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USE OF HIGH-GRADIENT MAGNETIC FIELDS FOR THE SEPARATION OF MACROMOLECULES

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ABSTRACT

High gradient magnetic field separation (HGMS) has been used to separate several types of > 1 μm sized ferro- or paramagnetic particles from bulk streams. The majority of the studies have been carried out using a single ferromagnetic wire or wire mesh to produce the field gradients necessary for particle capture. The purpose of this paper is to examine the possibility of using HGMS on < 1 μm entities for the purpose of macromolecular separations. Preliminary experimental results demonstrate that HGMS techniques can be used to capture 0.1 μm diam latex beads from a paramagnetic salt solution passing through a columnar bed of ferro-magnetic spheres.

INTRODUCTION

High gradient magnetic field separation processes (HGMS) have been utilized for a number of applications in the separation of > 1 μm ferro- magnetic and paramagnetic particles. Among the materials separated using this technique are red blood cells (1), ore tailings, (2), and sewage, (3). The normal mode of operation in these processes is to place a ferromagnetic wire mesh into a uniform magnetic field; thereby, creating large magnetic field gradients near the surface of the wires. The desired material is separated via magnetic-field-capture from the bulk stream onto the wire mesh.

In general, the ability of a HGMS system to capture particles depends upon the magnitude of the magnetic susceptibility of the

particles. The magnetic susceptibility is defined as the ratio of the magnitude of particle magnetization in a field, to the intensity of the imposed magnetic field. For ferromagnetic materials the susceptibility is large (on the order of 10^3 to 10^5); therefore, imposition of a moderate magnetic field can induce the material to create a more intense magnetic field in its vicinity. However, this behavior is not without bound. If the intensity of the imposed magnetic field is increased past a critical point, ferromagnetic materials will reach a state of magnetic saturation where all the atomic dipole moments are aligned. Hence, at field strengths past the magnetic saturation of the material there is no further increase in the contribution to the total magnetic field by the ferromagnetic entity.

Paramagnetic materials have small magnetic susceptibilities when compared to ferromagnetic materials. Paramagnetic susceptibilities are on the order of 10^{-3} and are represented by salts and transition elements. The qualitative behavior of paramagnetic substances in HGMS processes is the same as ferromagnetic substances; however, for an identically sized particle, interactions with the imposed magnetic field are much weaker. Diamagnetic materials include most organic and inorganic compounds. The magnetic susceptibility is small and negative, on the order of -10^{-6}.

In a high-gradient field system, ferro- and paramagnetic materials are drawn into regions of relatively high field strength while diamagnetic materials migrate into low-field-areas. Although ferro- and paramagnetic materials are usually considered appropriate for HGMS systems, it is possible to contemplate conditions under which magnetic field interactions with < 1 μm, diamagnetic particles could be appreciable. This would involve the use of intense magnetic fields (> 1 tesla) and may involve the use of magnetically susceptible additives in the carrier solution of the particles.

IMPORTANT CONCEPTS OF HGMS

The basic equations describing the interactions between particles and ferromagnetic wires placed in high intensity magnetic fields have been examined by a large number of authors (4, 5, 6). One of the most comprehensive on the subject was written by Watson (3). He investigated capture of paramagnetic particles by a ferromagnetic wire placed in a uniform magnetic field. In his treatment, Watson assumed potential flow (of fluid) in order to calculate trajectory equations for particle motion in the vicinity of the wire. Figure 1 contains a schematic diagram of a typical set of particle trajectories in a HGMS system. Dependant upon the relative magnitude of viscous and magnetic forces, a particle may be captured on the wire mesh packing or flow by the wire unaffected. Subsequent papers have verified and extended the results of this treatment.

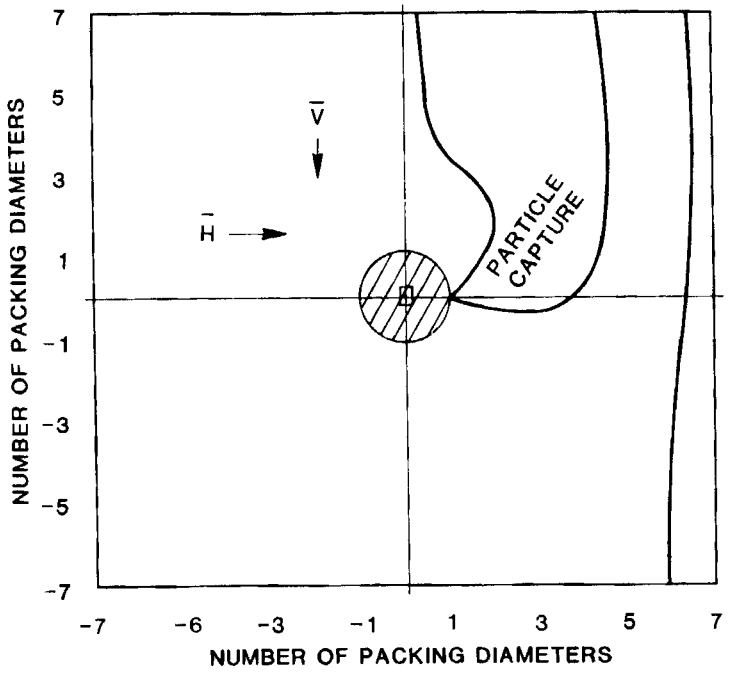


Fig. 1. Typical behavior of particles in HGMS separations schemes.

Numerous authors have addressed the concept of using ferromagnetic wires (meshes) to form a matrix for particle capture; however, only a few have investigated the interaction of diamagnetic particles with ferromagnetic spheres. Friedlaender, et al. (7) have published a limited amount of work in this area; hence, their contributions provide a good starting point for analysis. They derived and solved particle trajectory equations for the case of flow around a single ferromagnetic sphere placed in a magnetic field. It was assumed that the surrounding fluid was a solution containing a paramagnetic salt and entrained particles. Calculations were carried out for cases in which the magnetic susceptibility (of the particles in the fluid) was negative and one where the magnetic susceptibility was greater than zero. These systems were examined utilizing two models for flow around the sphere, (1) laminar and (2) potential flows. Experimental results tracing particle trajectories and noting particle capture indicated that the laminar flow model was in better agreement with experimental data than the potential flow model. The potential flow model at times predicted particle capture when experiment and the laminar flow model indicated no capture.

Although work has been published on modeling interactions with single ferromagnetic spheres, it appears that none has been carried out which contemplates the use of packed beds of ferromagnetic spheres for particle capture. There is some resemblance between the paramagnetic results and induced dielectric effects found in packed bed electrode systems, (8). There is, however, no parallel between diamagnetism and electric field effects.

In order to examine the potential for separation of diamagnetic material utilizing HGMS, one can perform a force balance on a particle flowing with a bulk liquid in the distorted magnetic field. Magnetic field gradients create relative motion between particles and the bulk fluid. For uncharged particles, the force balance is given by:

$$\overline{F}_m+\overline{F}_d+\overline{F}_g+\overline{F}_i=0, \tag{1}$$

where $\overline{F}_m$ is the magnetic force, $\overline{F}_d$ is the viscous force, $\overline{F}_i$ is the inertial force, and $\overline{F}_g$ is the gravitational force. Derivation of the expression for the magnetic force comes from looking at the change in energy by the introduction of the particle into the fluid (9). The work is proportional to the difference between the permeability of the particle and the fluid. The force is then the derivative of the work. The magnetic force is calculated using the following equation:

$$\overline{F}_m = 1/2\mu_o V_p(\overline{M}_p\cdot\nabla)\overline{H}, \tag{2}$$

where V_p is the volume of the particle, μ_o is the permeability of free space, $\overline{M}_p$ is the magnetization of the particle, and $\overline{H}$ is the magnetic field vector. The particle magnetization is given by the following equation (10):

$$\overline{M}_p = X\overline{H}/(1-X/3), \tag{3}$$

where X is the relative susceptibility of the solution and is represented by $(X_p - X_f)$ the difference between particle, X_p, and solution X_f, susceptibilities. Given that the susceptibilities of the particle and the salt solution are relatively small, the denominator in Eq. 3 is approximately equal to unity; therefore, the particle magnetization can be approximated as

$$M_p = XH \tag{4}$$

This relationship for particle magnetization makes sense when considering that the magnetic field produced by the diamagnetic or paramagnetic particle is small compared to the background field and the field produced by the ferromagnetic spheres. The magnetic field vector, $\overline{H}$, is obtained from solving Laplace's equation inside and outside the ferromagnetic sphere and adding contributions from the background field and the induced magnetic field.

The viscous force, F_d, is imposed on the particle as a result of the magnetic-field-induced relative motion of the particle with respect to flow of the bulk fluid. This relative velocity is assumed to be characterized by creeping flow; therefore, Stokes law can be used to calculate the viscous drag. This leads to the following relationship for the viscous force, where b is the

$$\overline{F}_d = 6\Pi\eta\ \overline{V}\ b \tag{5}$$

radius of the particle, η is the viscosity of the bulk fluid, and V is the relative velocity of the particle to the bulk fluid.

The force term due to gravitation is given by:

$$\overline{F}_g = V_p\ (\rho_p - \rho_f)\overline{G}, \tag{6}$$

where V_p is the volume of the particle, $\overline{g}$ is the gravitational field, and ρ_p and ρ_f are the densities of the particle and fluid, respectively. Because of the macromolecular (small) size of particles to be considered in this study, the force due to gravity can be neglected. Inertial forces should not come into play in this analysis because of the low Reynolds number flow assumed for the particle.

Substituting the proper expressions into Eq. 1 and solving for the components of the velocity vector of the particle yields the following equations (7):

$$\frac{dR}{d\tau} = (1 - \frac{3}{2R} + \frac{1}{2R^3})\cos\theta + \frac{AWK}{R^4}\ \frac{(1+3\cos 2\theta)}{2} + \frac{A(5+3\cos 2\theta)}{2R^3} \tag{7}$$

and

$$R\frac{d\theta}{d\tau} = -(1 - \frac{3}{4R} - \frac{1}{4R^3})\sin\theta - \frac{AKW}{R^5}(1 + \frac{A}{2R^3})\sin\theta \tag{8}$$

The characteristic parameters are defined below:

$$\tau = \frac{V_o t}{a} \tag{9}$$

$$R = r/a \tag{10}$$

$$A = M_s/3\mu_o H_o \tag{11}$$

$$W = \frac{\chi H_o^2 \mu_o}{\rho_p V_o^2} \tag{12}$$

$$K = \frac{2\rho_p b^2 V_o}{9a\eta} \tag{13}$$

where R and τ are the dimensionaless radial position and time, respectively, and θ is the angular position. "A" relates the saturation magnetization of the ferromagnetic material to the background field, K is the viscous term, and W relates the background field to inertial forces, a is the sphere radius, b is the particle radius, H_o is the background magnetic field, and V_o is the free stream velocity.

Multiplying A, W, and K together yields useful information about the ratio of the magnitude of the magnetic to viscous forces:

$$AWK = \frac{2M_s H_o \chi b^2}{27 V_o \eta a} \tag{14}$$

Examination of Eq. 14 reveals that the magnetic force is proportional to the effective magnetic susceptibility of the particle and to the square of the particle radius. Hence, if one is examining the possibility of effecting very small, diamagnetic particles, they must employ a very intense magnetic field (H_o), minimize the bulk fluid velocity (V_o) and size of the ferromagnetic packing (a), and enhance the effective magnetic susceptibility of the particle (χ) through the use of ferro- or paramagnetic additives.

EXPERIMENTAL SYSTEM

Placing a ferromagnetic sphere in a magnetic field causes large gradients in the magnetic field to occur near the surface of the sphere. If one imposes the same magnetic field upon a packed bed of ferromagnetic spheres, it causes large magnetic field gradients to occur in the interstitial volume of the bed. If a high-intensity magnetic field is imposed upon the bed, the combination of the high-intensity and high-gradient aspects of the magnetic field make possible the imposition of relative motion between small particles and the bulk fluid which are passing through the bed. The conditions under which a significant relative bulk fulid-particle motion can be developed is a function of (1) the magnetic susceptibility of the fluid and particles, (2) the size of the particles, (3) the size and magnetic susceptibility of the packing, and (4) the intensity of the magnetic field.

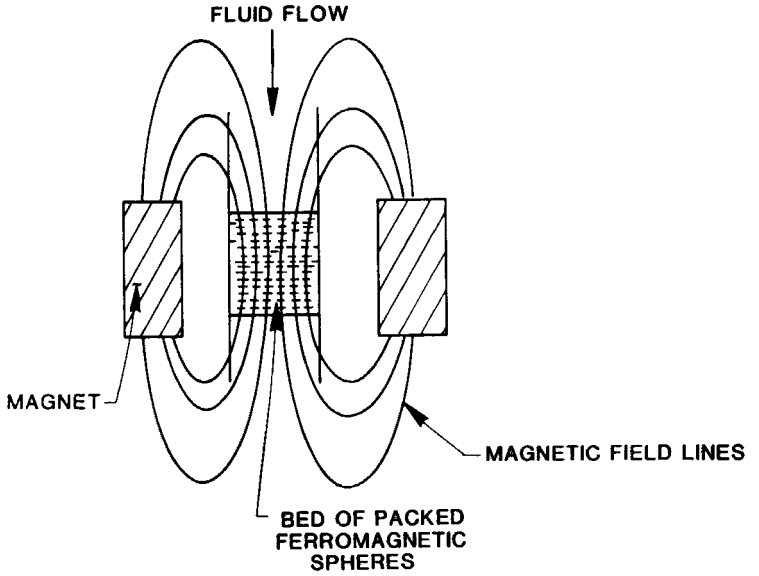
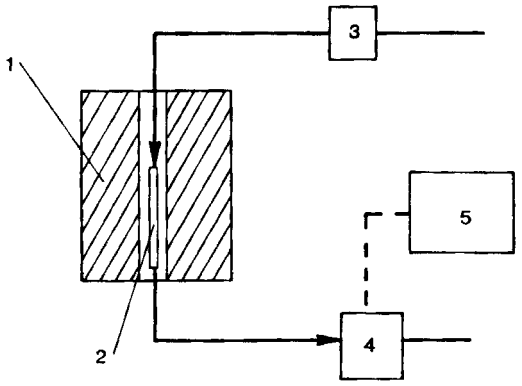


Fig. 2. Schematic diagram of the experimental packed-bed apparatus used in initial experiments.

Figure 2 is a schematic diagram of the experimental packed bed column in a magnetic field. A 6 tesla magnetic field is imposed on the bed of ferromagnetic steel spheres packed in a 3.0 in. long by 0.25 in. ID. Teflon column. The ferromagnetic spheres (Nuclear Metals Inc.) are made of 779 steel and have an average diameter of 100 μm. The magnetic field is generated by a superconducting solenoid manufactured by Cryomagnetics Inc.

Figure 3 contains a schematic diagram of the experimental system. An aqueous solution of 10 or 20% $MnCl_2$ was pumped into the packed column which is located in the magnet. A 70 μL sample may be injected into the salt solution and carried into the column. A spectrophotometer (620 nm) connected to an integrating recorder is used to determine the amount of material exiting from the column.

Latex beads, 0.1 μm in diameter, were used as model macromolecules. These beads were suspended in a 10 or 20% $MnCl_2$ solution at a concentration of 0.01 wt%. A succession of eight, 70 μL samples were injected into the column and the concentration traces were monitored by the integrating recorder. The flow rate of the bulk solution was set at either 0.1 or 0.5 mL/min which



1- MAGNET

2- COLUMN OF PACKED SPHERES

3- PUMP AND SAMPLE INLET

4- ABSORBANCE CELL

5- INTEGRATING RECORDER

Fig. 3. Schematic diagram of experimental HGMS system.

yielded average column residence times of approximately 20 and 4 min, respectively. Concentration versus time peaks were continuously collected and integrated in order to determine whether or not latex beads were being trapped in the column.

Table I contains the mass balance data from the eight successive injections. Runs 1 and 2 were carried out with water as the carrier solution in order to set a reference for the mass balance. These two runs showed that each injection results in 9.7 units of beads being put into the column. In these two runs, all of the beads flowed through the column even when the magnet was on because the water did not provide adequate background magnetic susceptibility for capture.

Runs 3, 4, and 5 were carried out to see if bead behavior could be affected by using a 10% $MnCl_2$ solution. The tabular data indicate that a significant fraction of the beads remained in the column even at the high flow rate (0.5 mL/min) and that total bead capture can be accomplished (Run 4). The behavior observed in these three runs appears to be inconclusive regarding elucidation of the particle capture mechanism. One possible explanation

Table I: Results for HGMS capture of 0.1 μm diam latex beads.

Run	Flow (mL/min	Recovered Material (bead units)	Comments Carrier	Magnetic Field (tesla)
1	0.5	9.70	H_2O	2
2	0.1	9.68	H_2O	6
3	0.5	5.70	10% $MnCl_2$	6
4	0.5	0.00	10% $MnCl_2$	6
5	0.1	2.19	10% $MnCl_2$	6
6	0.5	8.75	20% $MnCl_2$	6
7	0.1	27.91	20% $MnCl_2$	0
8	0.1	9.82	20% $MnCl_2$	0
Total injections (8)		77.6 in	95% recovery	
Total recovered		73.8 out		

is that after a small amount of magnetic field capture occurs (run 3) combined electrostatic (agglomeration) and magnetic interactions serve to induce further particle capture (run 4) until the column becomes fully loaded and displays breakthrough (run 5).

Runs 6, 7, and 8 were carried out using the 20% $MnCl_2$ solution. The data for Run 6 seem to indicate that the column is fully loaded with beads and subsequently breakthrough occurs. The liquid helium supply for the superconducting magnet was depleted 11 min. into Run 7. Elution of beads proceeded to occur immediately after the magnet was deactivated. Hence, the trace for this run represents unloading of the column by turning off the magnetic field. In run 7, 88% of the beads accumulated in the bed from runs 3-6 are accounted for by integration of the effluent stream signal. This apparent deficiency can be explained by the nonlinearity of the absorbance signal at the higher bead concentration vs absorbance curve begins to flatten out; thereby, somewhat underestimating the true concentration of beads that would be present. Corrective calculations were not undertaken in these preliminary runs.

Run 8 was performed to insure that all of the material was washed out of the column and that using the salt carrier solutions did not induce particle capture.

The overall mass balance for the eight runs was extremely good. The integrated peaks accounted for 95% of the material injected into the column. This result represents continuously

accounting for material injected into the column for a time period > 5.0 h.

CONCLUSIONS AND RECOMMENDATIONS

These initial experiments indicate that HGMS techniques may have applications in the separation of macromolecules. It was demonstrated that 0.1 100 μm diam diamagnetic beads can be captured in a high-gradient magnetic field if a suitable paramagnetic background solution is used. The captured beads can be released from the column by simply turning off the magnetic field.

Most macromolecules are much smaller than the latex beads used in these experiments. Typical diameters are < 10 nm; therefore, it is not certain that the magnetic field gradient mechanisms described in this paper can affect the molecules in solution. However, in the case of large biologically derived species (e.g. proteins) there often is an electric charge associated with the molecule; hence, it is possible that this electric charge will interact with the magnetic field through the Lorenz force and induce particle capture even though the molecular dimensions are quite small.

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