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Development of a Magnetic Filter System Using Permanent Magnets for Separating Radioactive Corrosion Products from Nuclear Power Plants

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ABSTRACT

Radioactive corrosion products generated through the neutron activation of general corrosion products at nuclear power plants are the major source of occupational radiation exposure. Generally, radioactive corrosion products exit in soluble and insoluble forms, and are removed by ion exchangers and purification filters. Most of the insoluble radioactive corrosion products have the characteristic of showing strong ferrimagnetism. With the development and production of permanent magnets (rare earth magnets) capable of generating a much stronger magnetic field than

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conventional permanent magnets, a new type of magnetic filter that can efficiently separate radioactive corrosion products and eventually reduce radiation exposure to personnel at nuclear power plants is proposed and developed. Magnetic separation by using permanent magnets has certain advantages, such as high flow purification, high pressure and temperature operation, and energy saving. The magnetic separator consists of an inner magnet assembly and an outer magnet assembly, a coolant channel, and a container surrounding the outer magnet assembly. The rotation of the inner and outer permanent magnet assemblies by a driving motor system produces moving and alternating magnetic fields in the coolant channel, which is located between the two magnet assemblies. The particulate in the radioactive corrosion products is separated from the coolant by the alternating magnetic fields as a result of the shifting arrangement of the permanent magnets. This study describes the preliminary experimental results of using different particle sizes and various materials at the different flow rates and rotation velocities of the magnet assemblies. This new magnetic filter performs well in filtering magnetite, cobalt ferrite, and nickel ferrite from aqueous coolant simulation.

Key Words: Radioactive corrosion products; Magnetic filter; Alternating magnetic field; Separation.

INTRODUCTION

In general, radioactive corrosion products are called Chalk River unidentified deposits (crud); they have been one of the unsolved problems of the nuclear power industry since first discovered at Chalk River, Canada. The reduction and removal of crud from reactor coolant systems is very important because crud is the major source of occupational radiation in nuclear power plants.

Major radioactive corrosion products such as ^{58}Co and ^{60}Co are known to contribute to more than 70% of occupational radiation exposure. The International Commission on Radiological Protection Publication 60 on radiation protection requires a much stricter reduction of the occupational radiation exposure that is, occupational: 5 rem $\Rightarrow$ 2 rem; general public: 0.5 rem $\Rightarrow$ 0.1 rem. To reduce exposure to radiation, the requirements for reducing the buildup of crud radioactivity and for increasing the removal rate of crud in the primary coolant system of nuclear power plants are becoming more demanding. There are several ways to reduce the radiation levels around the primary water system, for example, by improving the coolant purification system, by operating at high pH, by adopting materials with low levels of cobalt in the primary coolant system, and by decontaminating the primary system more frequently.[1] By the mid-1980s, although the technology based on electromagnetic fields for particulate removal in crud had been studied widely, the studies could not continue due to problems, such as the backflushing of electromagnetic filters. In recent years, the manufacturing technology of permanent magnets, especially rare earth magnets, has developed greatly: the new types of permanent magnets can inexpensively generate a stronger magnetic field than a conventional magnet. Thus, a new magnetic filter that uses these magnets has been proposed.

RADIOACTIVE CORROSION PRODUCTS

The materials that normally come into contact with the coolant streams of thermal power systems are metal alloys composed mainly of elements such as iron, nickel, copper, chromium, cobalt, aluminum, zinc, titanium, zirconium, carbon, and manganese. All these elements react with water and dissolved oxygen to form oxides (mixture of metal oxides).[2] The main crystal structure of these oxides is the spinel type similar to the structure of Fe^{2+} and Fe^{3+} in Fe_3O_4, with a partial substitution of Fe^{2+} with Ni^{2+} and Co^{2+}, and Fe^{3+} with Cr^{3+} and Co^{3+}. They are transported by the coolant stream and then deposited throughout the system, inducing adverse effects on the operation of the power plant.[3]

Initially, magnetite (iron ferrite: $FeOFe_2O_3 \Rightarrow Fe_3O_4$) was suspected as the main corrosion product. Further research suggested that corrosion products more likely consisted of nickel (cobalt)–ferrites ($Ni_xFe_{3-x}O_4$ or $Co_yNi_xFe_{3-x-y}O_4$). Both magnetite and nickel ferrite are included ferrite series, which has strong magnetism. A comparison of the types of magnetism for these and other important corrosion products is presented in Table 1.

Table 1. Magnetic properties of certain metals and metal oxides.

Ferromagnetic	Ferrimagnetic	Paramagnetic	Diamagnetic
ANSI 52100 chrome steel	Magnetite (Fe_3O_4)	Ferrous oxide (FeO)	Copper (Cu)
	Cobalt ferrite ($CoFe_2O_4$)	Cobaltous oxide (CoO)	Cuprous oxide (Cu_2O)
	Maghemite (γ-Fe_2O_3)	Chromic oxide (Cr_2O_3)	Zinc oxide (ZnO)
	Nickel ferrite ($NiFe_2O_4$)	Nickelous oxide (NiO)	
	Copper ferrite ($CuFe_2O_4$)	Cupric oxide (CuO)	

The primary long-term source of radiation in pressurized water reactors is ^{60}Co, which is formed by the neutron capture of ^{59}Co. The radioactive product ^{58}Co, formed from the reaction between ^{58}Ni and fast neutrons, is another important contributor to out-of-core radiation fields. The major source of nickel in the reactor coolant comes from the corrosion of the steam generator tubing, which is made of Alloy 600. The major sources of cobalt are the corrosion release of cobalt impurities from the Alloy 600 tubing, the mechanical wear of alloys of high cobalt content (used in control rod drive mechanisms, reactor coolant pumps, valves, and so on), and the corrosion release of cobalt from stainless steel piping and vessel internals. Cobalt and nickel are released to the primary coolant through the corrosion process and then deposited on fuel surfaces. The activation products ^{58}Co and ^{60}Co are gradually released to and circulated by the coolant in soluble, insoluble, or colloidal forms. During the power operational period, the crud in the primary coolant generally is removed at the chemical volume control system, which controls the concentration of chemicals, boron and lithium. At the shutdown, the primary coolant water is treated through various chemical processes for crud removal. Ion exchangers mainly remove the soluble form, while physical filters remove particulate. However, physical filters have the disadvantage of being unable to operate continuously because of the pressure drop caused by filter clogging. Furthermore, residual particulate may have adverse effects on the performance of ion exchangers, particularly through clogging, which shortens the useful life span of ion-exchange resins.

MAGNETIC FILTER USING PERMANENT MAGNETS

Most crud elements (nickel ferrite, magnetite, and so on) are strongly ferrimagnetic. A magnetic filter that uses permanent magnets can be applied efficiently under extreme conditions such as high temperature and high pressure.[4] The system we have developed comprises two main parts: a separator and a driving motor. The separator consists of an inner assembly and an outer magnet assembly, a fluid channel, and a container surrounding the outer magnet assembly. The fluid channel is located between the inner and outer permanent magnet assemblies. As corrosion products in the fluid pass through the channel between the magnet assemblies, they move toward nearby magnets because of the strong magnetic field. They then move toward the rotating direction of permanent magnets, which are rotated by a driving motor connected to the separator.

The rotation of the permanent magnet assembly and the shifted arrangement (S–N pole) of the permanent magnets generate the alternating magnetic field. The crud elements in the magnetic field, such as magnetite and nickel ferrite, are then magnetized. The crud that moves toward the rotating direction of the magnet assemblies is separated from the coolant at the boundary wall of the vessel; it is then collected in the corner of the fluid vessel near the outlet of the crud, as shown in Fig. 1. The effectiveness of the magnetic filter in separating particles in a fluid stream strongly depends on the magnitude of the magnetic force. In general, the competing forces are due to the hydrodynamic drag and inertial effects on the particle.

In a cross-sectional top view of the separator, Fig. 2 shows the inner and outer magnet assemblies and the fluid channel. To maximize the magnetic field, magnetic circuits are constructed with irons that strengthen the magnetic field between the inner and outer magnet assemblies. Each magnet faces a opposite polar magnet, located between the inner and the outer assembly, the fluid channel is influenced by the strong magnetic field produced by the coupled magnets.

Figure 1 shows a schematic drawing of the fluid channel. The movements of the crud particles in the fluid channel are shown in Fig. 3, which is a linearized version of Fig. 2. The crud in the coolant is captured by the magnetic field and moves to the rotating direction of the two magnet assemblies. Arriving at the boundary wall, the crud accumulates at the bottom corner of the fluid channel. The crud can be separated with an appropriate batch operation.

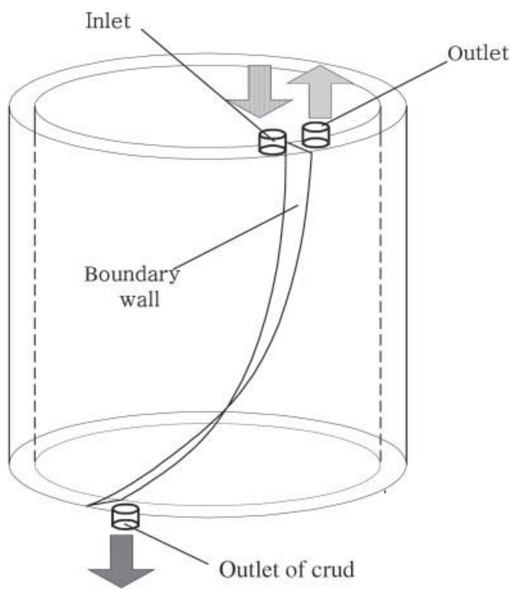

Figure 1. Schematic drawing of the fluid channel.

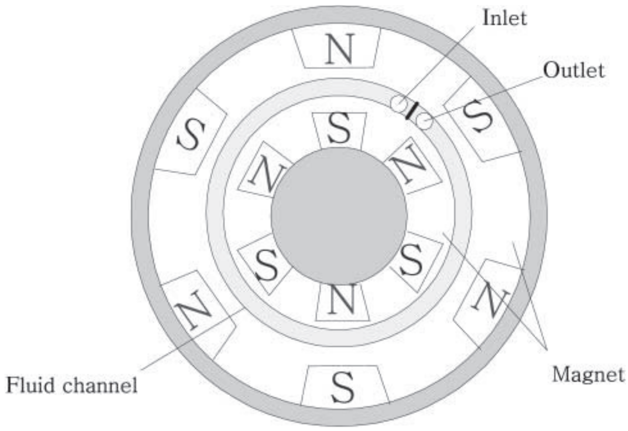

Figure 2. Cross-section of the separator (top view).

Magnet Assembly

The inner magnet assembly consists of six linear arrays of magnets (about 6000 Gauss, Nd) placed on an iron rod. Each array consists of 10 magnets whose magnetic poles are identically aligned. To improve separation, the arrays of magnets incline 15 degrees relative to the vertical direction. The iron rod is connected to a driving motor that rotates the inner magnet assembly. As shown in Fig. 2, the outer magnet assembly also contains six arrays, facing the arrays of the inner magnet assembly but in the opposite magnetic pole. Figure 4 shows the assembling sequence of the magnetic separator.

Fluid Channel

The fluid channel is located between the outer magnet assembly and the inner magnet assembly. It is made of SS-316, a kind of stainless steel, so as not to affect the magnetic field generated by the magnet assemblies. The magnetic field, throughout the fluid channel is about 3000 Gauss, governs the movement of the crud in the aqueous suspension.

Driving Motor

The driving motor rotates the magnet assemblies to separate corrosion products. The motor is connected to the inner and outer magnet assemblies and a speed controller is used to set the rotation velocity of the magnet assemblies.

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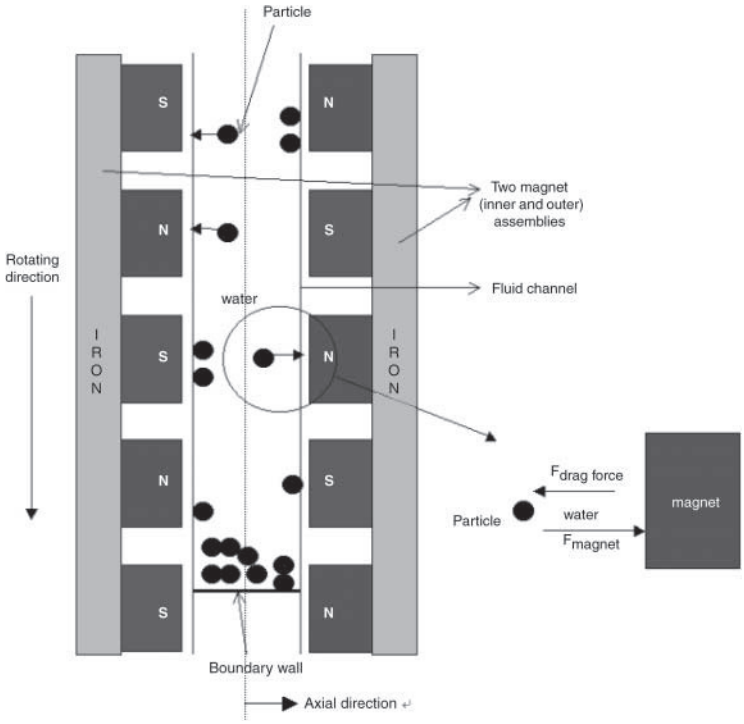


Figure 3. Schematic representation of particle motion and deposition in the separator.

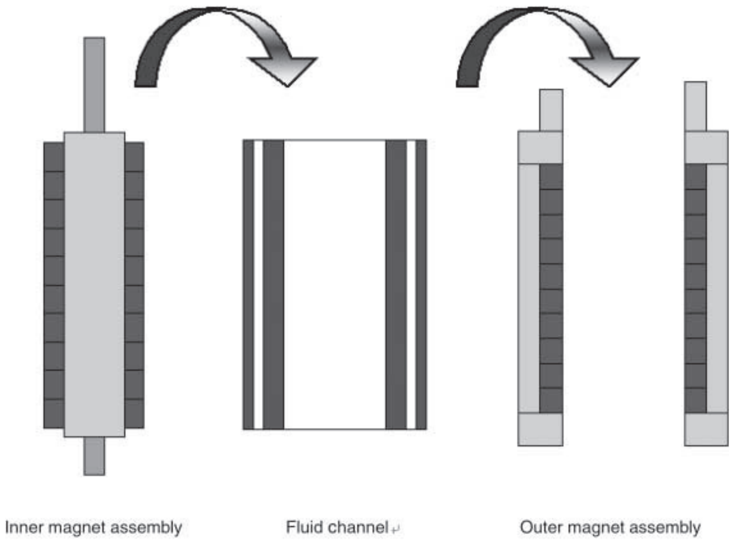
THEORETICAL APPROACH

Analysis of the movement of crud (magnetic particles) under a moving and alternating magnetic field is presented. First, when moving under a magnetic field, a particle experiences magnetic and viscous drag forces.[5–8] The equation showing movement of crud is considered a local area (in the circle of the upper Fig. 3).

The magnetic force is assumed to act only along one direction and can be described by

$$F_{\mathrm{m}} = V_0 \mu_0 \chi H \frac{dH}{dx} \tag{1}$$

where F_{m} is the magnetic force (N); V_0, the particle volume (m^3); μ_0, the magnetic permeability of free space ($\mathrm{T \cdot m/A}$); χ, the volumetric susceptibility; H, the magnetic field (A/m); and x, the axial distance (m).



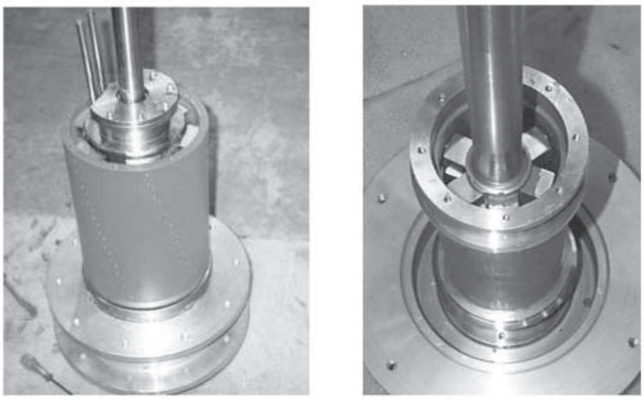


Figure 4. Separator assembly.

The viscous drag force experienced by the particle is,[9]

$$F_D = \frac{\rho V^2 A_p C_D}{2}, \quad C_D = \frac{24}{Re} \tag{2}$$

where F_D is the drag force (N); ρ, the density (kg/m^3); V, the particle velocity (m/sec); A_p, the projected particle area (m^2); C_D, the drag coefficient; and Re, Reynolds number.

The motion of the crud particles is given by the following equation:

$$m\frac{dV}{dt} = -F_{\mathrm{D}} + F_{\mathrm{m}} \tag{3}$$

where m is the particle mass (kg), and t is the time (sec).

Substituting Eqs. (1–3), into Eq. (4),

$$\frac{dV}{dt} + \frac{9\mu}{2a^2\rho_{\mathrm{p}}}V = \frac{\mu_0\chi H}{2\rho_{\mathrm{p}}}\frac{dH}{dx} \tag{4}$$

where μ is the fluid viscosity (kg/m·sec); a, the particle radius (m); and ρ_{p}, the particle density (kg/m^3).

If the inertial forces can be neglected, i.e., the particle velocity remains constant or $dV/dT = 0$, the particle velocity can be expressed as follows:

$$V = \frac{a^2\mu_0\chi H}{9\mu}\frac{dH}{dx} \tag{5}$$

The velocity of the particle varies the magnetic field strength, particle size, and viscosity. The other parameters of Eq. (5) are terms related to the magnetic properties of particle.

The filter coefficient related to separation factors (η) can be defined as the ratio of the water velocity to the particle velocity if the crud, arrived at the vessel walls (not the boundary wall) by attractive force of magnet, is separated. For instance, the filter coefficient is equal to 1 when V_{crud} is equal to V_{flow}. This means all particles move to the vessel wall by magnetic force before (or until) water flows out of some area. The water velocity, V_{flow} is

$$V_{\mathrm{flow}} = \frac{Q}{A} \tag{6}$$

where Q is the volumetric flow rate (m^3/sec); and A, the cross sectional area of the fluid channel (m^2).

The filter coefficient (ε) can be expressed as

$$\eta \propto \varepsilon = \frac{V_{\mathrm{crud}}}{V_{\mathrm{flow}}} = \frac{Aa^2\mu_0\chi H}{9\mu Q}\frac{dH}{dx} \tag{7}$$

EXPERIMENT

A schematic drawing of the experimental equipment is shown in Fig. 4. Water flows from the inlet tank to the outlet tank through the pump, the separator, and the flow meter. The driving motor, which is equipped with a

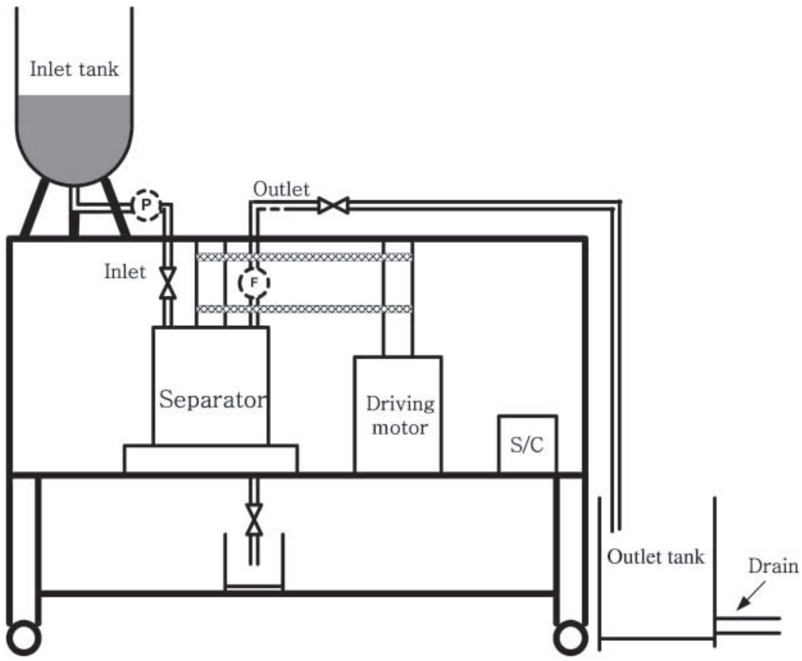


Figure 5. Schematic drawing of the magnetic filtering system.

controller to adjust the rotating velocity, rotates the magnet assembly in the separator. Details of the system components are given in Table 2. All experimental runs were conducted at room temperature and atmospheric pressure because the experimental system, which is at a preliminary stage, is not of the closed-loop type. Figure 5 shows a schematic drawing of the magnetic filter system.

Magnetite (Fe_3O_4), cobalt ferrite ($CoFe_2O_4$), and nickel ferrite ($NiFe_2O_4$) particles were used as crud simulants. Hematite (α-Fe_2O_3) particles with an antiferromagnetic property also were used to prepare the suspensions. The magnetite and hematite particles were supplied by the Aldrich Chemical Company, and the cobalt ferrite and nickel ferrite were obtained from the Kojundo Chemical Laboratory Co. Ltd, Sakado, Saitama, Japan. The relevant properties of their particles are given in Table 3. The particle size distributions are shown in Fig. 6. The size ranges of real crud are known as 1.5–18 μm, and the average size is about 3 μm (Gori unit #1 in Korea). The size is different from the sampling point. The particle size ranges that are used in this study are 2–25 μm, and an average size is about 5–6 μm, which is a little larger than the size of real crud.

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Table 2. Details of the components of the experimental system.

	Material	Size and capacity	Comment
Magnet			
Inner assembly	Nd	25 mm × 30 mm × 20 mm, ~6000 G	60 EA (rare earth)
Outer assembly	Nd	56 mm × 30 mm × 20 mm, ~4500 G	60 EA (rare earth)
Frame	Al	1040 mm × 1530 mm × 600 mm,	
Flow meter		~2.5 gpm	1 EA (rotameter)
Valve	SS 316		4 EA (ball type) control of flow rate
Motor		~120 rpm	
Reservoir	SS 304	60 L	
Pump		15 L/min	1 EA
Pipe	SS 304		
Fluid channel	SS 316	Diameter: 180 mm Height: 300 mm Fluid capacity: 1.5 L	1 EA
Vessel	Al		1 EA

Experimental results were obtained for varying flow rates, the rotating velocities of the magnet assembly, and suspension concentrations. The experimental conditions are summarized in Table 4. The particle counter was used to measure the concentrations of crud simulants in the outlet water. The particle counter used in this research is the instrument used to measure the particle size, which is between 2 and 250 μm (multichannel counter). Since it is difficult to measure a particle whose size is below 2 μm, the experiment was

Table 3. Physical properties of crud simulants used in this study.

	Magnetic property	Type (mean size)	Intensity of magnetization (emu/g)	Density (g/cm^3)
Magnetite (Fe_3O_4)	Ferrimagnetism	Powder (5–6 μm)	92	5.16
Nickel ferrite ($NiFe_2O_4$)	Ferrimagnetism	Powder (3–4 μm)	50	5.38
Cobalt ferrite ($CoFe_2O_4$)	Ferrimagnetism	Powder (2–3 μm)	80	5.29
Hematite (Fe_2O_3)	Antiferromagnetism	Powder (~1 μm)	low	5.24

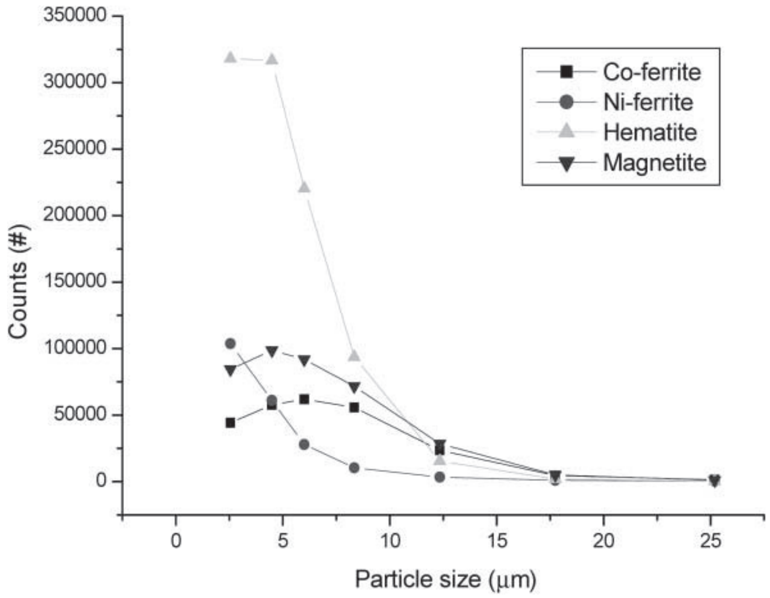

Figure 6. Size distribution of powders used in the study.

carried out with the particle whose average size is 5 and 6 μm and slightly bigger than the real size of crud in the plant, which is 3 μm in average, and we tried to reduce the error with the experiment for the measurable range. It is considered that the influence of the particle whose size is below 2 μm to the separation efficiency is neglected because of the measurable particle size within the limits of 2–25 μm.

Table 4. Experimental conditions.

Environment	Room temperature	
	Atmospheric pressure	
Material	Magnetite (Fe_3O_4)	
	Ni-ferrite ($NiFe_2O_4$)	
	Co-ferrite ($CoFe_2O_4$)	
	Hematite (Fe_2O_3)	
Flow rate	0.5 gpm,[a] 0.9 gpm, 1.3 gpm	Valve
Rotation velocity of magnet assembly	30 rpm, 50 rpm, 70 rpm	Motor controller
Concentration	2 ppm, 10 ppm, 50 ppm	Only magnetite
Particle size	1.3–25 μm	Analysis by particle counter

[a]gpm = gallon per minute (1 gallon = 3.7853 L).

RESULTS AND DISCUSSION

Effect of Water Flow Rate and Magnet Assembly Rotating Velocity

Experimental results with respect to the flow rate and rotating velocity are shown in Figs. 7–11. Figures 7–10 show the separation efficiency of each input material depending on the flow rate and the rotating velocity of the magnet assembly. The results of a comparison of four input materials under the condition of 50 rpm are shown in Fig. 11. All cases except the hematite case show comparatively good separation efficiencies. Differences in the separation efficiency of each material are expected to be caused by differences in magnetic susceptibility (or intensity of magnetization), especially since hematite is an antiferromagnetic material with weak magnetic properties. Basically, the separation efficiency tends to decrease as the flow rate increases, a known characteristic of magnetic filters or electromagnetic filters.

This new type of magnetic filter has a unique parameter: the rotating velocity of the magnet assembly. Figures 6–9 show that changes in the rotating velocity of the magnet assemblies slightly improve the efficiency of the magnetic filter under the condition of the constant flow rate. Thus, the

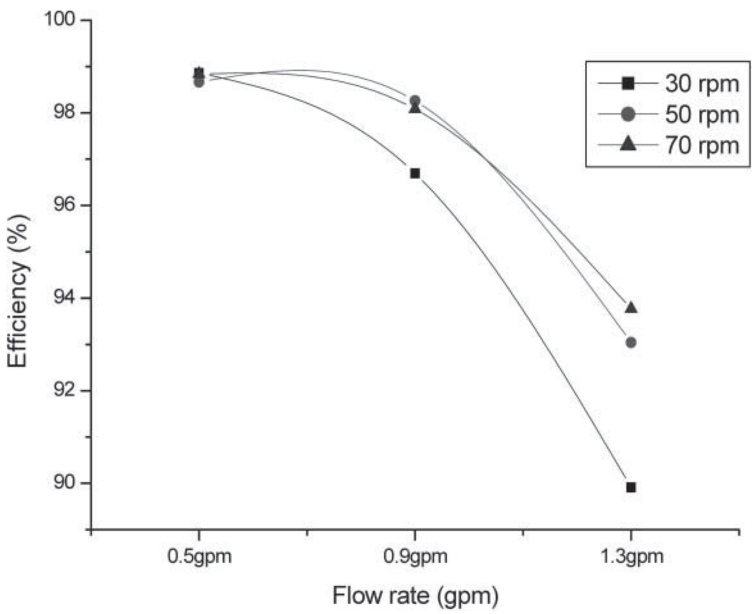

Figure 7. Separation efficiency vs. water flow rate (magnetite suspension).

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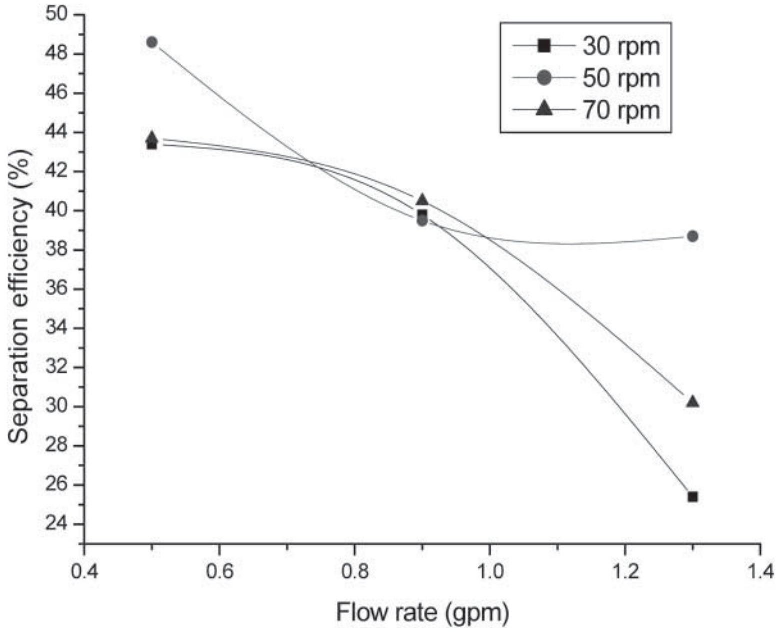

Figure 8. Separation efficiency vs. water flow rate (hematite suspension).

rotating characteristic of the magnet assemblies may be an advantage and could provide useful flexibility of this magnetic filter system. However, the experimental results show that a faster rotating velocity for the magnet assemblies does not always increase the separation efficiency.

Effect of Suspension Concentration

The suspension concentrations have no relation to the theoretical formula [Eq. (7)], because the suspension concentrations are the experimental or operational facts related to sample analysis. In other words, under low concentration conditions, samples are very difficult to analyze accurately, whereas, high concentration conditions differ from real coolant conditions. Conducting the experiments with few errors, while keeping the concentration as low as possible, is very important. The experiments were performed only for the magnetite case. The separation efficiency for the various concentrations of inlet water is shown in Fig. 12. In the case of 50 and 10 ppm, the experimental results show a similar tendency. In the case of 2 ppm, however, the separation efficiency is lower than that of other cases. This phenomenon

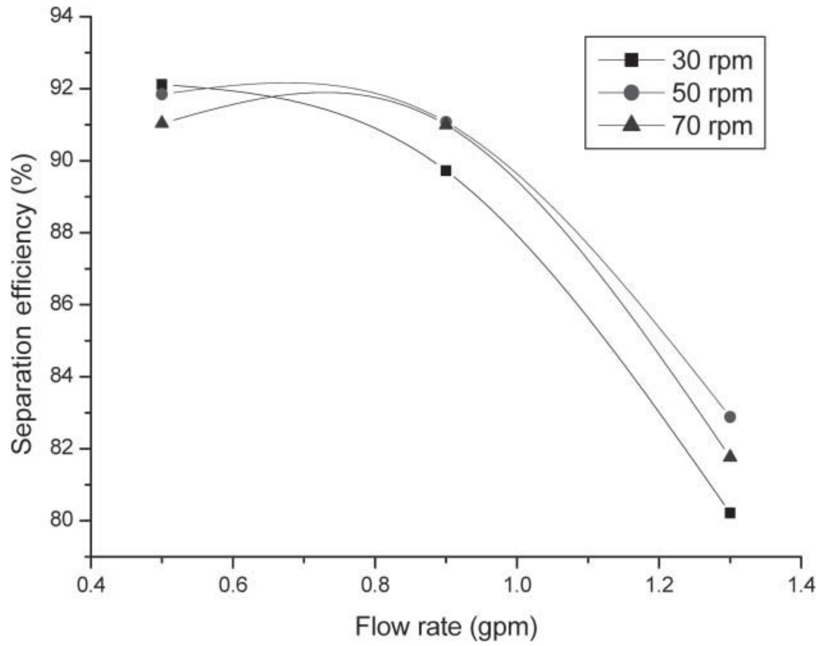

Figure 9. Separation efficiency vs. water flow rate (nickel ferrite suspension).

results from impurities in the bulk water and inside the magnetic filter system, as well as from the experimental error. Due to use of large amounts of water at one experiment, service (city) water is used for the experiments, not distilled water. The concentration of impurities (particle form) contained in bulk water is about ~0.2 ppm. This value can affect the separation efficiency.

Effect of Particle Size

The separation efficiency for the particle size of magnetite and the flow rate is shown in Fig. 13. Figure 14 shows the separation efficiency of the magnetic filter for various particle sizes of four input materials under the condition of 0.9 gpm and 50 rpm. The bigger the particle size, the better the separation efficiency of the magnetic filter system. Above 5 μm, the separation efficiency is more than 90%. Thus, studying for the development of a particle size enlarger based on the magnetic properties of crud is necessary to increase the separation efficiency.

Figure 15 shows the size distribution of magnetite at the input stage, the output stage, and the separated stage where the crud is collected. Small particles

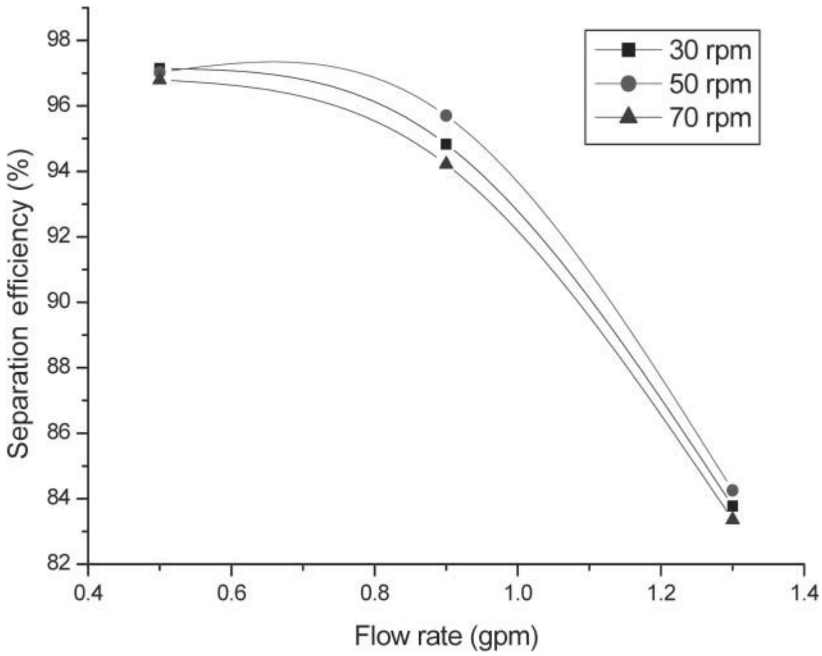

Figure 10. Separation efficiency vs. water flow rate (cobalt ferrite suspension).

are more abundant in the output stage (outlet water) than in the input stage (inlet water), while large particles are more abundant in separated stage. This result implies that "the larger the particle size, the better the separator efficiency."

Discussion

The magnetic filter proposed in this paper has some merits. For example, it uses permanent magnets instead electromagnets, it has a novel design for easily separating the crud, and it performs well.

The efficiency of a magnetic filter tends to improve as the flow rate slows and the particle size increases. For a magnet assembly with a constant rotating velocity, the magnet filter performs better with a slower flow rate. On the other hand, the efficiency rapidly decreases when the flow rate accelerates. The rotating velocity of a magnet assembly has some influence on the separation efficiency. This new type of magnetic filter performs relatively well, compared with other conventional magnetic filters. In the case of a conventional magnetic filter or electromagnetic filter, only the flow rate turned out to be a

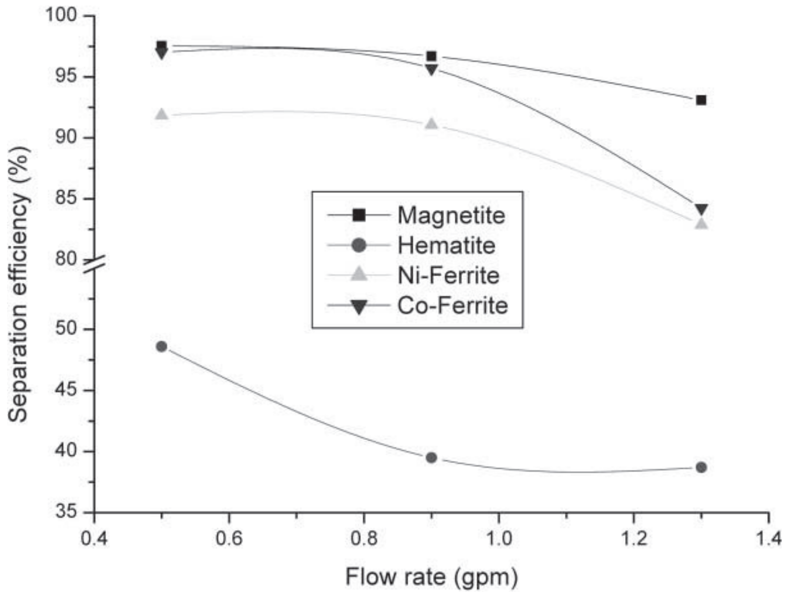

Figure 11. Separation efficiency under the same condition (50 rpm, 10 ppm).

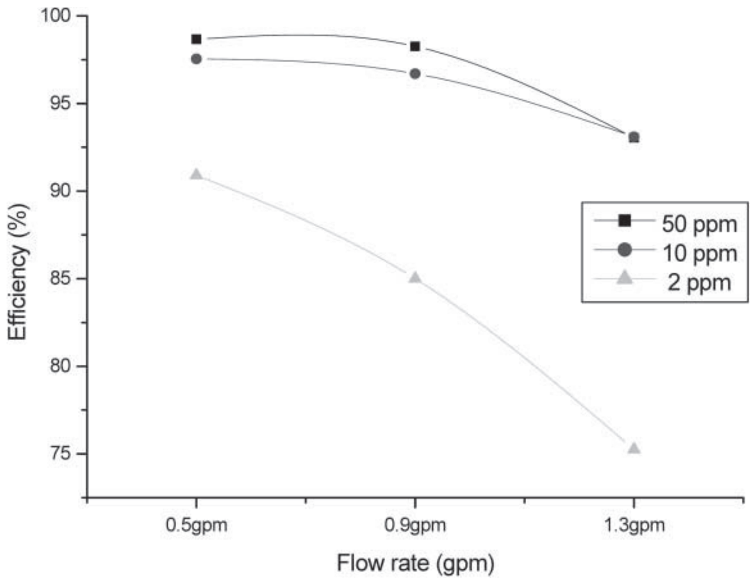

Figure 12. Experiment result for various concentrations of magnetite in the inlet water.

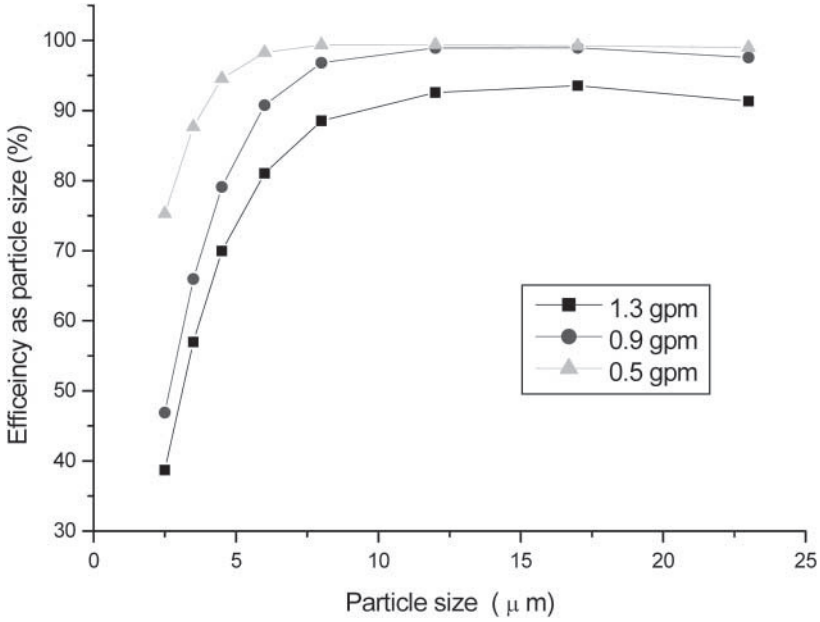

Figure 13. Separation efficiency vs. particle size (magnetite suspension; rotating speed: 50 rpm).

dominant parameter along with the strength of the magnetic field. For the magnetic filter proposed in this study, however, the rotating velocity of magnet assemblies also is the parameter that influences separation efficiency.

Further study is necessary to optimize the rotating velocity of the magnet assembly, especially to overcome the problem related to the flow stream of the water in the fluid channel. This problem is exacerbated by the drag force of the fluid, which is minimized when the moving velocity of the particle is the same as the velocity of the main stream in the fluid channel.

Data acquired through experiments have some variations under the same experimental conditions. The main reasons are the sample analysis system and cleaning of the filter system.

- *Sample analysis system*: There is a possibility that the method using the particle counter for sample analysis makes some errors by the sampling method or the analysis condition. It is regarded that the precision of sample analysis is relatively poor since the atomic absorption spectrophotometer (AAS) and inductively coupled plasma (ICP) cannot be used due to the characteristics that the ferrite metal is little dissolved in a solvent.

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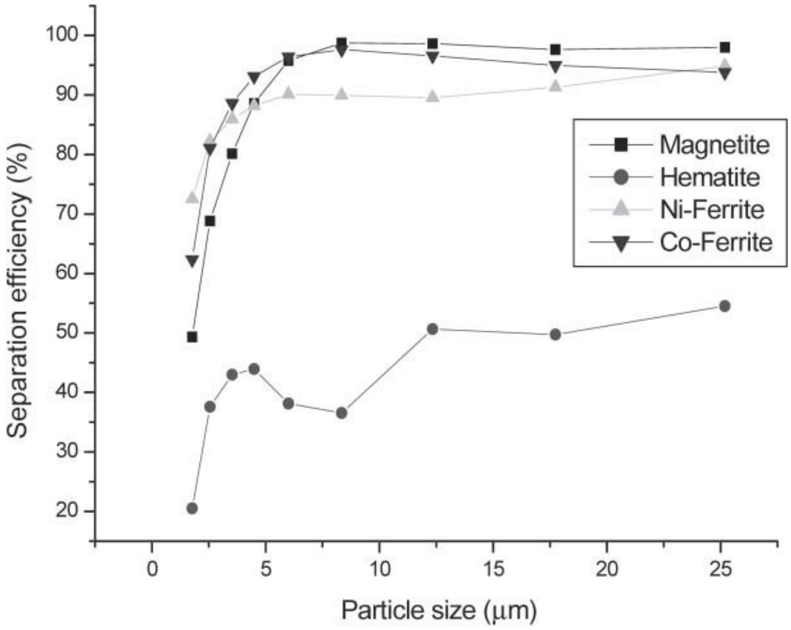

Figure 14. Separation efficiency vs. particle size (four input material; 50 rpm, 0.9 gpm, 10 ppm).

- *Cleaning of filter system*: After each experiment, in general, the filter system is cleaned for the next experiments. It is possible that the impurities are not removed from some component, since the only method used applies a high-speed water flow to remove the remnants and impurities in the system.

To produce more accurate results and to reduce experimental error, better cleaning techniques are required for the magnetic filter system, especially the fluid channel.

CONCLUSION

In this study, a new design of a magnetic filter is proposed, described, and analyzed with preliminary experimental results. The following conclusions can be drawn from the experiments:

- A moving and alternating magnetic filter that uses permanent magnets can provide good operational properties and has potential, and advantages, for removing crud.

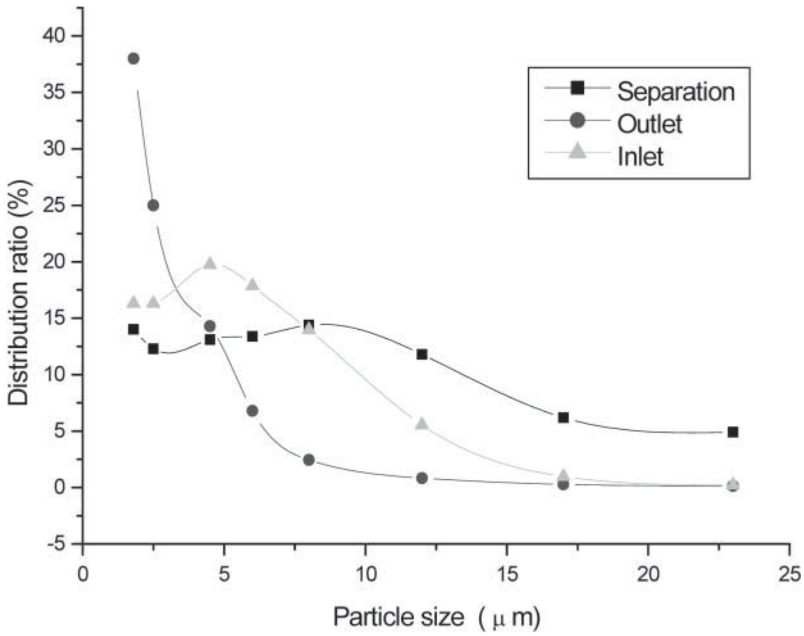

Figure 15. Particle size distribution at each stage.

- The efficiency of a magnetic filter tends to increase with slower flow rates, higher magnetic susceptibility, and bigger particles.
- In addition to the flow rates, the rotating velocity of magnet assemblies could be another parameter for varying the efficiency of the magnetic filter, providing useful flexibility for better operation and performances.
- The magnetic filter could be applied to the chemical volume control system and the steam generator blowdown system in nuclear power plants, as well as to prefilters (or auxiliary filters) for micro- or nanofiltration and membrane or ion exchangers.

The application of the alternating magnetic filter could provide an effective method for the removal and separation of crud. The proposed method of removing crud could also be applied, in general, to other industrial fields when the current and various future experiments are proved to be successful and the performance of the magnetic filter is verified by prospective improvements.

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