

Application of Electromagnetic Fields to Improve the Removal Rate of Radioactive Corrosion Products

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Abstract

To comply with increasingly strict regulations for protection against radiation exposure, many nuclear power plants have been working ceaselessly to reduce and control both the radiation sources within power plants and the radiation exposure experienced by operational and maintenance personnel. Many research studies have shown that deposits of irradiated corrosion products on the surfaces of coolant systems are the main cause of occupational radiation exposure in nuclear power plants. These corrosion product deposits on the fuel-clad surface are also known to be main factors in the onset of axial offset anomaly (AOA). Hence, there is a great deal of ongoing research on water chemistry and corrosion processes.

In this study, a magnetic filter with permanent magnets was devised to remove the corrosion products in the coolant stream by taking advantage of the magnetic properties of the corrosion particles. Experiments using permanent magnets to filter the corrosion products demonstrated a removal efficiency of over 90% for particles above 5 μm . This finding led to the construction of an electromagnetic device that causes the metallic particulates to flocculate into larger aggregates of about 5 μm in diameter by using a novel application of electromagnetic flocculation on radioactive corrosion products.

Key Words : corrosion products, permanent magnet filter, magnetic separation, flocculation, electromagnets, cohesive device, particle size

1. Introduction

Any nuclear power plant (NPP) is a complex technological system that can comprise up to

100,000 components, most of which operate under extreme conditions, including high temperature and pressure, mechanical and electrical stresses, and irradiation. The parts and

assemblies of NPPs are affected by water and steam flows. The main problem in ensuring the reliable and safe operation of NPPs lies in the operational reliability of both the NPP equipment and the metallic pipeline, both of which are affected by water chemistry conditions.

The materials of an NPP structure that usually come into contact with the primary coolant streams of the power systems are metal alloys that have iron, nickel, copper, chromium, and cobalt as their major components. All of these materials react chemically with water and dissolved oxygen to form oxides known as "corrosion products." An important class of corrosion products is known as the ferrites, which are derivatives of magnetite or of a 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]. The corrosion products are normally transported by the coolant stream and are deposited throughout the power systems where they may cause problems such as axial offset anomaly (AOA). Corrosion products are also recognized as one of the major sources of occupational radiation exposure (ORE) for NPP workers. Consequently, there is a considerable demand for the application of new technology to effectively control corrosion products.

Methods that have been adopted to control corrosion products in NPPs include: improving the coolant purification system, operating the reactor under high pH conditions, adopting materials with low levels of cobalt in the primary coolant system, and decontaminating the primary system more frequently to reduce the radiation levels around the primary coolant system. However, all of these

methods emphasize preventing material corrosion and improving thermo-hydraulic efficiency, rather than eliminating and removing corrosion products in the primary coolant system.

Contrary to the methods above, the present study developed a conceptual design for a permanent magnet filter (PMF), which would remove corrosion products using a magnetic field to actively improve the water quality of the primary coolant. The novel magnetic filter, devised with permanent magnets, generated a relatively strong magnetism and demonstrated a removal efficiency for corrosion products of greater than 80% [2, 3]. An electromagnetic filter of about 5 μm in diameter was set up as a cohesive device before the PMF, causing the corrosion particles to flocculate into larger aggregates and improving the corrosion product removal rate to over 90%. This paper focuses on the application of electromagnetic flocculation to radioactive corrosion products and presents the results of several experiments that show how the proposed cohesive device results in the successful agglomeration of corrosion products.

2. Theory of Magnetization

In order to design a practical permanent magnet filter (PMF) and electromagnetic device, it is essential to understand the physics of magnetization and the trapping principles of magnetic filtration. All materials interact with an applied magnetic field in some way and to some extent, and this interaction plays a significant role in magnetic filtration. Fortunately, nuclear power plant corrosion products display a relatively strong ferromagnetism.

In a large class of materials there exists a constitutive relation that both magnetic density \mathbf{B} and the magnetization \mathbf{M} are proportional to the magnetic intensity \mathbf{H} . If the material is isotropic as well as linear [4], then

$$B = \mu H \quad (1)$$

and

$$M = \chi_m H \quad (2)$$

where μ and χ_m are the permeability and susceptibility of the material, respectively. If χ_m is positive, the material is called "paramagnetic," and the magnetic induction is strengthened by the presence of the material. Even though the atoms of this material have a net magnetic field, the macro effect is zero due to the random orientation of the atoms. If χ_m is negative, the material is called "diamagnetic" and the magnetic induction is weakened by the presence of the material. Contrary to the magnetism remarked upon above, ferromagnetism and ferrimagnetism have 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.

The effectiveness 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 an idealized one-dimensional isotropic case, the forces acting on a particular particle in a magnetic field can be described as [5]

$$|F_m|_x = \frac{1}{2} \mu_0 V M \frac{dH}{dx} \quad (3)$$

where $|F_m|_x$ is the magnetic force for x coordinates, μ_0 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

$$F_d = 6\pi\eta_f r_p u \quad (4)$$

where F_d is the drag force, η_f is the viscosity of the medium, u is the velocity of a particle relative to the fluid stream, and r_p is the radius of the particle. The gravitational or buoyant force is given by

$$F_g = \frac{4}{3} \pi r_p^3 (\rho_p - \rho_f) g \quad (5)$$

where F_g is the gravitational force, 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.

$$|F_m|_r > |F_g|_r + |F_d|_r \quad (6)$$

The above equation indicates 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. These considerations impose theoretical limits on the performance of a cohesive device and magnetic filter.

3. Principles of Magnetic Separation System

3.1. Open Gradient Magnetic Separation and High Gradient Magnetic Separation

Open gradient magnetic separation (OGMS) utilizes the field gradient of the external magnetic field produced by the magnet itself to deflect the magnetic particles from the main stream [6]. In contrast to high gradient magnetic separation (HGMS), OGMS has neither a magnetic filter matrix made of fine ferromagnetic wires nor iron beads for generating the strong magnetic field, but it is possible to apply a continuous process. Since the outlets for concentrates and filtrates are

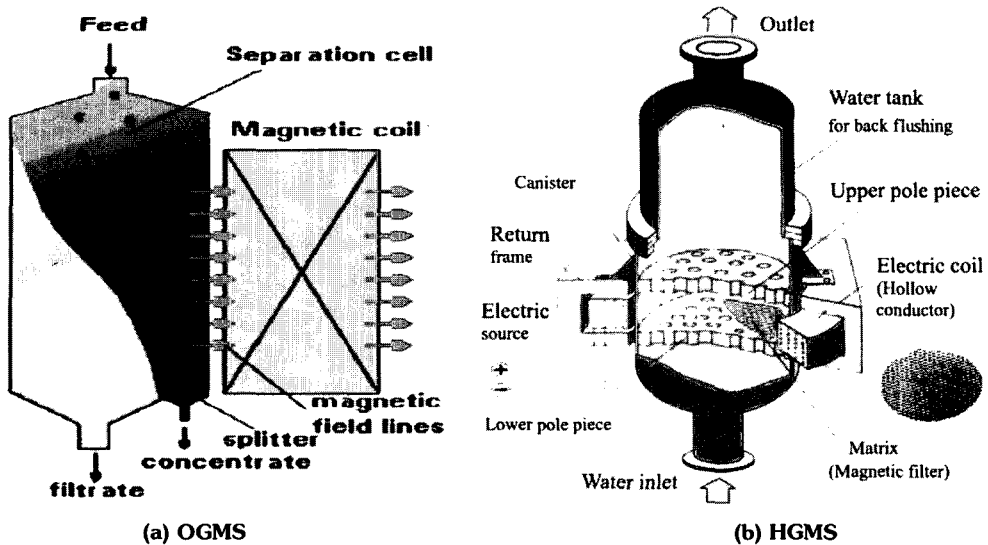


Fig. 1. Principle of OGMS and HGMS

divided, no particle accumulation takes place inside the magnetic separator. Figure 1(a) illustrates the principle of OGMS.

High gradient magnetic separation takes advantage of large magnetic forces around fine ferromagnetic wires or iron beads in the magnetic filter to collect the magnetic particles in matrices. The matrix locally distorts the magnetic field produced by surrounding solenoid magnets and creates large magnetic field gradients in the vicinity of the ferromagnetic surfaces [7]. These surfaces then become trapping sites for magnetic particles. For HGMS, the removal of captured particles by flushing the filter matrix is necessary to avoid blockage of the filter matrix since the accumulation of particles sometimes occurs inside the separator. The principle of HGMS is shown in Figure 1(b).

4. Permanent Magnetic Filtration

4.1. Design for the Practical Permanent Magnet Filter (PMF)

Permanent magnets have several advantages over

conventional electromagnets. The primary advantage is that they can provide a relatively strong magnetic field over an extended spatial region for an indefinite period, with no expenditure of energy. Another advantage of permanent magnets is that they can be fabricated with a wide range of structural properties, geometric shapes, and magnetization patterns, owing to the rapid growth of manufacturing techniques for such magnets. Therefore, in this study, a permanent magnet filter (PMF) was devised that can separate metallic particles from the main stream and, applying the principle of OGMS, can remove the corrosion products in the coolant stream.

The PMF devised in this research adopts the wiggler configuration and uses cylindrical magnet bars. The arrangement of the magnet assembly is shown in Figure 2 [2]. In this wiggler configuration, there are two rows of magnets arranged parallel to one another and separated by a certain distance. 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

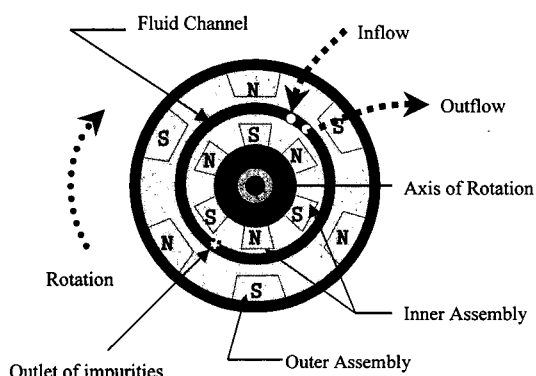


Fig. 2. Cross Section of the PMF Separator

the magnetic field [8].

The PMF is composed of two main parts: a separator and a driving motor. The separator 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 (Fig. 2). Because of the strong magnetic field, corrosion particles in the fluid are magnetized as they pass through the channel between the magnet assemblies. The particles then move in the direction of the permanent magnets, which are 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.

4.2. Results of Experiments with the Permanent Magnet Filter (PMF)

Several experiments were conducted with various

parameters, including the class of particles, the flow rates, the rotating velocities of the magnet assembly, and the concentration of the input solution. Experiments were usually carried out at room temperature and atmospheric pressure since, in this study, the PMF was devised for preliminary research in the laboratory. Nickel ferrite (NiFe_2O_4), cobalt ferrite (CoFe_2O_4), magnetite (Fe_3O_4), and hematite (Fe_2O_3) were used to simulate corrosion products. The typical concentration of the input solution was 10 ppm.

Previous research papers have given experimental results of the PMF according to the flow rate and the particle size [2, 3, 9]. The present experimental results showed separation efficiency for the PMF of more than 80% for all particles under the following conditions: a 0.9 gallon/min (gpm) flow rate and 50 rotation/min (rpm) rotating velocity of the magnet assembly. The experimental results also reflect that the hydrodynamic drag force and the gravitational force, and not the magnetic force, are the major forces influencing very small particles. Some differences in separation efficiency are attributed to the disparities in the magnetic susceptibility of particular particles, especially hematite. As expected, the filter performance depends mainly on the flow rate and the particle size. The experimental results show that the PMF separation efficiency increases for a lower flow rate of the fluid stream or for a larger particle size. In particular, the PMF separation efficiency is over 90% for particles larger than $5\ \mu\text{m}$ with a 0.9 gpm flow rate and a 50 rpm rotating velocity of the magnet assembly. Thus, it is necessary to design a cohesive device that uses an electromagnetic field to flocculate particles into larger aggregates of about $5\ \mu\text{m}$ in diameter. In addition, the rotating velocity of the magnet assemblies could also be a parameter affecting the efficiency of the magnetic filter; however, this variable showed little influence

on the separation efficiency and its effect is not constant.

5. Electromagnetic Flocculation

5.1. Design for a Practical Cohesive Device with Electromagnetic Filtration

Electromagnetic filtration technology has reached a level of development in which filters can be designed to conduct their intended performance with predictable efficiency. The cost of an electromagnetic filter (EMF) installation is comparable to the cost of installing other major water treatment equipment. Operation and maintenance of the device will impose no unusual constraints on overall plant operations. A major advantage of an EMF is that it can operate in circuits located in higher temperatures than the conventional means of feedwater filtration [1]. In addition, the EMF offers more design flexibility for the filter elements to achieve the required efficiency related to the number of magnetic trapping sites per unit filter volume. In this study, the primary objective of 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.

The cohesive device designed in this study 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. The whole vessel is supported concentrically to an electromagnetic solenoid (Fig. 3). 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 that allows the electromagnetic solenoid to be cooled directly by oil that surrounds it, filling 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 have a uniform intensity throughout the volume concentric to the magnets and no agglomeration would occur. The ferromagnetic matrix assembly adopts the ball type of steel matrix, with a case 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 an on-off timer and a programming device, which automatically control the sequence of run-stop operations.

The principle behind the operation of the cohesive device is simple. The important component of the cohesive device is the magnetizable matrix made from ferromagnetic materials. If the applied magnetic field is uniform, there will be no net magnetic force on the particles. The presence of the matrix, with a positive magnetization inside the electromagnetic

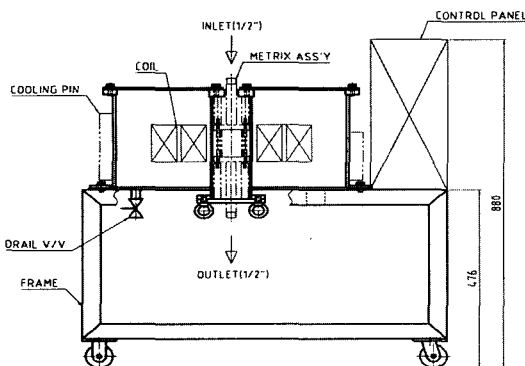


Fig. 3. Schematic Diagram of the Cohesive Device

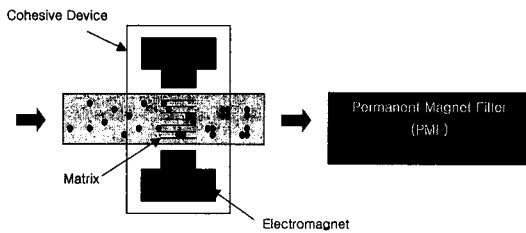


Fig. 4. Operation of the Cohesive Device

solenoid, causes an increase in field intensity due to the magnetic field added by the magnetization of this matrix. Thus, if an external magnetic field exists, this ferromagnetic matrix generates a strong magnetic field gradient in the vicinity of the ferromagnetic surface.

As corrosion particles in the fluid pass through the matrix assembly along the pipe line, they are magnetized by the background magnetic field gradient generated by electromagnetic solenoids 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 and, after this period, the timer turns off, and the magnetized corrosion aggregates flow out from the matrix assembly (Fig. 4). The magnitude of the field gradient is affected by the geometry of the ferromagnetic matrix and, in this experiment, the magnetization of the particles varied from approximately 2500 Gauss to 5000 Gauss.

5.2. Experimental Results

Several experiments were performed with changes to the following variables: the class of particles, the flow rates, the concentration of the input solution, the operation time of the

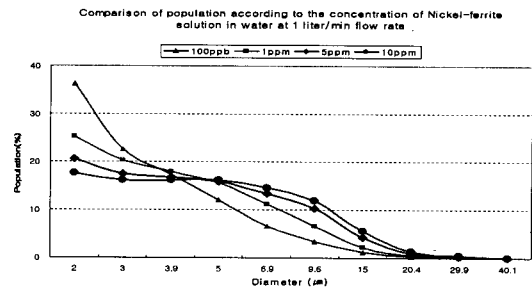


Fig. 5. Distribution of Particle Size According to the Concentration of Nickel Ferrite Solution

electromagnets, and the applied magnetic field. Experiments were normally carried out at room temperature and atmospheric pressure. The operating background fields ranged from 5000 Gauss to 2500 Gauss, and the typical applied magnetic field in this study was 5000 Gauss. Before the experiments, the original size of each particle was measured and most particles were found to be within a range of 2 to 3 μm in diameter. Most of the materials involved had amorphous structures while some metal oxides, such as hematite, were crystalline spheres.

Experiments with the cohesive device display a good performance when most aggregates of the particles are 4 to 5 μm in diameter. However, as the particle concentration is lowered, the size of aggregates becomes smaller: 4.93 μm at 10 ppm, 4.73 μm at 5 ppm, 3.64 μm at 1 ppm, and 3.38 μm at 100 ppb for nickel ferrite, with a 1 liter/min flow rate (Fig. 5). With a lower particle concentration, the number of particles aggregated by the magnetic field decreases; therefore, few aggregates form at very low concentrations, and the size of these aggregates is small compared to those found with higher particle concentrations. The operation time of the electromagnets also plays a key role in the flocculation of particles. Figure 6 illustrates the distribution of particle size for nickel ferrite for a 10 ppm concentration and a 1 liter/min flow rate. As the operation time is

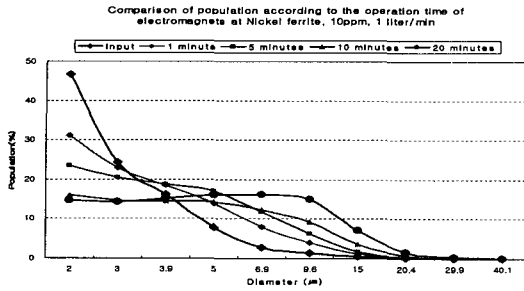


Fig. 6. Distribution of particle size according to the operation minute of electromagnets

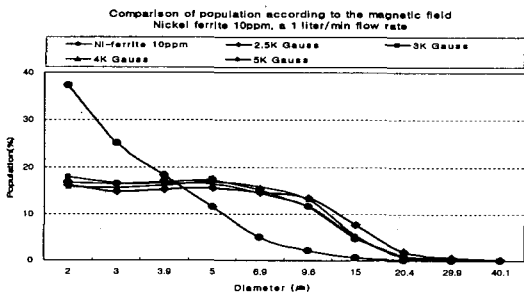
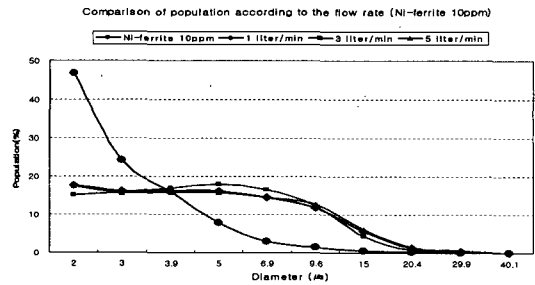
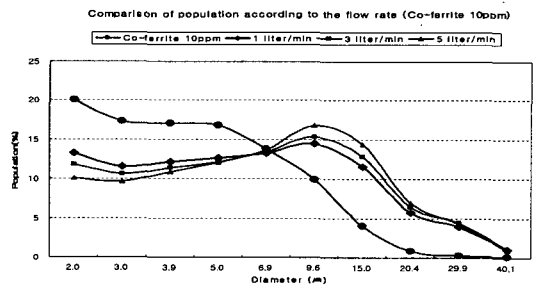


Fig. 7. Distribution of Particle Size According to the Applied Magnetic Field

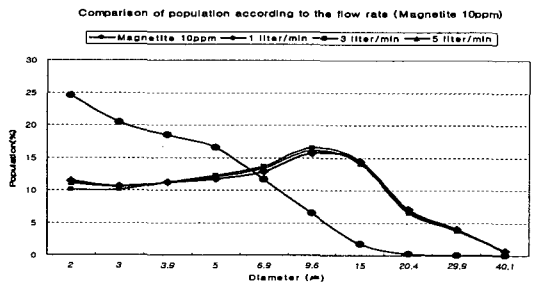
lengthened, the population of large particles is increased. The size of the aggregates varies with the operation time of the electromagnets: $3.42 \mu\text{m}$ after 1 minute, $3.87 \mu\text{m}$ after 5 minutes, $4.63 \mu\text{m}$ after 10 minutes, and $5.33 \mu\text{m}$ after 20 minutes of operation. Incidentally, the applied magnetic field and the flow rate do not contribute to the agglomeration of particles. Thus, most magnetic fields of 2500 Gauss and over are considered to have reached magnetization saturation. Figure 7 displays the population results of nickel ferrite, according to the applied magnetic field. The flow rate varies from 1 liter/min to 5 liter/min, but there is no difference in the particle size for the high and low flow rates. The distribution of particle size, according to the flow rates for each particle, is shown in Figure 8. At 10 ppm of concentration, nickel ferrite, which is known as the most dominant corrosion product, forms



(a) Nickel ferrite, 10ppm



b) Cobalt ferrite, 10ppm



c) Magnetite, 10ppm

Fig. 8. Distribution of Particle Size According to the Flow Rates

aggregates that are approximately $5 \mu\text{m}$ in diameter; magnetite and cobalt ferrites form aggregates that are about $8 \mu\text{m}$, due to the high magnetic susceptibility of these materials.

6. Discussion

As expected, experiments showed that the proposed cohesive device that uses an electromagnetic field flocculated particles into the

intended particle sizes relatively well. During subsequent normal operation in a nuclear power plant, the concentration of the corrosion products in the primary coolant system is approximately 100 ppb, rising to approximately 20 ppm during downtime. Therefore, in experiments conducted with the proposed cohesive device, the concentrations of the input solution were varied and included a concentration of 100 ppb in order to reflect the typical concentration of corrosion products in a real nuclear power plant. The experimental data in Figure 5 shows the flocculation of particles at 100 ppb, which is a very low concentration of particles. However, performing the experiment with such a low concentration is difficult since many factors, particularly the residual impurity in the fluid channel, can result in incorrect values. In this study, several experiments were carried out to obtain reasonable data values; incorrect values were eliminated.

As stated above, the typical applied magnetic field in this study is 5000 Gauss, which is high enough to magnetize the corrosion particles and to flocculate them into agglomerations. However, since this magnetic field is relatively strong, magnetism occasionally remains in the matrix after the power supply timer of electromagnets is shut off. This magnetism sometimes interrupts the flow of corrosion particles from out of the matrix assembly. These residual aggregates become obstacles to the flocculation of particles and should be cleaned before and after operation of the cohesive device.

7. Conclusions

This study involved the design and manufacture of a magnetic filter with permanent magnets to reduce the risk of occupational radiation exposure (ORE) and to help meet increasingly demanding

safety requirements in nuclear power plants. Previous research papers [2, 3] have demonstrated that the general removal efficiency of permanent magnet filters (PMF) is greater than 80% for various corrosion products. Preliminary experimental results showed that the removal efficiency was higher if the flow rate of the 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 removal efficiency in this study. The PMF was found to achieve a high removal efficiency of over 90% for particles with diameters of 4 μm and over.

There is high demand for a means to increase the size of corrosion product particles so that the removal of corrosion products can be performed more efficiently. This task was performed relatively well by the device developed in this study, a cohesive device that uses an electromagnetic field. After passing through the cohesive device, most corrosion particles within a range of 2 to 3 μm in diameter were aggregated into particles of 4 to 8 μm . Experiments with magnetite and cobalt ferrite displayed outstanding results as the average size of aggregates was approximately 8 μm in diameter due to the high magnetic susceptibility of those materials. The flow rate did not play an important role in the agglomeration of particles, and there was no difference in the particle sizes of high and low flow rates. The applied magnetic field also did not influence particle sizes, and it was considered that a magnetic field of over 2500 Gauss reached magnetization saturation for corrosion particles. The flocculation depended mainly on the particle concentration and on the operation time of the electromagnets, although some influence was attributed to the original disparities in the magnetic susceptibility of the particles involved. With a

higher particle concentration and a longer operation time, there is an increase in the number and the size of the aggregates.

Therefore, if the cohesive device devised in this study is adopted and positioned to flocculate particles before they enter the PMF, then the removal efficiency of the PMF for very fine corrosion products can be increased from 80% to over 90%, under various flow rates and concentrations.

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