Progress in Magnetic Separation Technology for Processing Large Quantities of Dilute Suspensions

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Abstract

Magnetic separation using permanent magnets and/or electromagnets has been increasingly used for purification and recycling of suspended solids and water since the mid 1970’s. The objectives behind the development of such magnetic separation systems include purification of kaolin clay in the paper-coating industry, wastewater recycling in the steel industry, and recycling of glass-grinding sludge in the CRT-polishing industry. Due to direct, selective, high-intensity magnetic forces on particles to be separated, the filtering speed of High Gradient Magnetic Separation (HGMS) systems, developed in the 1970’s, is now 15–300 times faster than conventional filtration systems. Consequently, magnetic separation is now being applied to a broad class of weak paramagnetic materials, down to submicron particle size. Such HGMS systems have been used for large-scale manipulation of colloidal materials (i.e., up to 50 kg/hr for drainage purification and reuse systems in steel mills). In the 1980’s, large superconducting magnets were adopted for field coils of HGMS systems used for kaolin clay purification. After development in the 1990’s of less expensive, liquid-helium-free superconducting magnets, national research projects were initiated in Japan, Korea and China to expand their application. A particularly pressing application is the environmental remediation of soil polluted with radioactive particles in Japan.

Key Words: Magnetic force, High gradient magnetic separation, Drainage purification, Reuse of waste fine particles

I. INTRODUCTION

At the plenary session of the fifth international cryogenic engineering conference held in Kyoto in 1974, Henry H. Kolm [1] from MIT stated that, “Superconductivity represents a breakthrough comparable to the invention of the wheel: it offers frictionless transport of electricity in quantities heretofore inconceivable, and lossless generation of magnetic fields of intensities and sizes beyond a previous hope of economic feasibility.” and that “Superconductivity makes high gradient magnetic separation economically feasible on a large scale, and this constitutes a very significant breakthrough in that it provides a complete new technique for the large-scale manipulation of colloidal materials. It seems likely at present that magnetic separation will provide the first, the largest, and the most vitally important application of superconductivity.”

In 1995, economically and operationally favorable liquid-helium-free superconducting magnets with excellent operability were developed in Japan by adopting high critical temperature superconductors discovered in 1986. Consequently, “the large-scale manipulation of colloidal materials by using a superconducting magnet” is no longer a prediction, but a realistic target. This introductory paper discusses the progress and present status of such application.

II. FEATURES OF MAGNETIC SEPARATION AND HGMS

In magnetic separation, magnetic forces act directly on fine particles suspended in dispersion medium (water, air, oil, organic solvents, etc.). The force vanishes when the externally applied magnetic field is turned off, so that the system can continuously be regenerated by cycling it on and off. Unlike conventional filtration that uses the blocking-type filtration method, high gradient magnetic separation (HGMS) systems do not discharge any secondary waste. Furthermore, because HGMS systems involve magnetic forces that are 1,000 to 10,000 times higher than those in conventional magnetic separation techniques, HGMS systems can also be used to rapidly separate large quantities of dilute suspensions. For example, in the geothermal water purification system at the Kakkonda power plant in Japan, the flow velocity in filtration in the HGMS system to remove arsenic is 5.1 m/min (10 L/min), and the arsenic concentrations before and after filtration are 0.05–0.35 mg/L and less than 0.07 mg/L, respectively, when only 80 mg/L of Fe(III) hydroxide is used as an additive to co-precipitate with arsenic [2].

Magnetic forces acting on a particle do not require a uniform magnetic field, but rather a non-uniform, or a gradient magnetic field. Such a magnetic field exists near the magnetic pole of permanent magnets arranged in series with alternating orientation of the magnetic poles. In an HGMS system, the field gradient reaches 1.6×10¹⁰ A/m² (20,000 T/m), and the magnetic force is enhanced by a factor of 1,000–10,000 [3] in comparison to that generated with a permanent
magnet. This high gradient magnetic field can be created by applying a magnetic field to a grid of 100μm-diameter ferromagnetic thin wires.

According to precise calculation [4], the magnetic force acting on a particle in an HGMS system depends not only on the magnetic field gradient but also on the (1) particle volume, (2) relative magnetization between the dispersoid and the dispersion medium, and (3) strength of the applied magnetic field. Thus, an effective technique to increase the magnetic force is to use a (1) cohesive material such as poly aluminum chloride (PAC), (2) a magnetic scavenger such as surface-activated magnetite, and (3) a superconducting magnet capable of generating a higher magnetic field than that by a permanent magnet or an electromagnet.

### III. PARTICLE SIZE LIMIT OF HGMS

Experiments [5] and the generalized theory [6] reveal that HGMS systems have a particle-size limit based on the force balance between the magnetic and diffusion forces. The diffusion force is caused by Brownian motion, which dominates the kinematics of suspended particles as the particle size decreases. In contrast, the finer the particle, the weaker the balance between the magnetic and diffusion forces. The diffusion force is to use a (1) cohesive material such as poly aluminum chloride (PAC), (2) a magnetic scavenger such as surface-activated magnetite, and (3) a superconducting magnet capable of generating a higher magnetic field than that by a permanent magnet or an electromagnet.

To estimate this particle size limit d for HGMS systems, Geber et al. [6] derived the criterion $d \leq \frac{6\alpha kT}{\mu_0 H_0} \left( \frac{9(\chi_p - \chi_t)}{(3 + \chi_p)(3 + \chi_t)} (\pm r_a^{-3} + \frac{M^\alpha}{2H_0 r_a^{-3}}) \right)$. We adopted Gerber’s criterion at $r_a=5$ (x/a=5 or y/a=5) although the strength of a magnetic force $F_p$ that attracts a paramagnetic particle onto the surface of the ferromagnetic wire is maximum at the surface ($r_a=1$) marked “A” in Fig. 1, and the force $F_d$ that attracts a diamagnetic particle is maximum at “B.”

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Fig. 2 shows the numerically calculated results from the values in Table I where the ferromagnetic wire is placed in water, and $H_0$ is either weak (2.0/μ₀ A/m) or strong (10.0/μ₀ A/m). We adopted Gerber’s criterion at $r_a=5$ (x/a=5 or y/a=5) although the strength of a magnetic force $F_p$ that attracts a paramagnetic particle onto the surface of the ferromagnetic wire is maximum at the surface ($r_a=1$) marked “A” in Fig. 1, and the force $F_d$ that attracts a diamagnetic particle is maximum at “B.”

![Fig. 1 Magnetic force near the ferromagnetic wire surface.](image)

![Fig. 2 Particle-size limit of HGMS.](image)
The calculation results reveal the following: (1) the larger the absolute value of particle susceptibility, the smaller the diameter of a particle captured on the ferromagnetic wire, (2) the infinite value of \( d \) at the water susceptibility means that particles with the same magnetic susceptibility as the dispersion medium cannot be captured, and (3) increase in \( H_0 \) enhances the magnetic force between particles and the ferromagnetic wire, thus enabling the capture of smaller particles, and (4) because the term \( 9(\chi_p - \chi_0)/\{3+\chi_p\} (3+\chi_0) \) in Eq. (1) is larger, the magnetic force on a diamagnetic particle is larger than that on a paramagnetic particle where \( |\chi_p| > 5 \times 10^{-2} \), and hence \( d \) of diamagnetic particles is smaller than that of paramagnetic particles. For example, the calculated \( d \) is 45.1 nm for diamagnetic particles and 53.6 nm for paramagnetic particles at \( |\chi_p|=1.0 \) and \( H_0=10^7 \times \).

**IV. CHRONOLOGICAL PHASES OF R&D MAGNETIC SEPARATION ACTIVITIES**

In terms of magnetic properties and particle size of the target particle to be separated, the historical development of magnetic separation can be classified into three chronological phases as shown in Fig. 3.

**A. Phase I: Ferromagnetic and Large Particles Mainly in Mines**

Phase I is the period up to 1970. Magnetic separation had been used for many years prior to collect ferromagnetic materials such as steel chips accidentally mixed into food or magnetic iron ore. The application was however limited to sorting iron or iron-containing ore such as titanium or tin. The sorting devices equipped with weak permanent magnets could exert a magnetic force on large, ferromagnetic particles whose diameter was down to around several millimeters. Rotating–drum magnetic separators have mainly been used in mines. As an example, the device called an open gradient magnetic separation (OGMS) shown in Fig. 4 removed ferromagnetic impurities and enriched titanium in mines in Brazil. The picture was taken in 1984, and it is uncertain whether this OGMS is still being used.

**B. Phase II: Ferromagnetic and Small Particles in Mines and Industry**

Phase II starts around 1970. The turning point was the invention of high gradient magnetic separation (HGMS), and its application for refining kaolin clay in the paper industry in Georgia in the USA [7]. In HGMS, the magnetic force on particles is enhanced by a factor of 1,000–10,000 in comparison with OGMS. By using HGMS, weakly magnetized particles with the same magnetic susceptibility as the dispersion medium cannot be captured, and (3) increase in \( H_0 \) enhances the magnetic force between particles and the ferromagnetic wire, thus enabling the capture of smaller particles, and (4) because the term \( 9(\chi_p - \chi_0)/\{3+\chi_p\} (3+\chi_0) \) in Eq. (1) is larger, the magnetic force on a diamagnetic particle is larger than that on a paramagnetic particle where \( |\chi_p| > 5 \times 10^{-2} \), and hence \( d \) of diamagnetic particles is smaller than that of paramagnetic particles. For example, the calculated \( d \) is 45.1 nm for diamagnetic particles and 53.6 nm for paramagnetic particles at \( |\chi_p|=1.0 \) and \( H_0=10^7 \times \).
titanium oxide particles down to tens of microns in diameter can be magnetically separated in practical systems. HGMS has also been used as a drainage recycling system for treating rolling-mill waste water in the steel industry [8, 9], and such systems of various sizes have been fabricated, with the largest canister’s inner bore of 3 m. The flow velocity in filtration is 600 m/h and 15 times faster than that of a conventional filter. Since its inception, about 100 HGMS systems have been in operation in the steel industry. A glass-grinding sludge recycle system [10] shown in Fig. 3 is also in operation in Japan.

In 1986, R&D in magnetic separation substituted superconducting magnets for electromagnets in kaolin refinement processes in the USA. [11, 12] The design concept was simply retro-fitting to save electricity consumption for generating a magnetic field on the HGMS filter. Despite being liquid-helium cooled, with its generated magnetic field of 2.0 T and energy storage of 3.53 MJ, compared with previous HGMS systems, this “superconductorized system” resulted in a 58% reduction in equipment weight, a 66% reduction in volume, and a 95% reduction in power consumption. Moreover, the field coil charging/discharging time was shortened from several minutes to 1 min or less, and operational efficiency was also substantially improved.

Even this striking example however does not fully show the merits of superconductivity. The reason is that in this application, the charging/discharging cycling of superconducting magnet occurred every 15 min, thus burdening the superconducting magnet and its cryogenic system with heavy ac power losses. Eventually only three of these systems with superconducting magnets were produced.

C. Phase III: Weak Magnetic and Fine Particles in Water Processing

Phase III starts in the late 1980s. R&D focused mainly on applying superconducting magnets to generate a sufficient magnetic field to process large amounts of water containing weakly magnetized fine particles at high rate. One example is the kaolin refinement system developed in the UK [13, 14] that was completed in the early 1990s in which three main improvements were made to superconducting magnets: (1) Operation in a persistent current mode without an iron yoke, which reduced burden on the cooling system; (2) a magnetic field of 5 or 6 T, which increased the processing speed; and (3) reciprocating movement of two canisters packed with HGMS filter, which shortened the “dead” time for the cleaning of the HGMS filters by rinsing without being subjected to a magnetic field. When one HGMS filter was separating particles, the other was being cleaned, thereby improving the operation efficiency. The resulting separation capacity increased by 24 times compared to non-reciprocating devices. As of February, 1999, there were about 30 such systems in operation in the world [15].

Another example is the river water purification system that uses a continuously rotating disk-type HGMS filter. The filter is mounted on the cryostat and is installed outwardly from the center axis of a solenoidal superconducting magnet bore, and the magnet is energized using a permanent current switch, which consumes less electric power. The test plant with a processing capacity of 500 ton/day operated for 21 months at Jiganebori River in Kashiwa city, in Chiba prefecture, Japan, and showed good performance, evidenced by phosphorous and suspended solid removal rates of more than 85% and less than 5 mg/L, respectively, at a water flow speed of 0.1 m/s [16]. Other applications of this type of HGMS system are the removal and recovery of phosphorus and organic dyes from wastewater, recovery of nickel from...
electroless-plating waste fluid, and wastewater processing using highly concentrated magnetic activated sludge.

The 1995 invention of economically and operationally favorable cryocooler-cooled superconducting magnets followed the 1986 discovery of high critical temperature cuprate superconductors, and thus enabled applications for handling large amounts of water contaminated with thinly suspended solids (on the order of several ppm). Applications include removal of biogenic and anthropogenic pollutants from aquatic systems and industrial waste from streams. From 2000 to 2004, R & D on arsenic removal from geothermal water was undertaken [17-20]. In 2003, a wastewater purification system of a paper factory started operation in Japan [21]. As shown in Fig.4, numerous symposiums on the R&D have been held in Japan. In June 2013, the symposium on removal of radioactive particles from the soil by use of superconducting magnetic separation technology was held in Fukushima, Japan, and the strong possibility of using clay as a scavenger was reported.

In addition, extensive R&D has been done on superconducting magnetic separation systems. In Germany, OGMS-type devices have been manufactured and tested. One sample that was tested in a coal mine is a rotary drum magnetic separator using a multi-pole superconducting magnet soaked in liquid helium with a maximum generation field of 3.0 T [22]. Another example is a device equipped with a liquid-helium-free 5 T superconducting magnet directly cooled by a cryocooler [23], and tested for removal of heavy metals from laboratory discharge effluents. These two devices have no magnetic wire filters, and the magnetic forces are due to the magnetic field gradient simply generated with superconducting magnets alone. In China and Korea, R&D has been done in the fields of kaolin purification, hazardous material removal from coal, and recovery of rare earth elements from waste sludge.

V. CONCLUDING REMARKS

Magnetic separation is a primary application field for superconducting magnets, and also a promising technology for efficient processing of large quantities of dilute suspensions. The full-scale application of magnetic separation processes will significantly contribute to the preservation of the global environment. A particularly pressing application is the environmental remediation of soil polluted with radioactive particles in Japan.

REFERENCES