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Development of Moving Alternating Magnetic Filter Using Permanent Magnet for Removal of Radioactive Corrosion Product from Nuclear Power Plant

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Abstract

Radioactive Corrosion Products (CRUD) which are generated by the neutron activation of general corrosion products at the nuclear power plant are the major source of occupational radiation exposure. Most of the CRUD has a characteristic of showing strong ferrimagnetisms. Along with the new development and production of permanent magnet (rare earth magnet) which generates much stronger magnetic field than the conventional magnet, new type of magnetic filter that can separate CRUD efficiently and eventually reduce radiation exposure of personnel at nuclear power plant is suggested. This separator consists of inner and outer magnet assemblies, coolant channel and container surrounding the outer magnet assembly. The rotational motion of the inner and outer permanent magnet assemblies surrounding the coolant channel by driving motor system produces moving alternating magnetic fields in the coolant channel. The CRUD can be separated from the coolant by the moving alternating magnetic field. This study describes the results of preliminary experiment performed with the different flow rates of coolant and rotation velocities of magnet assemblies. This new magnetic filter shows better performance results of filtering the magnetite at coolant (water). Flow rates, rotating velocities of magnet assemblies and particle sizes turn out to be very important design parameters.

Key Words : CRUD, magnetic filter, alternating magnetic field, separation

1. Introduction

It is important to reduce the build-up of CRUD (Chalk River Unidentified Deposit) or to remove the CRUD from the reactor coolant system because major source of occupational radiation exposure is coming from the CRUD in the nuclear power plant. Major radioactive corrosion products, Co^{58} and Co^{60}, are known to contribute approximately more than 70% of the total occupational radiation exposure. The ICRP 60 for the radiation protection for the public requires much strict reduction of the occupational radiation exposure. It is more demanding requirement to reduce the build-up of CRUD radioactivity and to increase the removal rate of CRUD in the primary coolant system for the radiation exposure reduction. There are several passive ways to reduce the radiation levels around the primary water system, i.e. improvement of coolant purification system, high pH operation, adoption of low corrosive and low cobalt containing materials in the primary coolant system and the more frequent periodical decontamination of the primary system. [1] By the middle of 1980s, the technology using electromagnetic field for CRUD removal was studied world widely, it was not, however, continued due to some problems of electromagnetic filter. In Japan, the filter using permanent magnet instead of electromagnetic field was developed in the latter of 1980s. [2] It was performed tests at the Halden Test Reactor in Norway. Although test results of magnetic filter were not so bad, that project was stopped in the reason of economic problem by the support authority. After that time, the study related CRUD removal is stagnated except water chemistry control. Recently, manufacturing technology of permanent magnet(especially, rare earth magnet) has been advanced greatly, and these new magnets can generate inexpensively stronger magnetic field than the conventional magnet. Thus, the magnetic filter using these magnets has been introduced and proposed and the basic principle of this magnetic filter is similiar to that developed in Japan.

The materials that normally come into contact with the coolant streams of the thermal power systems are metal alloys containing the elements such as iron, nickel, copper, chromium, cobalt, aluminum, zinc, titanium, zirconium, carbon, and manganese as major constituents. All these elements react chemically with water and the dissolved oxygen to form oxides (mixture of metal oxides). [3] The structure of these oxides is known as a spinel, which comprise of Fe^{2+}, Fe^{3+}, and Fe_3O_4 with partially substituted of Fe^{2+} with Ni^{2+} and Co^{2+} while Fe^{3+} with Cr^{3+} and Co^{3+} cations. The oxides show relatively low solubilities and display quite varied magnetic properties and are transported by the coolant stream and then deposited throughout the systems where these oxides induce adverse effects on the power plant operation. [4]

An important class of the oxides is known as the ferrites, in which iron is the major metallic constituent. It was based on the assumption that the corrosion product was magnetite (Fe_3O_4). However, a more careful research confirms that corrosion products are more like composed of nickel-ferrite ($Ni_xFe_{3-x}O_4$). The ferrites show strong magnetic properties in contrast to the other corrosion products that show very much weaker magnetic properties. The stable form of Fe_2O_3 at low temperatures is the well-known hematite, α-Fe_2O_3 [5] This oxide, or its hydrated form, is the usual iron corrosion product found in the condensate or feedwater systems of fossil and

Table 1. Magnetic Properties of Metals and Metal Oxides

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

nuclear power plants during startup, when the exposure to the atmospheric oxygen and temperatures are low. At high temperatures in the presence of oxygen, another valence-3 oxide known as maghemite or γ-Fe_2O_3 may form. [6,7,8] A comparison of magnetic property of the elements at the major important corrosion products is presented in Table 1.

2. Magnetic Filter Using Permanent Magnet

Most of the CRUD (nickel ferrite, magnetite, etc.) possesses the characteristic of showing strong ferrimagnetisms. The magnetic filter using permanent magnets can be a new type of magnet filter system and can be used efficiently at rigorous conditions such as high temperatures and high pressures. [2] The system is composed of two main parts; a separator and a driving motor. The separating part consists of the inner and outer magnet assemblies, fluid(coolant) channel and container surrounding the outer magnet assembly. Fluid channel is located between the inner and outer permanent magnet assemblies. Corrosion products (CRUD) in the fluid are flowing through the channel between the magnet assemblies and collected by the strong magnetic field, and then moved down along with the rotation of driving motor connected to the separator. Rotation of permanent magnet assembly and shifted arrangement of permanent magnets generate the moving alternative magnetic fields. CRUD, such as magnetite in the magnetic field, will then be magnetized. Thus, CRUD could be transferred and move along with the forward direction of the moving alternating magnetic field. CRUD which are collected and divided from the coolant could be deposited inside sludge at the vessel wall. Then, CRUD can be separated from the coolant by the moving alternating magnetic force. The effectiveness of the magnetic filter to separate the particles from a fluid stream strongly depends on the relative magnitudes of the magnetic attractive force. In general, the competing forces are those due to the hydrodynamic drag and inertial effects on the particle. Fig. 1 is a cross sectional top view of the separator which shows the inner and outer magnet assemblies, and fluid channel. By maximizing magnetic field with iron to prevent magnetic field leakage in other direction, each of magnet faces a respective opposite polar magnet. Located between the inner and outer assembly, the fluid channel is influenced by the strong magnetic field produced by the coupled magnets. Fig. 2 shows schematic drawing of fluid channel

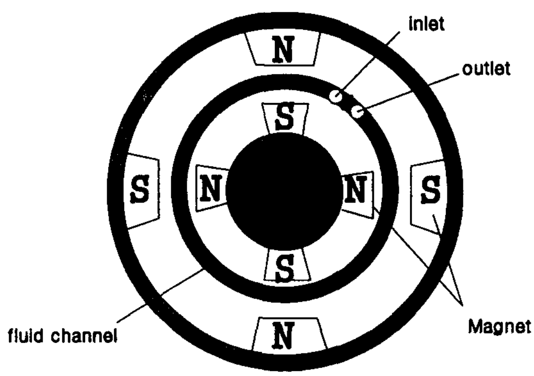

Fig. 1. Cross Sectional Top View of Separator

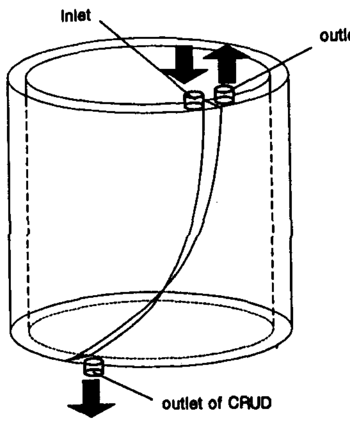

Fig. 2. Schematic Drawing of Fluid Channel

which displays inlet and outlet of fluid, outlet of CRUD, and inclined boundary wall. CRUD in the fluid(coolant) is captured by magnetic field and moves to the rotating direction. Arriving at boundary wall, CRUD could be accumulated at the corner of fluid channel. As stated earlier, CRUD can then easily be separated with an appropriate batch operation.

3. Theoretical Approach

The analysis of the movement of CRUD (magnetic particle) under moving alternating magnetic field is performed as follows. First, when the particle moves under the magnetic field, the particle is influenced by both magnetic force and viscous drag force. [9,10,11,12]
The magnetic force can be described by

$$F_m = V_0 \mu_0 \chi H \frac{dH}{dx} \ , \qquad (1)$$

where
F_m=magnetic force (N), V_0=particle volume (m^3),
μ_0= magnetic permeability of free space (Tm/A), χ = susceptibility,
H = magnetic intensity (A/m), x= axial distance (m).

The strength of the moving alternating magnetic field H can be expressed by the following equation. [13]

$$H = H_0 \sin(kx - wt) \qquad (2)$$

where
k = frequency of magnetic field (m^{-1}),
w = angular frequency (sec^{-1}),
t = time (sec),

The viscous drag force yields the following equation of motion, [14]

$$F_D = \frac{\rho V^2 A_p C_D}{2} \ , \qquad (3)$$

where
F_D = drag force (N),
ρ = density (kg/m^3),
V = velocity of particle (m/sec),
A_p = area (m^2),
C_D = drag coefficient.

Then finally the CRUD motion is governed by the following equation:

$$m\frac{dV}{dt} = -F_D + F_m \ , \qquad (4)$$

where
m = mass of particle (kg).

With eqs. (1) ~ (3), eq. (4) becomes

$$\frac{dV}{dt} + \frac{9\mu}{2a^2\rho_p}V = \frac{\mu_0 \chi k H_0^2}{2\rho_p}\sin 2(kx - wt) \ , \qquad (5)$$

where
μ = viscosity (kg/msec),
a = radius of particle (m),
ρ_p = density of particle (kg/m3).

If the moving velocity of particle (CRUD) is considered as constant, the particle velocity can be expressed as follows:

$$V_{CRUD} = \frac{ka^2 \mu_0 \chi H_0^2 \sin 2(kx - wt)}{9\mu} \qquad (6)$$

For a constant magnetic strength, the velocity of the particle varies with the frequency(related to k), which depends on the rotating velocity of magnet assembly, magnetic field strength and drag force of fluid (viscosity). Other parameters are terms which is related to magnetic properties of particle. The separation factor can be defined as the water flow to the particle velocity ratio.

$$V_{flow} = \frac{Q}{A} \qquad (7)$$

where

Q = flow rate (m^3/sec),

A = flow area (m^2).

Finally, the separation factor (η) can be expressed as

$$\eta \propto \frac{V_{CRUD}}{V_{flow}} = \frac{Aka^2\mu_0\chi H_0^2 \sin 2(kx - wt)}{9\mu Q} \qquad (8)$$

4. Experiment

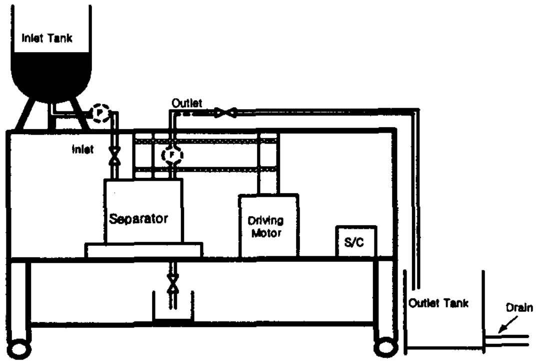

Fig. 3. Schematic Drawing of Magnetic Filtering System

Fig 3. depicts a schematic drawing of experimental equipments. Water in inlet reservoir flows from inlet tank to outlet tank through the pump, separator and flow meter. The driving motor, which equipped with controller to adjust rotating velocity, rotates magnet assembly in separator. Details of each component are described in Table 2. Test runs are conducted at room temperatures and atmospheric pressures because the experimental systems, which are in the preliminary stage, is not yet loop type but open-pool type. Magnetite (Fe3O4) with the ferrimagnetic property is used as dummy CRUD. The magnetite used in these test runs are manufactured by Aldrich Chemical Company and its size is around 5 micrometer. Fig 4 shows the distribution of particle size of magnetite used in experiment. The particle size of magnetite powder is measured by a particle counter.(PMS 20 channel) Three grams of magnetite (about 50ppm)

Table 2. Description of Each Component of the System

		Material	Size & capacity	Comment
Magnet	Inner assembly	Nd	25×30×20, ~ 4000G	60 EA (rare earth)
	Outer assembly	Nd	56×30×20, ~ 4000G	60 EA (rare earth)
Frame		Al	1040×1530×600	
Flow meter			~ 2.5 gpm	1 EA(rotameter)
Valve		SUS 316		4 EA(ball type) control of flow rate
Motor		~ 120 rpm		
Reservoir		SUS 304	60 liter	
Pump			15 liter/min	1 EA
Pipe		SUS 304		
Fluid channel		SUS 316		1 EA
Vessel		Al	1 EA	

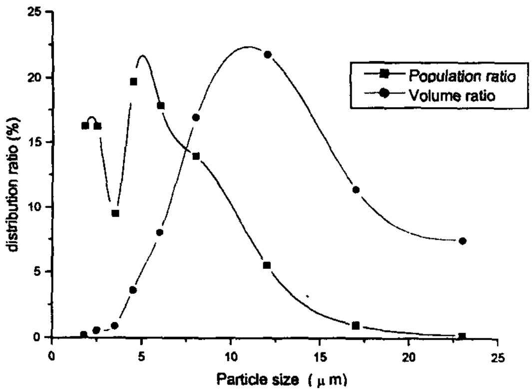

Fig. 4. Particle Size Distribution of Magnetite Powder

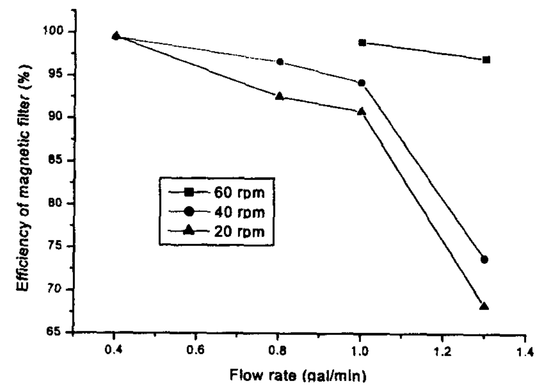

Fig. 5. Experiment Result with Respect to Flow Rate

is mixed in inlet water at each test run. Experimental results have been obtained for varying flow rates (0.4~1.3 gallon per minute) and rotating velocities (20~80 revolutions per minute) of magnet assembly. A highly strong magnet (~5000Gauss), which is rod type, has been used for measuring the concentrations of dummy CRUD of outlet water. The magnet rod is inserted and stirred in the outlet water tank, then most of the magnetite in water is captured or sticked to the magnet surface. Finally, the dried mass of the magnetite is measured.

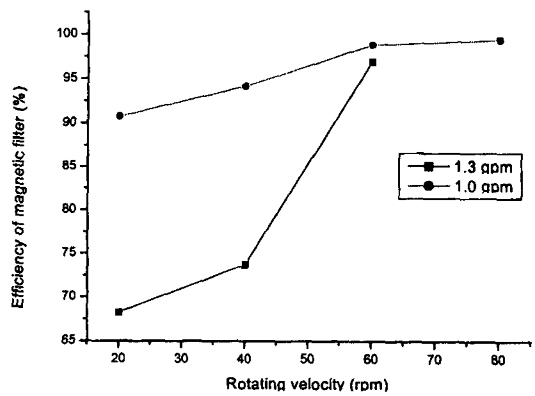

Fig. 6. Experiment Result Respect to Rotating Velocity of Magnet Assemblies

5. Results and Discussion

As shown in equation (8), the flow rate of fluid (Q term), rotating velocities (k term) and particle size (a term), which affect the efficiency of the magnetic filter of magnet assemblies, are very important parameters. Experimental results with respect to the two parameters (flow rate and rotating velocity) are shown in Fig. 5 and 6. Fig.7 and 8 show the efficiency of magnetic filter as particle size which is measured by particle counter and the size distribution of magnetite at each stage(input, output, separated stage). Basically, the efficiency of magnetic filter shows the tendency to increase as the flow rate is lower, rotating velocity is higher, and particle size is bigger. This new type of magnetic filter showed a good performance relatively compared to the other conventional magnetic filter. In the case of a conventional magnetic filter or electromagnetic filter, only the flow rate turned out to be a dominant parameter along with the magnetic field strength. For the magnetic filter proposed in this study, however, the rotating velocity of magnet assemblies as well as the flow rate is a dominant and important parameter. For a constant rotating velocity of

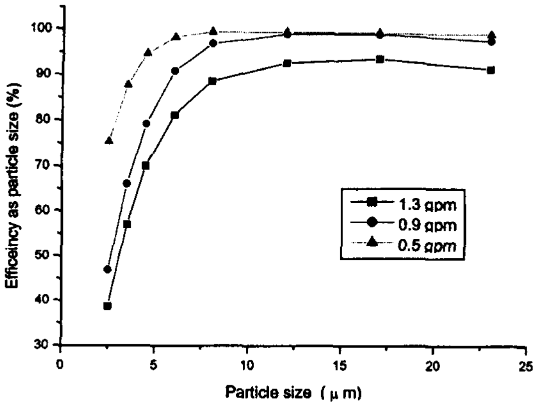

Fig. 7. Removal Efficiency of Magnetic Filter as Particle Size

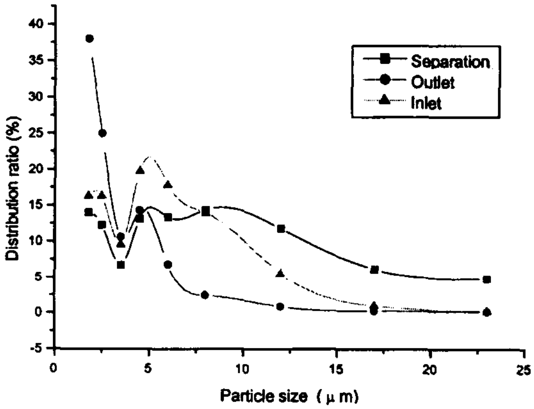

Fig. 8. Particle Size Distribution at Each Stage

magnet assemblies (Fig. 5), the efficiency of magnet filter shows better performances at lower flow rate condition. However, on the other hand, the efficiency rapidly decreased when the flow rates are increased. Fig. 6 shows that an increase in rotating velocity of magnet assemblies improves the efficiency of magnetic filter at constant flow rate condition. Thus, the rotating characteristic of magnet assemblies could be a major advantage and provide a useful flexibility of this magnetic filter system. This may resolve the problem arising with restricted operational conditions of flow rates with moving alternating magnetic field.

The concentrations of the magnetite in outlet water are directly measured through routine procedures. In other words, either ICP or AAS method is used to measure the concentrations of metal elements. Magnetite, however, is oxide form that is hardly soluble. Moreover, it is very difficult to sample homogeneously since magnetite is precipitated as powder form in the outlet tank. Furthermore, one or two samples cannot be counted as representative samples. The analysis method to use particle counter is studying. In the future study, more accurate analysis method will be suggested and applied.

6. Conclusions

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

- Moving alternating magnetic filter using permanent magnets can provide better operational properties than conventional magnetic filters and shows some potential and advantage for the CRUD removal.
- The efficiency of magnetic filter shows the increasing tendency at lower flow rates, higher rotating velocities of magnet assemblies and bigger particle size.
- In addition to the flow rates, the rotating velocity of magnet assemblies also could be an important parameter to improve the efficiency of magnetic filter and provide the useful flexibility for better operation and performances.
- The proposed magnetic filter can resolve the problems associated with the limited operational conditions of flow rate by taking the advantage of the moving alternating magnetic field.

The application of moving alternating magnetic filter could provide an effective method for the removal and separation of CRUD. The proposed method of removing CRUD also could be applied in general to other industry fields, once the current and various future experiments are proved to be successful and the performances of the magnetic filter are verified by the future improvements.

Acknowledgement

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