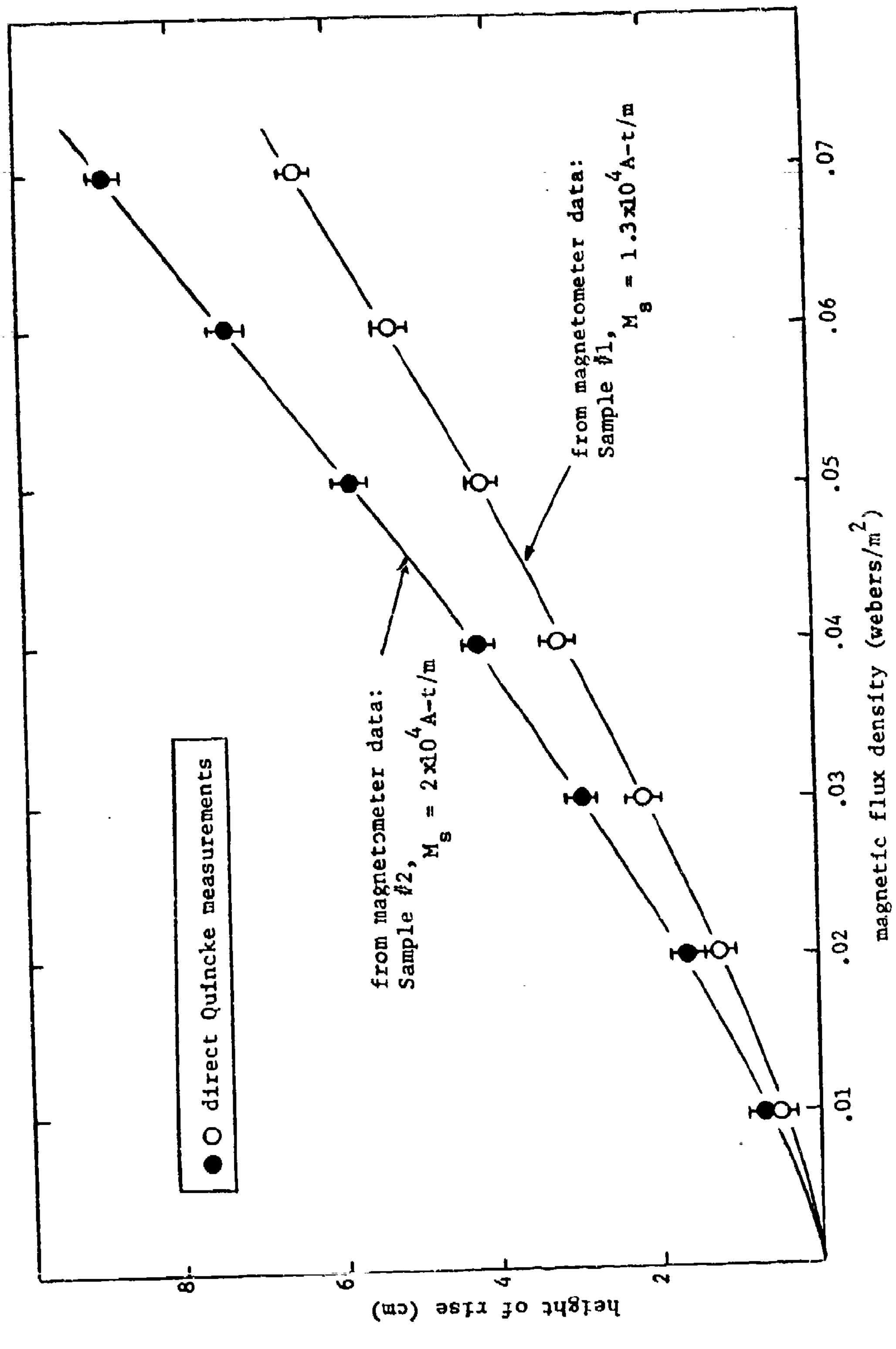

Fig. 3.2 Height of rise versus external DC magnetic flux density.

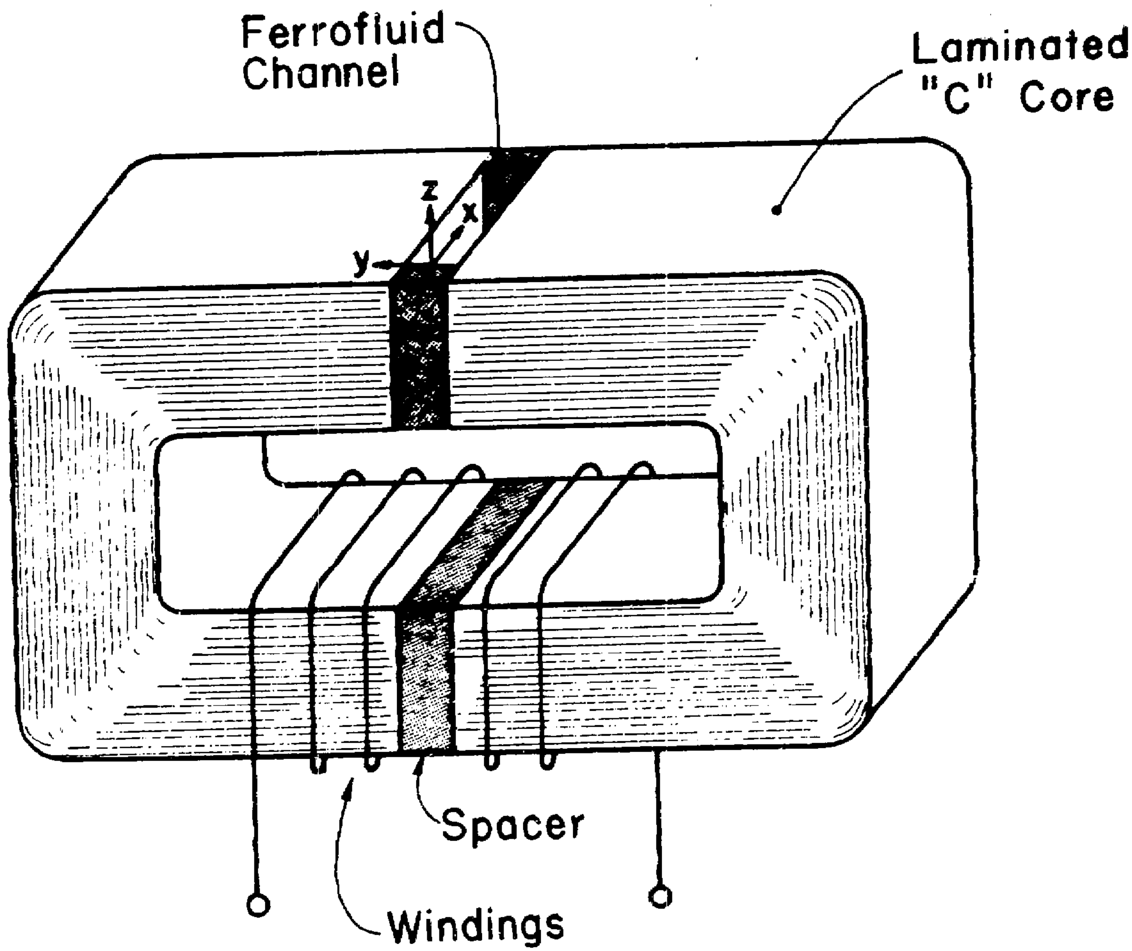

Fig. 3.3 Sketch of transformer core, windings and liquid channel for parametric surface instability experiment.

perturbations spontaneously arise which grow to a certain amplitude and take the form of a standing wave pattern with an easily discerned wavelength. This wavelength also depends on magnetic field frequency and magnitude.

The theory of interfacial parametric ferrohydrodynamics is analogous to that of electrohydrodynamics [16], except for the non-linearity of ferrofluids. Therefore, an analytical model for the M(H) relation was required. The superparamagnetic theory, generally considered to be accurate for ferrofluids, was employed using a single Langevin function:

$$M(H) = M_s[\coth(\alpha H) - 1/(\alpha H)] \tag{3.8}$$

Eq. (3.8) ignores the particle size distribution, substituting an average particle size which best fits the measured M(H) curve. This approach is as much a curve-fitting as a modeling exercise.

Perturbations of the surface are assumed to be spatially periodic. Using Eq. (3.8) and balancing forces at the interface gives the surface equation of motion. This equation fits the canonical form of Hill's equation. At given magnetic field frequency and magnitude, a certain band of wavelengths is unstable and one wavelength value has the fastest growth rate. Wavelengths predicted by this "most unstable mode" method are compared to the experimentally observed standing wave pattern in Fig. 3.4. Given the approximations used in the theoretical model, the agreement is reasonable, suggesting that the "most unstable mode" hypothesis accurately describes the development of the non-linear, amplitude-limited standing wave pattern.

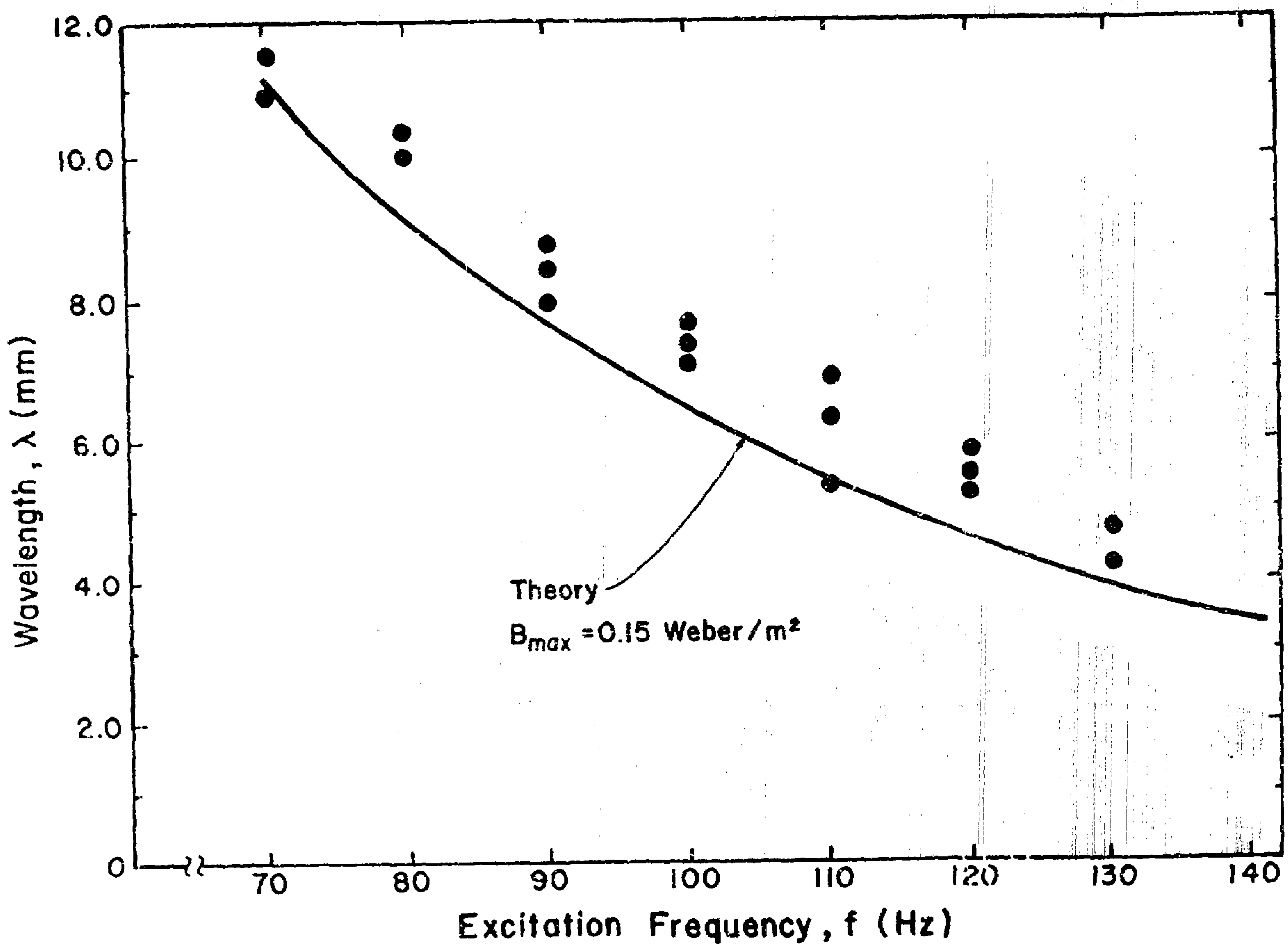

Figure 3.4 Plot of experimental data and most unstable wavelength theory versus frequency for parametric *ferrohydrodynamics* experiment.

C. Ferrofluid Seals

Interest in shaft seals for power plants and propulsion systems suggested the investigation of ferrofluid seals. Presently, a line of small (≈ 3 cm diameter) ferrofluid shaft seals is commercially available and, further, larger ferrofluid shaft seals for cryogenic and vacuum systems are being developed. Very little literature on the design and operation of ferrofluid rotary shaft seals exists. The study reviewed below was initiated to gain understanding of how these seals work, to identify important design parameters, and to develop a model which explained the observed behavior.

Static Plugs

First experiments were conducted with static seals in order to gain understanding of the essential features of ferrofluid sealing. Refer to Fig. 3.5. The static plug is established by concentrating the magnetic field along a short section of a glass tube. Ferrofluid is injected into this region. Then the plug is loaded from above by an immiscible liquid. Using this very simple arrangement, the important features of static ferrofluid plugs could be observed. Because of the liquid loading, plug failure is not catastrophic; no appreciable ferrofluid is lost. A failure channel as illustrated in Fig. 3.6 forms and the upper liquid passes through the seal along the tube 90° off the axis of the imposed magnetic field, where the field is the weakest. These leaks can usually be stopped by increasing the magnetic field strength.

The model developed to describe the behavior of the ferrofluid plug is based on the so-called modified Bernoulli equation [17]:

$$p + \rho v^2/2 - \mu_o \int_0^H M(H')dH' = \text{constant}, \qquad (3.9)$$

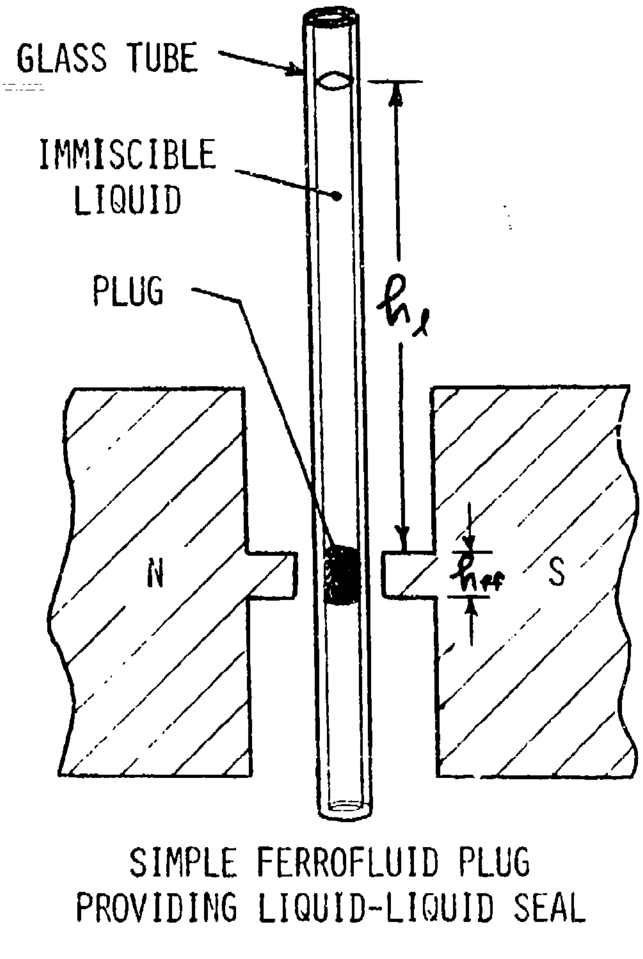

Fig. 3.5 Ferrofluid Plug Experiment

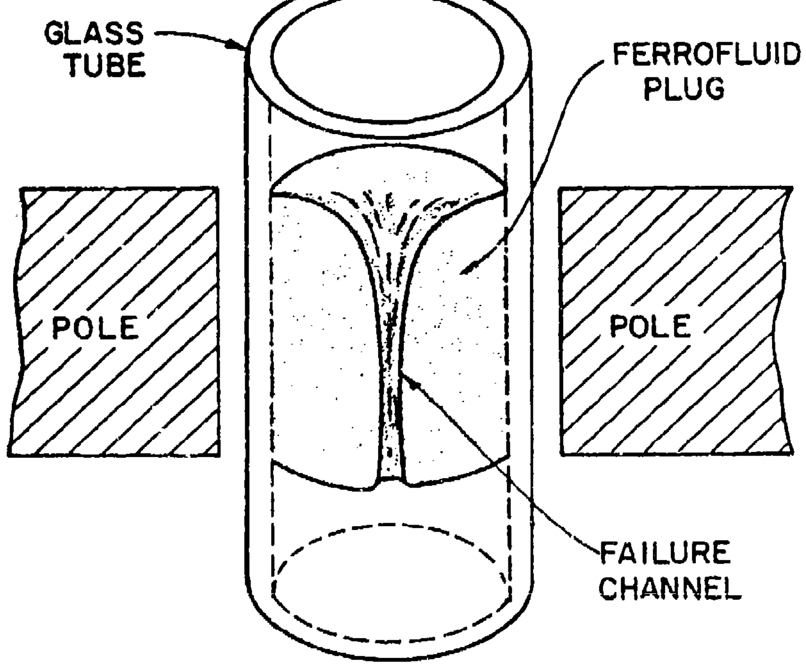

Fig. 3.6 Details of failure of ferrofluid plug when maximum loading capacity is slightly exceeded. Small channel is formed 90° away from poles because of small variations in magnetic field within the plug.

where p is the pressure and v is the fluid velocity (zero in the hydrostatic limit considered here). Using Eq. (3.9) for the idealized plug shown in Fig. 3.7, we have

$$p_1 - p_2 = \mu_o \int_{H_1}^{H_2} M(H')dH' \tag{3.10}$$

where p_1 and p_2 are the hydrostatic pressures on either side of the plug, and H_1 and H_2 are the essentially tangential magnetic field strength magnitudes at the two interfaces. This model clearly shows how the plug is displaced as it is loaded. The maximum loading occurs when the interface on the high pressure side is located at the maximum field intensity with the interface on the low pressure side in the region of zero magnetic field:

$$\Delta p_{max} = \mu_o \int_0^{H_{max}} M(H')dH'. \tag{3.11}$$

Strictly speaking, this limit is not achieved, but in a properly designed plug it is closely approached. Note that Δp_{max} can not be approached if the inventory of ferrofluid is too small. The experimental static plug burst pressure is compared to the prediction of Eq. (3.11) in Fig. 3.8.

As shown in Fig. 3.8, the model described above successfully predicts the burst pressure of static seals. This success is deceptive, because the theory does not account for interfacial stability. Under certain conditions, Eq. (3.10) will predict successful sealing, yet experimentally the seal fails. This apparent discrepancy is invariably due to interfacial instability which permits the fluid being sealed to

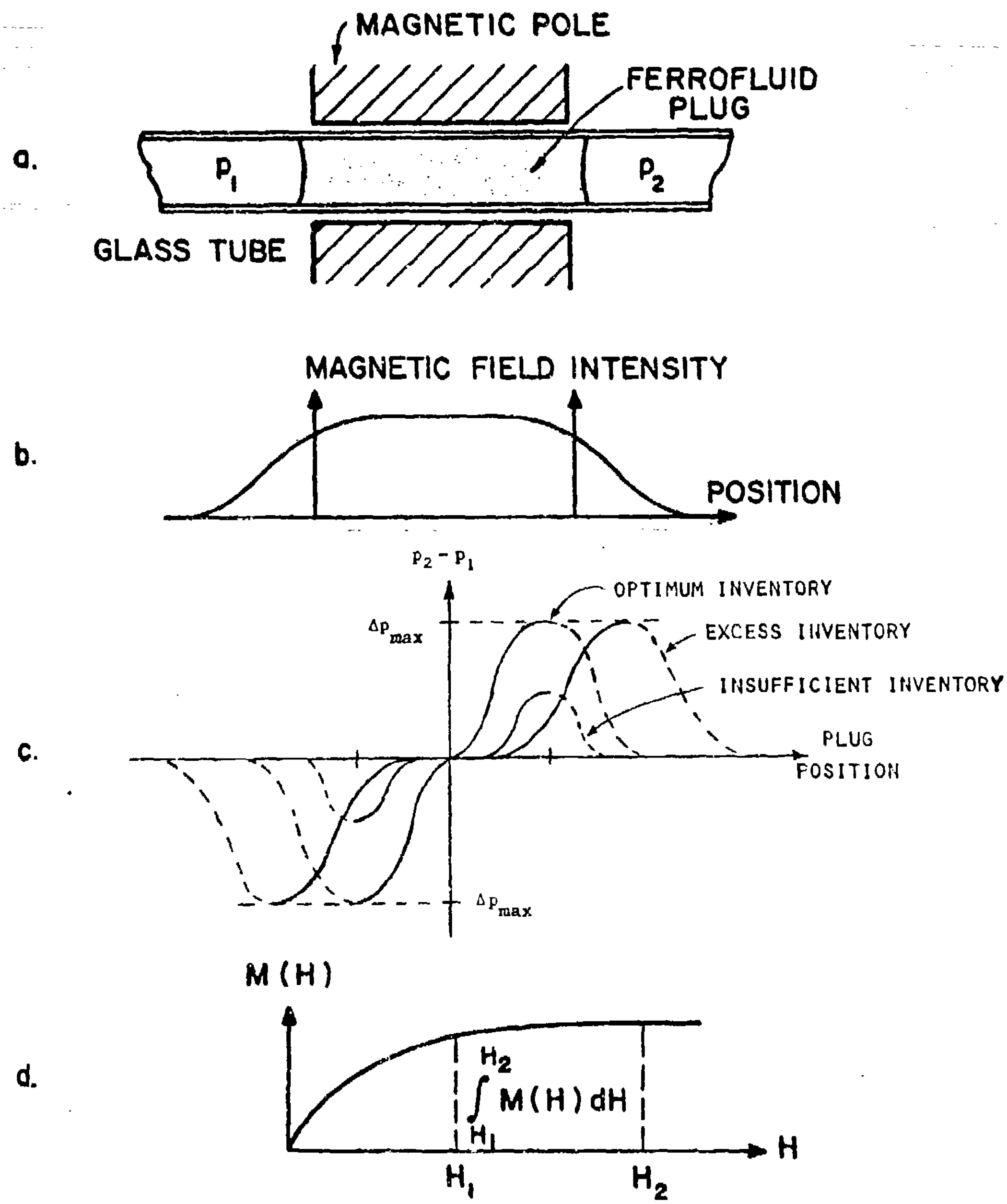

Fig. 3.7 a) Static ferrofluid plug between opposite poles.
b) Symmetric field distribution versus position.
c) Hydrostatic loading for varying amounts of ferrofluid versus seal center position.
d) Typical M vs. H relationship for ferrofluid.

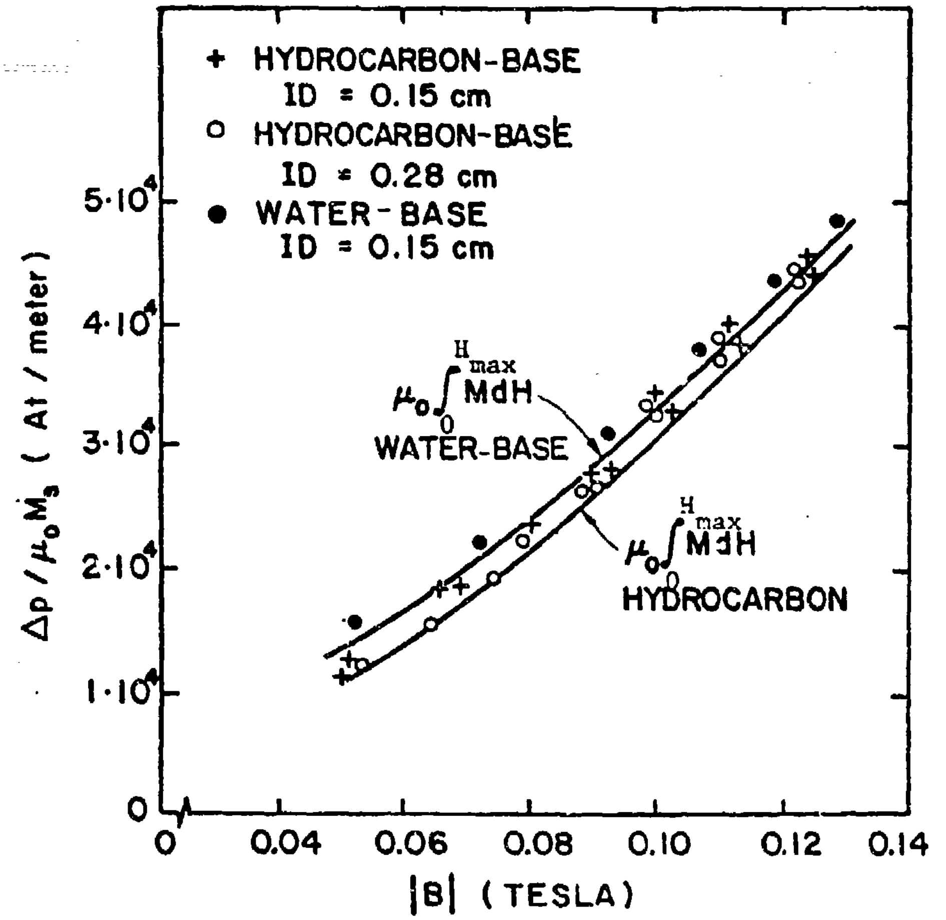

Fig. 3.8 Maximum hydrostatic loading of ferrofluid plug versus external (transverse) flux density for 190 gauss hydrocarbon-base and 250 gauss water-base ferrofluid. Maximum inventory is employed.

pass through the ferrofluid layer. This behavior is more common in liquid-liquid sealing where the low interfacial tension permits the Rayleigh-Taylor instability to destabilize the ferrofluid-liquid interface. This instability can be suppressed if the uniform magnetic field region is kept very small, so that the interfaces are stabilized by the non-uniform magnetic field. This matter is discussed in some detail by Perry [18].

Dynamic Rotary Seals

A simple version of a rotary shaft seal employing ferrofluid is shown in Fig. 3.9. This seal lacks the multiple stages commonly employed in commercial units, but it is useful as an experimental tool. A dynamic seal is in many ways similar to a static plug; however, the rotational fluid dynamics complicates the device behavior. Burst pressure measurements for the device shown in Fig. 3.9 were made under a variety of conditions. These included variable gap spacings, variable magnetic flux, and variable speed. Some representative data are shown in Fig. 3.10. The different curves are for different ferrofluid inventory values. Note that the seal performance is dependent on inventory as predicted by the static seal model (see previous section). Also note that rotation effects are prevalent only at lower inventory. The static burst pressure for very low inventory is found to be a small fraction of the value for the dynamic (rotating) case. Both the rotation-dependent effects seem to be related to ferrofluid redistribution caused by centrifugal forces, however, this hypothesis is difficult to verify due to the difficulty in observing the ferrofluid location within the seal during rotation. Likewise, the temperature of the ferrofluid is difficult to monitor *in situ*.

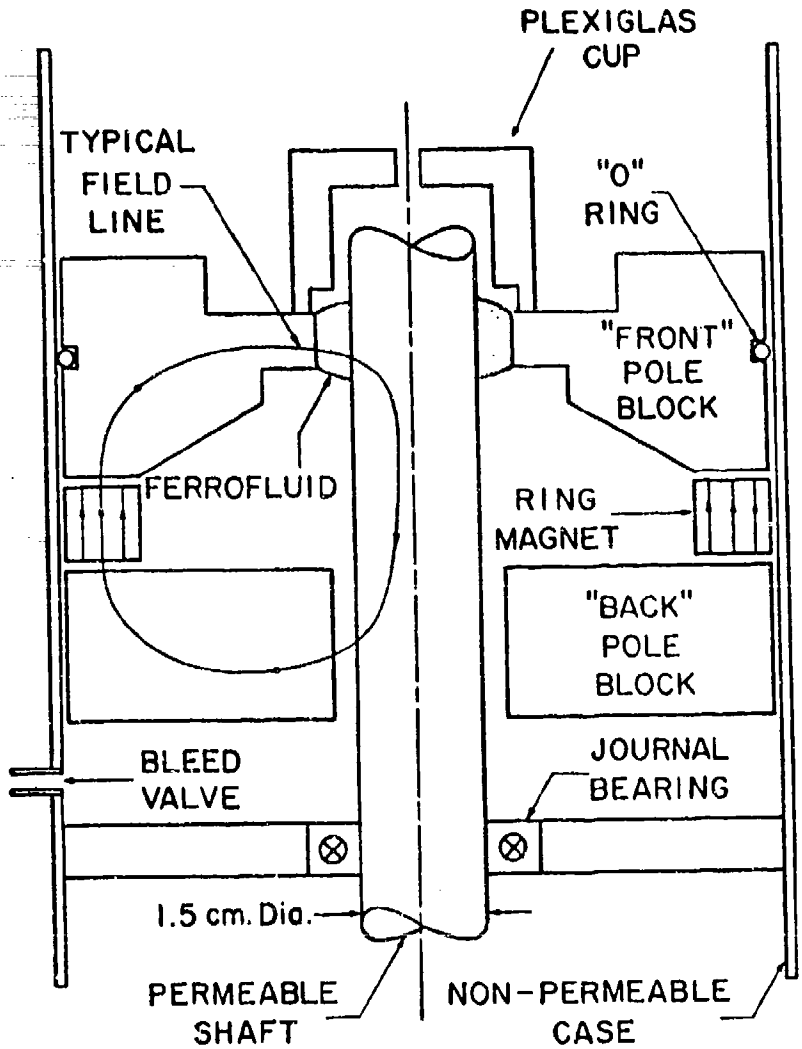

Fig. 3.9 Cross-section of experimental ferrofluid seal and test fixture. Seal is loaded from below by evacuating region above seal.

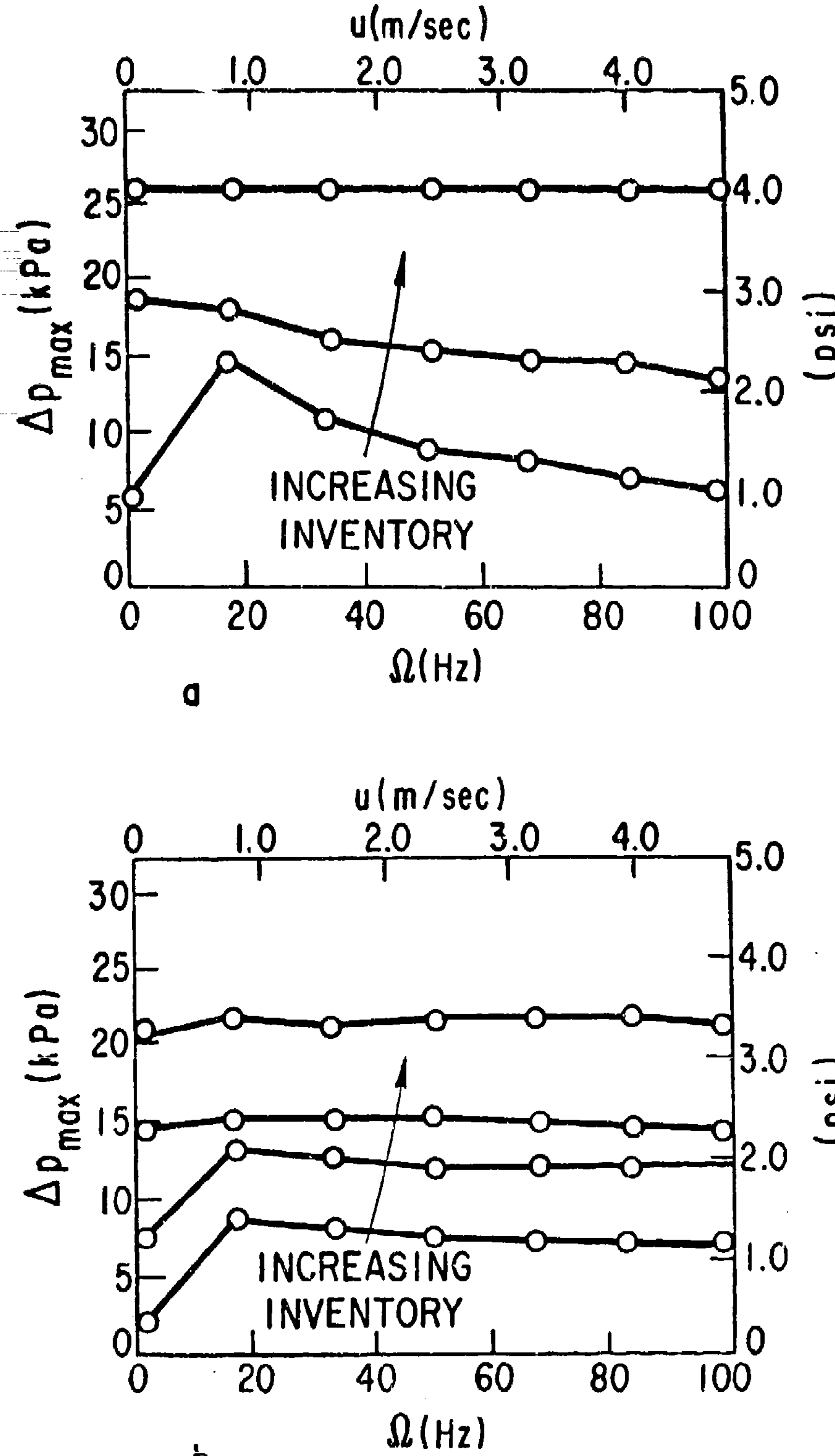

Fig. 3.10 Maximum capacity of a single stage constant inventory ferrofluidic seal, a) Alnico magnet (substantial leakage fields) with clearance of .038 mm (.0015 in) versus rotation speed, b) Plastiform® magnet (reduced leakage fields) with clearance of .15 mm (.006 in) versus rotation speed.

Attempts to develop a realistic theoretical model for the seal are frustrated by the complex nature of the problem. For the device tested, magnetic field leakage due to pole block saturation made the true location of the seal uncertain under load. Further, the influence of the rotation on the ferrofluid location and dynamics was dependent on the ferrofluid location. If leakage is neglected, the model is not capable of explaining many of the observed phenomena. Thus, the modeling exercise hinges on solution of the non-linear problem of ferrofluid in a gap with strong leakage fields, variable inventory, and variable rotation speed. As yet, only Bailey, et.al. [19] and Perry [18] have attempted the task of modeling a dynamic seal. Bailey, et.al. [19] modeled a conventional type of seal for a large diameter shaft, using an energy method. Perry used the surface force balance method based on the ferrofluid plug model to analyze a ferrofluid seal with a transverse magnetic field and a non-magnetic shaft. Perry's model, though very crude, predicted the effect of rotation on seal burst pressure. No one has yet developed a solution for a conventional ferrofluid seal (with magnetized shaft) which predicts the interfacial profile of the ferrofluid.

IV. Summary and Conclusions

Over the three year period of the research contract, the basic magnetization properties of ferrofluids have been studied. Reversible agglomeration effects not previously reported have been observed in water-base, ester-base and hydrocarbon-base ferrofluids. This agglomeration is dependent on the applied magnetic field, and it occurs in uniform and non-uniform fields as low as 5 Oe. From the measured settling velocities of approximately 10^{-3} cm/sec, the apparent agglomerate sizes have been inferred in the range of 10^9 particles. The agglomerates are elongated in the direction of the applied magnetic field. Theoretical predictions of the maximum agglomerate size are based on a hydrodynamic model which permits drag calculation plus an energy minimization technique which permits determination of the particle size and shape. The work is substantiated by independent observation of agglomerate particles made by Hayes [20]. The agglomeration of water-base ferrofluid has potential implications in scrap-metal separation technology where large volumes of magnetic liquid are employed to separate out non-ferrous metals using strong magnetic fields. The requirement of cheap ferrofluid, the large volumes involved, and the strong magnetic field imply that proper recognition of agglomeration and its effect on magnetization in ferrofluids is essential. The useful static shelf life of certain ferrofluid seal and/or bearing components is also a cause for concern if the ferrofluid is subject to agglomeration. All the ferrofluids tested showed a complete recovery from agglomeration upon gentle mixing, so, once recognized, agglomeration problems are likely to be easily solved in any application.

Over the same period, primarily basic studies of the electromechanics of ferrofluids have been conducted in order to test the applicability of present continuum models for the force on a ferrofluid. Miller and Resler [21] have questioned the generally accepted models. No experiments similar to those of Miller and Resler were attempted in this study; the experiments conducted did not indicate any breakdown of present continuum models, nor did they rule out the possibility that the discrete nature of magnetic liquid colloids exhibit non-continuum behavior. Certain experiments, notably the height of rise measurements did uncover a time-dependent effect on magnetization, probably attributable to agglomeration which affected the ferrofluid response to strong fields. This effect in itself is not a breakdown of the continuum model, but rather a feature of the magnetization. However, the distinct possibility exists that the large agglomerate particles might lead to a breakdown of the continuum model. This is because the agglomerates are up to 10^9 times larger than the individual sub-domain ferrite particles themselves.

The parametric surface wave dynamics have been studied, and are fairly well understood at this point. The potential for application of this work to either synchronous interfacial pumping of ferrofluid or magnetic liquid jet printing has not yet been pursued.

A model for a static ferrofluid plug has been developed and tested by experiment. The model is sufficiently simple and general to describe all the essential features of ferrofluid sealing. Intensive experiments have been conducted with a single-stage rotary shaft seal. The influences on the burst pressure of such device parameters as ferrofluid inventory, gap spacing, magnetic flux and rotation have been examined carefully. Some of the features of the observed behavior have been found to be consistent with the static plug model and some have not. The complexity of the rotational seal problem has thus far prevented the successful modeling of the rotary seal. The successful application of ferrofluid seals to larger shaft sizes and higher surface velocities, as proposed for shaft seals in cryogenic power system applications, propulsion systems, etc. depends on improved understanding of the basic mechanisms involved. Considerably more work on the modeling of dynamic ferrofluid seals is warranted.

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VI. Other Work

A. International Advanced Course and Workshop on the Thermomechanics of Magnetic Fluids

In response to a request from Dr. B. Berkovsky, one of the investigators (T. B. Jones) has prepared a series of three lectures to be presented during October, 1977, at the "International Advanced Course and Workshop on the Thermomechanics of Magnetic Fluids". This course is sponsored by UNESCO and is being held at the International Centre for Mechanical Sciences in Udine, Italy. The lectures to be presented concern the theoretical treatment of problems in ferrofluids, with emphasis on application to seals. The lecture series, entitled: "Theory and Application of Ferrofluid Seals", is divided into three sections which are 1) "Force density formulations for magnetic fluids", 2) Auxiliary fo~ e calculation methods and solution of simple ferrohydrostatic problems", and 3) "Static ferrofluid seals and plugs". The abstract is reproduced below:

Theory and Application of Ferrofluid Seals

These lectures on the theory and application of ferrofluid seals are divided into three topics including the formulation of forces in ferrofluids, solution of various ferrohydrostatic equilibria, and consideration of static ferrofluid seals and plugs. The important ferrofluid force formulations include $\mu_0 \overline{M} \cdot \nabla \overline{H}$, and certain force densities derived from energy considerations. Each of the force densities has associated with it an appropriate stress tensor. In the case of incompressible magnetic fluids, ambiguities in the force density arise which can be shown to be associated with the arbitrariness of hydrostatic pressure. Despite the diversity of these body force formulations, they can all be reconciled and shown to be consistent if integrated over the entire volume of the ponderable body in question. Application of these various force formulations and demonstration of their mutual consistency is best shown by working various simple examples of ferrohydrostatic equilibria. Certain of these geometries may be solved for ferrofluids with arbitrary, non-linear magnetization characteristics. Carefully working these problems provides a solid basis for understanding the electromechanical response of magnetizable liquids in non-uniform magnetic fields. The single most important application of ferrofluids right now is in vacuum and pressure sealing. Static plugs and rotational seals are illustrative examples of ferrohydrostatic and ferrohydrodynamic equilibria which are amenable to solution by a number of different approaches, some more convenient to use than others. Models developed to analyze the behavior of static ferrofluid seals lead the way to the design of seals with performance characteristics suited to various applications.

B. Bibliography for Ferrofluid Research

E. A. Peterson [23] compiled a bibliography of 176 references on ferrofluid research, which Perry updated in 1976 [18], adding 26 new references. This bibliography has been updated once again, by the addition of 26 new papers. The bulk of these papers have appeared within the past 15 years, indicating widespread interest and activity in ferrofluid research.

SECOND SUPPLEMENT TO BIBLIOGRAPHY FOR FERROFLUID RESEARCH

September 1977

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VII. Reports, Papers, Presentations

Reports

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