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A Study on the Method for the Removal of Radioactive Corrosion Products Using Permanent and Electric Magnets

영구자석과 전자석을 이용한 방사성 부식생성물 제거방안 연구

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Abstract

The removal of radioactive corrosion products from the reactor coolant through a magnetic filter system is one of the many approaches being investigated as a means to reduce radiation sources and exposures to the operational and maintenance personnel in a nuclear power plant. Many research activities in water chemistry, therefore, have been performed to provide a filtration system with high reliability and feasibility and are still in process. In this study, it was devised the magnetic filter system with permanent and electric magnets to remove the corrosion products in the coolant stream taking an advantage of the magnetic properties of corrosion particles. Permanent magnets were used for separation of corrosion products and electric magnets were utilized for flocculation of colloidal particles to increase in their size. Experiments using only permanent magnets, in the previous study, displayed the satisfactory outcome of filtering corrosion products and indicated that the removal efficiency was more than 90 % for above 5 ㎛ particles. Experiments using electric magnets also showed the good performance of flocculation without chemical agents and exhibited that most corrosion particles were flocculated into lager aggregates about 5 ㎛ and over in diameter. It is, thus, expected that the magnetic filter system with the arrangement of permanent and electric magnets will be an effective way for the removal of radioactive corrosion products with considerably high removal efficiency.

Key words: radioactive corrosion products, magnetic filter system, permanent magnets, electric magnets, particle size

요 약

자기필터시스템을 이용한 원자로 냉각재로부터의 방사성 부식생성물 제거는 원자력 발전소의 운전 및 유지보수 종사자에 대한 방사선 피폭 준위를 낮추는 방법으로 많은 연구가 이루어지는 분야 중 하나이다. 그 결과, 보다 높은 신뢰성과 여과성능을 갖춘 자기필터를 개발하고자 수화학 분야에서는 많은 연구가 이루어지고 있다. 본 연구에서는 부식생성물의 자기적 성질을 이용하여 원자력 발전소 냉각재내의 방사성 부식생성물을 제거하기 위해 영구자석과 전자석이 조합된 자기필터시스템을 개발하였다. 영구자석은 부식생성물의 여과를 위해 사용되며 전자석은 아주 미세한 콜로이드 부식생성물 입자의 크기를 증가시키기 위한 응집에 이용된다. 선행연구에서 영구자석만을 사용한 필터 실험결과 대부분의 부식생성물 입자에 대해 만족할만한 수준의 제거효율을 달성하였으며 특히, 크기가 5㎛ 이상인 입자의 경우 제거효율은 90%를 상회하였다. 전자석을 이용한 응집 실험결과 화학응집제의 첨가 없이 대부분의 부식생성물 입자가 전자기장에 의해 응집하여 크기가 5㎛ 이상으로 증가되어 응집실험에 대해 전반적으로 만족스러운 결과를 도출하였다. 따라서, 영구자석과 전자석이 조합된 자기필터시스템은 방사성 부식생성물 제거를 위한 효과적인 방법으로 높은 제거효율을 보여주리라 여겨진다.

중심단어: 방사성 부식생성물, 자기필터시스템, 영구자석, 전자석, 입자의 크기

Ⅰ. Introduction

In a pressurized water reactor (PWR) power plant, high-purity water is used as neutron moderator, as reactor coolant, and as heat transport medium all over the steam water cycle. Thus, it is always the same water or steam which comes in contact with a variety of components during circulation in this single circuit arrangement. At all material surfaces, corrosion and possible erosion-corrosion process occur. The rate of these has to be minimized by selecting suitable material combinations on one hand and by maintaining well-defined water chemistry conditions on the other hand. Even extremely slow corrosion processes generate corrosion products which are to some extent released from the metal surfaces to the following medium in ionic, colloidal and particulate form. Owing to large surface areas, these impurities create significant corrosion product concentrations. Once released to the process stream, corrosion products can be transported to the reactor core and can deposit on the fuel, thus leading to the formation of activated nuclides, such as the long-lived cobalt-60. The subsequent release of activated corrosion products from the core to the coolant makes them available for incorporation in corrosion films on the material surfaces, not only of reactor primary and auxiliary systems, but throughout the steam-water cycle.

It is recognized that about 70 % of the radiation sources in a nuclear power plant (NPP), contributing to occupational radiation exposure (ORE), is attributed the deposits of irradiated corrosion products that cover all internal surfaces of the reactor coolant system (RCS). These corrosion products arise from the ex-core plant internal surfaces and are irradiated by neutron flux on the reactor core. An important class of corrosion products is known as the ferrites, which are derivatives of magnetite or metallic constituent and these ferrites have a comparatively low solubility and varied magnetic properties. For example, the corrosion product formed in the primary coolant system and deposited on the reactor core of a pressurized water nuclear reactor plant has been identified by X-ray spectrometry to be a nonstoichiometric nickel ferrite [1]. Hence, corrosion products are easily removed from suspension by a magnetic filter. The magnetic filter has a relatively low inherent hydraulic resistance and is capable of filtering under RCS operating temperature and pressure.

The present study developed a conceptual design to construct the novel magnetic filter that would remove corrosion products, for an active method of improving the water quality of the primary coolant. The devised permanent magnet filter (PMF) with strong magnetism demonstrated the removal efficiency for corrosion products of greater than 80 % and particularly over 90 % for particles larger than 5 ㎛ [2]. The cohesive device using electric magnets was also developed to flocculate the corrosion particles into larger aggregates without chemical agents. Experimental results of the cohesive device showed that most corrosion particles within the range of 0.1 to 3 ㎛ in diameter was aggregated into particles of 5 ~ 8 ㎛ [3, 4, 5]. Thus, the cohesive device was positioned to flocculate particles before the PMF to increase the removal efficiency of the PMF for very fine corrosion products from 80 % to over 90 %. This paper focuses on the application of permanent and electric magnetic field to remove radioactive corrosion products and presents the results of several experiments that show how the novel magnetic filter using permanent and electric magnets improves the removal efficiency for corrosion products.

Ⅱ. Basic Principles

1. Magnetic Filtration

Magnetic materials are classified into the following types of magnetisms: paramagnetism, diamagnetism, ferrimagnetism, and ferromagnetism. These magnetisms result from the motion of electrical charges, either electron spin or orbital motion. Every electron in the atom generates an element of magnetism. Fortunately, nuclear power plant corrosion products show a relatively strong ferromagnetism. Ferromagnetism has a net magnetic moment in the absence of an external magnetic field and this spontaneous magnetization causes a strong magnetic interaction among corrosion product materials [6].

The efficiency of a magnetic filter in trapping particles from a fluid stream depends on the relative magnitude of the magnetic attractive force and on the combined forces tending to keep the particles in suspension. In general, the competing forces are those due to hydrodynamic drag and to the gravitational field. In the idealized one-dimensional isotropic case, the forces acting on a particular particle in a magnetic field can be presented as [7]

$$|F_m|_x = \frac{1}{2}\mu_o VM\frac{dH}{dx} \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots \text{(1)}$$

where $|F_m|_x$ is the magnetic force for x coordinates, μ_o is magnetic permeability at free space, V is the particle volume, and M is the magnetization. The product, VM, is the magnitude of the particle acted on by the field gradient, dH/dx. The hydrodynamic drag force for particles in the present case can be represented by Stoke' s Law [8]

$$F_d = 3\pi\eta_f d_p u \quad \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots \text{(2)}$$

where F_d is the drag force, η_f is the viscosity of the medium, u is the velocity of particle relative to the fluid stream, and d_p is the diameter of particle. The gravitational force is given by

$$F_g = \frac{4}{3}\pi r_p^3(\rho_p - \rho_f)g \quad \cdots\cdots\cdots\cdots (3)$$

where F_g is the gravitational force, r_p is the radius of the particle, ρ_p is the particle density, ρ_f is the fluid medium density, and g is the acceleration of gravity. Therefore, the criterion for successful trapping a particle in the magnetic filter can be described as follows.

$$\left|F_m\right|_r > \left|F_g\right|_r + \left|F_d\right|_r \quad \cdots\cdots\cdots\cdots (4)$$

where the subscript r represents the radial component. The above equation shows that the relative component magnitudes of the three forces normal to the filter element surface determine whether a particle is trapped on the surface or whether it remains suspended, or becomes resuspended, in the fluid stream.

2. Electromagnetic Flocculation

Coolant water from a nuclear power plant usually contains some dissolved and suspended solids, such as corrosion products. The size distribution of most suspended particles is within a range of 0.001 to 10 micron in diameter. Suspended particles at the lower end of the size spectrum do not readily settle. A suspension of particles that will not settle is known as a stable suspension and the particles that make up these suspensions are known as colloids. Colloidal particles are defined by size. Their size range is generally considered as being from 0.001 to 1 micron [9]. The unique behavior of colloidal particles is a result of surface phenomena. The colloids have a very large ratio of surface area to mass. Their mass is so small that the gravitational force has little effect on their behavior. The principal phenomena controlling the behavior of colloids are electrostatic forces, van der Waals forces, and Brownian motion.

Electrostatic force is the principal force contributing to the stability of the colloidal suspension. Most colloids are electrically charged. The nature of this charge varies somewhat, depending on the nature of the colloid. Metallic oxides are generally positively charged, while nonmetallic oxides and metallic sulfides are generally negatively charged. The result of this electrical charge is that colloids of similar charge will repeal each other. The surface charge on the colloids attracts ions of opposite charge, known as counter ions. These ions, which include hydrogen and other cations, form a dense layer adjacent to the particle known as the stern layer. Water molecules are also attracted to the colloidal particle. The attraction of water is due to the asymmetric electrical charge of water molecules. A second layer of ions, known as the diffused layer, is also attracted to the colloid. In this layer, ions of both electrical charges are attracted, but counter ions predominate. The two layers together are often referred to as the double layer.

A force of attraction exists between any two masses. The magnitude of this attraction is a function of the mass of the two bodies and the distance between them. The attraction is known as the van der Waals forces. In colloidal chemistry, van der Waals forces are the antithesis forces. As indicated in Figure 1(a), the force of repulsion due to the electrical charge will normally repel the colloids before they can move close enough for van der Waals forces to become significant [9]. If the magnitude of the electrostatic forces could be reduced, the particles would move close enough for van der Waals forces to predominate.

Electromagnetic flocculation is a treatment process to destabilize colloidal particles. In this process, electromagnetism plays the vital role to break down the stabilizing forces of colloids in water. Electromagnetic flocculation process typically occurs by a combination of two mechanisms: compression of the double layer and counter-ion adsorption [9].

Compression of the double layer can be accomplished by the addition of electromagnetism to the water. The electromagnetic field gradient generated by electromagnetic solenoids and ferromagnetic matrices destabilizes the balance of surface charge on the colloidal particles and attracts the ions of opposite charge toward the colloid surface. As the concentration of counter ions on the colloid surface increases, the counter-ions cause the net charge in the diffused layer to neutralize and result in the compression of this layer. This compression affects the thickness of the entire double layer and so allows colloids to come closer together. If the colloids can be caused to come close enough together for van der Waals forces to predominate, the colloids will agglomerate into a floc. This phenomenon is illustrated in Figure 1(b).

The counter-ions can also be adsorbed onto the surface of the colloidal particles. In this way, the repulsive charges on the surface of the particles may be fully neutralized by the charges carried by the counter-ions. Therefore, the destabilized colloidal particles can adhere to each other to form colloidal-colloidal complexes, by van der Waals attractions or by further adsorption of counter-ions.

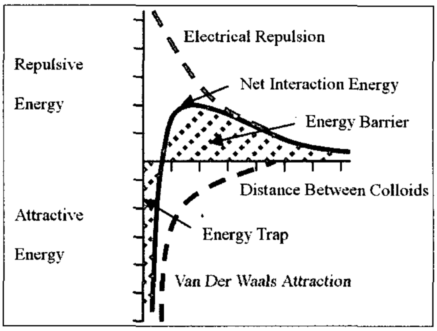
Ⅲ. Magnetic Filter System

1. Mechanical Design

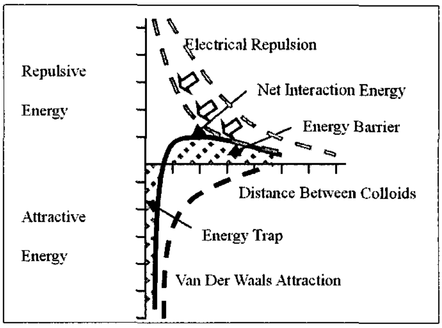
The magnetic filter using permanent and electric magnets was devised to improve the corrosion product removal rate. The magnetic filter is composed of two main devices: the permanent magnet filter (PMF) and the cohesive device using electric magnets (Fig. 2) [3, 10]. The PMF was manufactured to separate metallic particles from the main stream. The cohesive device was developed to cause the very fine suspended magnetic particles to flocculate into larger aggregates that can be separated easily in the PMF [4, 5].

The PMF adopts the wiggler configuration, using cylindrical magnet bars. In this wiggler configuration, there are two rows of magnets arranged parallel to one another and separated by a certain distance [10]. Once inside the wiggler configuration, the particles are influenced by a spatially varied magnetic field that causes them to oscillate back and forth in a plane perpendicular to the magnetic field [11].

The PMF is composed of two main parts: a separator and a driving motor. The separator



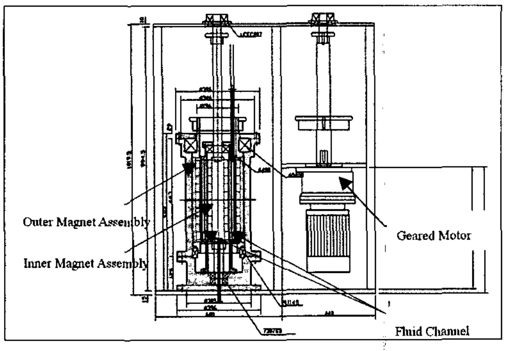
(a) A Stable Colloid



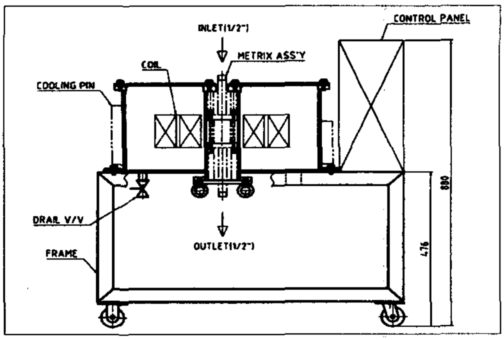
(b) A Colloid after Compression

Fig. 1. Forces Acting on Colloids

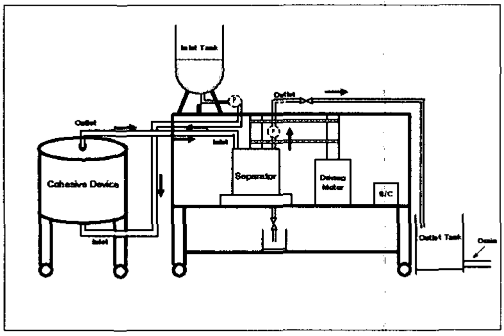
consists of inner and outer assemblies, fluid channels, and a container surrounding the outer magnet assembly. The fluid channel is located between the inner and outer permanent magnet assemblies. The rotation of the permanent magnet assemblies and the shifted arrangement of the permanent magnets generate the alternating magnetic field in the fluid channel. To maximize the magnetic field each magnet faces an opposite respective polar magnet. Corrosion particles in the fluid are magnetized as they pass through the channel between the magnet assemblies, due to the strong magnetic field; the particles then move in the direction of the permanent magnets, rotated by a driving motor connected to the separator. Finally, the corrosion products are accumulated at the bottom corner of the fluid channel and are separated from the fluid stream at the boundary wall of the vessel.



(a) Permanent MagnetFilter



(b) Cohesive Device



(c) Combined Magnetic Filter

Fig. 2. Mechanical Design of the Magnetic Filter

The cohesive device is composed of three main parts: a vessel with inlet and outlet pipe connections, which are fitted to the inside with a solenoid to generate a background magnetic field and a structure to cool the magnet coils; the magnetizable matrix assembly; and the control panel [3, 4, 5]. The whole vessel is supported concentrically to an electromagnetic solenoid. The vessel is made of nonmagnetic stainless steel to avoid any short-circuiting of the magnetic field. Since the magnetizing current generates considerable heat and since additional heat is produced from the vessel itself during the operation, the vessel is furnished with a cooling system; the electromagnetic solenoid is cooled directly by oil surrounding it, filled into the vessel. The ferromagnetic matrix is a critical design feature of this cohesive device and plays a key role in the agglomeration effectiveness. Without the ferromagnetic matrix or with a nonferromagnetic matrix, the magnetic field would be uniform in intensity throughout the volume concentric to the magnets, and no agglomeration effect would occur. The ferromagnetic matrix assembly adopts the ball type of steel matrix, and its case is made of an acrylic material to avoid corrosion release from the matrix assembly. The control panel functions as the necessary protective interlocks to prevent damage to the electromagnet under fault conditions. The control panel is also equipped with on-off timer and a programming device, which automatically controls the sequence of run-stop operations.

2. Operational Principal

The principle behind the operation of the magnetic filter is simple. First, the input particles in the water tank flow into the cohesive device by the pump. As corrosion particles in the cohesive device pass through the matrix assembly along the pipe line, they are magnetized by the background magnetic field gradient generated by electric magnets and ferromagnetic matrices. This gradient in the background field causes the attraction of particles with positive susceptibility toward the surface. The very fine magnetized particles on the matrix surface eventually flocculate into larger aggregates by a process attributable to the potential differences between each particle while the power supply timer is on. Power is supplied for a specific interval, after which the timer turns off, and the magnetized corrosion aggregates flow out from the matrix assembly and into the PMF. Corrosion aggregates in the PMF are also magnetized as they pass through the channel between the magnet assemblies, due to the strong magnetic field; the particle aggregates then move in the direction of the permanent magnets, rotated by a driving motor connected to the separator. Finally, the corrosion products are accumulated at the bottom corner of the fluid channel and are separated from the fluid stream at the boundary wall of the vessel.The operation of the magnetic filter is shown in Figure 3.

3. Experiments and Results

To be consistent with previous experiments, the experimental environment is established similar to that of the PMF and the cohesive device, and experiments were usually performed under room temperature and atmospheric pressure. Various experiments were conducted with changes to the following parameters: the class of particles, the flow rates, the rotating velocity of permanent magnet assembly, the concentration of the input solution, and the particle size. Nickel ferrite ($NiFe_2O_4$), cobalt ferrite ($CoFe_2O_4$), and magnetite (Fe_3O_4) were used to simulate corrosion products. Before the experiments,

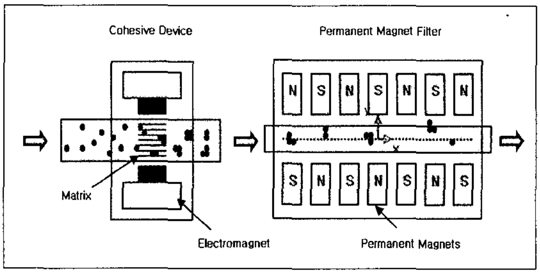


Fig. 3. Operation of the Magnetic Filter

Table 1. Experimental Conditions of Magnetic Filter

Conditions	Classes and values
Particles	Nickel ferrite ($NiFe_2O_4$, Purity 99.9 %) Cobalt ferrite ($CoFe_2O_4$, Purity 99 %) Magnetite (Fe_2O_4, Purity 98 %)
Flow rates	1 ℓ /min, 3 ℓ /min, 5 ℓ /min
Rotating velocity of magnet assembly	30rpm, 50rpm, 70rpm
Particle concentrations	1ppm, 10ppm, 20ppm
Particle size	0.1 ~ 35 μm
Operation time of electromagnets	1 min-ON and 10 sec-OFF (Fixed parameter)
Applied magnetic fields	5K Gauss (Fixed parameter)

the original size distribution of each particle was measured, and particles were found to be within the range of 0.1 to 35 ㎛ in diameter. The detailed experimental parameters are presented in Table 1.

To acquire the distribution of particle size, the particle counter which counts the number of particles in an aqueous solution was used. The particle counter uses a halogen lamp for irradiating the sample particle in a solution and its shadow equivalent to the sample size throws an image on the photodiode. These numerical values acquired from the photodiode are processed by computer numerical analysis and the particle counter finally displays the distribution of particle size.

The magnetic filter displays the satisfactory filtration ability that the removal efficiency for all corrosion products is highly improved. Figure 4 illustrates an increase of removal rate of the combined filter compared with that of the PMF only. The increased removal rate reaches over 90 %.for all particles with a 1 liter/min flow rate and a 50 rpm rotating velocity of the magnet assembly. Consistent with the results of the PMF only, the flow rate plays a dominant role in magnetic filtration of the combined filter. As the flow rate is decreased, the filtration efficiency for corrosion products is increased. This result also indicates the higher flow rate, the higher increase of removal rate (Fig. 5). This phenomenon is attributed to the flocculation of particles; the particles, which are not separated in the PMF due to the high flow rate, flocculate into lager aggregates passing through the cohesive device and these larger particles are easy to be removed in the PMF. Incidentally, the rotating velocity of permanent magnet assembly and the concentration of the input solution were not important parameters for determining the removal rate for corrosion particles. There is no remarkable difference in the removal efficiency for the high and low rotating velocity of permanent magnet assembly. The result of magnetic filtration according to the rotating velocity of permanent magnet assembly

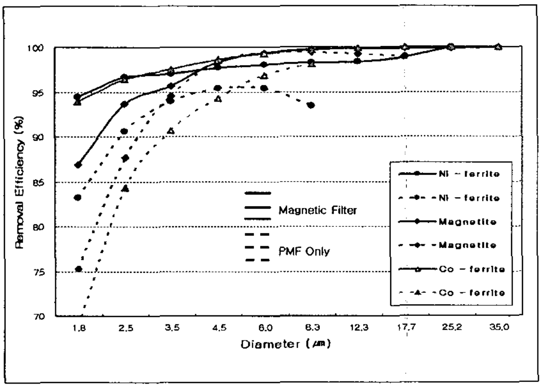


Fig. 4. Removal Efficiency according to the Class of Particles, 10ppm, 50rpm, 1 liter/min, 5K Gauss

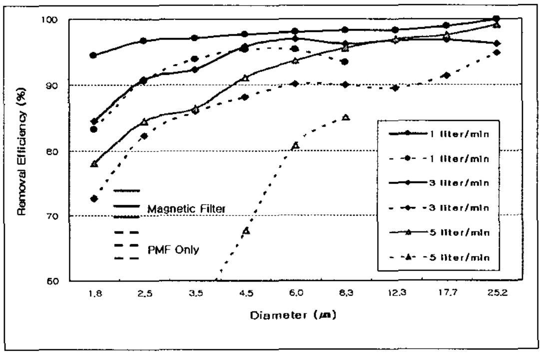


Fig. 5. Removal Efficiency (Flow Rate, Nickel ferrite, 10ppm, 50rpm, 5K Gauss)

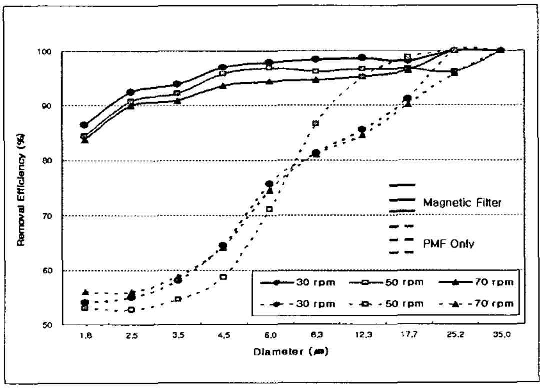


Fig. 6. Removal Efficiency according tothe Rotating Velocity of Magnet Assembly, Nickel ferrite, 10ppm, 3 liter/min, 5K Gauss)

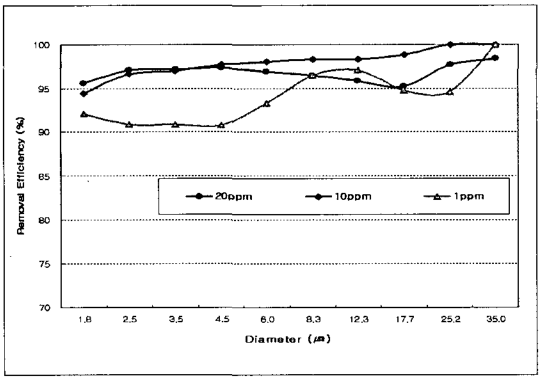


Fig. 7. Removal Efficiency (Concentration of Input Solution, Nickel ferrite, 1 liter/min, 50rpm, 5K Gauss)

for nickel ferrite is shown in Fig. 6. The results also showed removal efficiency of more than 90 % for nickel ferrite regardless of the concentration of the input solution under the following conditions: a 1 liter/min flow rate and a 50 rpm rotating velocity of the magnet assembly (Fig. 7). Some disparities in the removal efficiency between the high and very low concentration, such as 1ppm, is attributed to the decrease in the number of corrosion particles filtered by the magnetic filter. The particle size made great contribution to the removal rate. Corresponding to the result of PMF only, the removal efficiency increases for a lager particle size. Figure 8 demonstrates the increased removal efficiency on the average for nickel ferrite, magnetite, and cobalt ferrite. In particular, there was a higher increase of removal rate for a smaller particle size and it was considered that the cohesive device was to cause the very fine suspended magnetic particles to flocculate into larger aggregates that could be separated easily in the PMF. To compare the experimental results of combined magnetic filter with the previous results of the PMF, both results are plotted in the same figure; a solid and dotted line indicates the results of combined magnetic filter and the PMF only, respectively.

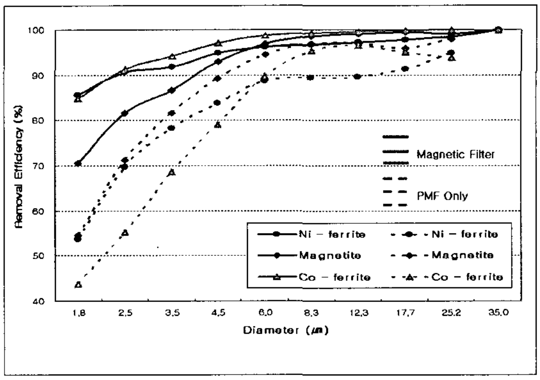


Fig. 8. Removal Efficiency (Particle Size, 50rpm, 5K Gauss

Ⅳ. Conclusion

This study involved the experiments of a magnetic filter with permanent and electric magnets to reduce the risk of occupational radiation exposure (ORE) in nuclear power plants. Previous researches have showed that the removal efficiency of permanent magnet filters (PMF) to be greater than 80 %, for various corrosion products [2]. Preliminary experimental results demonstrated that the removal efficiency was higher if the flow rate of fluid stream was lower and the size of the particles was larger. Although the rotating velocity of the magnet assemblies had a slight influence on the removal efficiency, the flow rate and the particle size were the main parameters for determining the removal efficiency in previous study. The PMF was found to achieve a high removal efficiency of over 90 % for particles with diameters of 5 ㎛ and over.

There was high demand for a means to improve the efficiency of removing corrosion products by increasing the size of corrosion product particles. This task was performed relatively well by the cohesive device developed in the previous study, which uses an electromagnetic field. After passing through the cohesive device, most corrosion particles within the range of 0.1 to 3 ㎛ in diameter was aggregated into particles of 5~8 ㎛ [3, 4, 5]. The flocculation depended mainly on the particle concentration and on the operation time of the electric magnets, exclusive of some influences attributed to the original disparities in the magnetic susceptibility of particles involved. With a higher particle concentration and longer operation time, there is an increase in the number and the size of the aggregates.

Thus, in this study, the cohesive device was set up before the PMF to increase the corrosion product removal rate. Experiments with the novel magnetic filter using permanent and electric magnets showed the highly improved removal efficiency for corrosion products. The removal efficiency of the PMF for very fine corrosion products was increased from under 80 % to over 90 %, under various flow rates and concentrations. Consistent with the results of the PMF, the flow rate and particle size played an important role in magnetic filtration for particles, and the removal efficiency increased for a lower flow rate of the fluid stream or a larger particle size.

With due consideration of the characteristics of the primary coolant in nuclear power plants, the magnetic filter flocculates the corrosion particles using only electromagnetism without any chemical agents and then separate them from the fluid stream. This device also shows the remarkable removal rate of corrosion products. The potential of utilization for nuclear power plants is very high due to the several advantages of the novel filter system: no pressure drop, the possibility of application under high temperature and pressure, and no back flushing. Therefore, the novel magnetic filter using permanent and electric magnets, developed in this study, can be applied to an active method to improve the water quality of the primary coolant in nuclear power plants. This filter is especially worthy of consideration from the standpoint of reducing reactor corrosion iron input.

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