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A Study on the Method with Electromagnetic Field for Improving the Removal Efficiency of Radioactive Corrosion Products

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Abstract

A number of different solid particles found in nuclear reactor coolant have been identified as one of the major sources of the occupational radiation exposure in a nuclear power plant. These corrosion product deposits at the fuel clad surface have also been known to contribute to the onset of Axial Offset Anomaly, AOA, considered as a barrier to optimal reactor power. In general, the corrosion products have extremely low solubility and display the strong magnetic properties. It is therefore performed the conceptual design to devise the filter which removes the corrosion products by magnetic field generated by an arrangement of permanent and electric magnets. Experiments using permanent magnets display the good performance of filtering corrosion products and indicate that the removal efficiency of magnetic filter is more than 90% for above 5 micrometers particles. Thus, this study is focusing on the enlarging the particle size using an Electromagnetic Filter (EMF) as a cohesive device before the Permanent Magnet Filter (PMF), the magnetic filter using permanent magnets, for improving the removal efficiency of radioactive corrosion products.

Introduction

The materials of nuclear power plant structure that usually come into contact with the primary coolant streams of power systems are metal alloys containing as major components iron, nickel, copper, chromium, cobalt, and so on. All these materials react chemically with water and dissolved oxygen to form oxides known as corrosion products. The corrosion products have comparatively low solubility and show varied magnetic properties. An important class of corrosion products is known as the ferrites which are derivatives of magnetite or metallic constituent. 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]. This is also a representative of the circulating particulate corrosion product in a nuclear power plant. The corrosion products are transported by the coolant stream and deposited throughout the power systems where they may cause troubles for optimal nuclear power plant operation, such as Axial Offset Anomaly, AOA, considered as a barrier to optimal reactor power. They are also recognized the major source for the occupational radiation exposure to nuclear power plant workers. There have been therefore significant demands for the application of new technology to control and restrain the corrosion products effectively and reasonably.

There are several ways in use including the improvement of the coolant purification system, the operation condition at high pH, the adoption of materials with low levels of cobalt in the primary coolant system, and decontamination of the primary system more frequently to reduce the radiation levels around the primary coolant system in a nuclear power plant. For instance, elevated pH is known to reduce ex-core corrosion release, transport, and deposition of corrosion products on nuclear fuel. However, the increased boron requirements for modern core designs demand significant increase in lithium concentration to attain desired constant-elevated pH conditions. Therefore, a careful engineering evaluation of the possible impact of these lithium increases on corrosion susceptibility of both fuel and structural components is necessary [2]. The numerated ways above place more weight on the prevention against corrosion of material and the improvement of thermo-hydraulic efficiency than the elimination and removal of corrosion products in the primary coolant system basically.

Contrary to the conventional ways mentioned above, it is performed the conceptual design to develop the filter which removes the corrosion products by magnetic field that is generated by an arrangement of permanent and electric magnets as an active method to control the primary coolant waste. In this study, the new type of magnetic filter devised with permanent magnets generates strong magnetic field and shows the good result of removal efficiency for corrosion products more than 80% [3]. However, it is necessary to use an electromagnetic filter, which causes the corrosion particles to flocculate into larger aggregates about 5 micrometers, as a cohesive device before the permanent magnet filter for improving the removal efficiency more than 90%. This paper focuses on the several results of experiments with electromagnetic filter resulting in agglomeration of corrosion particles.

Theory of Magnetization

In order to design the practical permanent and electromagnetic filter, it is essential to understand a physics of magnetization and 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, the nuclear power plant corrosion products display a comparatively strong ferromagnetism.

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

$$B = \mu H$$

and

$$M = \chi_m H$$

where μ and χ_m are the permeability and susceptibility of the material, respectively [4]. 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 diamagnetic, and the magnetic induction is weakened by the presence of the material [5]. Contrary to the magnetism remarked 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 the combined forces tending to keep the particle in suspension. In the idealized one dimensional isotropic case, the forces experienced by a particle in a magnetic field can be described as

$$\left|F_m\right|_x = \frac{1}{2}\mu_o V M \frac{dH}{dx}$$

where $|\boldsymbol{F}_m|_x$ is the magnetic force for **x** coordinates, $\boldsymbol{\mu}_o$ is magnetic permeability at free space, $\boldsymbol{V}$ is the particle volume, and $\boldsymbol{M}$ is the magnetization. The product, $\boldsymbol{VM}$, is the magnitude of the particle acted on by the field gradient, $\boldsymbol{dH/dx}$. The hydrodynamic drag force for particles in the present case can be represented by Stoke’s Law [1]

$$F_d = 6\pi\eta_f r_p u$$

where $\boldsymbol{F}_d$ is the drag force, $\boldsymbol{\eta}_f$ is the viscosity of the medium, $\boldsymbol{u}$ is the viscosity of particle relative to the fluid stream, and $\boldsymbol{r_p}$ is the radius of particle. Then finally the motion of corrosion particles is governed by [3]

$$m\frac{dV}{dt} = -F_d + F_m$$

and

$$\frac{dV}{dt} + \frac{9\mu}{2r_p^2\rho_p}V = \frac{\mu_o \chi H}{2\rho_p}\frac{dH}{dx}$$

where χ is magnetic susceptibility, $\boldsymbol{\rho_p}$ is the particle density. The above equation indicates that the relative magnitudes of the components of the two 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.

Principles of Magnetic Separation System

1) Open Gradient Magnetic Separation (OGMS)

Open gradient magnetic separation achieves the separation goals by deflecting the magnetic particles from the main stream. Contrary to High Gradient Magnetic Separation (HGMS), OGMS has no ferromagnetic matrix for generating the strong magnetic field, but it is possible to apply for a continuous process since no particle accumulation takes place inside the magnetic separator due to two or more outlets (Figure 1) [6]. In OGMS, the magnetic force acts in antiparallel direction to flow within a certain area. If the magnetic force acting on a particle exceeds the counteracting hydrodynamic resistance forces, the particle is retained inside the area of the magnetic field. The area of high magnetic forces acts on these particles like a barrier that cannot be overcome. During initial operation, magnetic particles increasingly accumulate in the magnetic separator. In order to achieve a continuous operation of the magnetic separator, it is therefore necessary that the accumulated particles be removed from the magnetic field area via a concentrate discharge. This technology is applied for magnetic separation in metal processing industry and municipal sewage treatment plants.

2) High Gradient Magnetic Separation (HGMS)

High Gradient Magnetic Separation achieves the separation goals by collecting the magnetic particles in ferromagnetic matrices. The basic principle of HGMS is simple and similar to that of conventional deep-bed filters. The main difference lies in the direct attraction between particles and separation matrix, which is achieved by the magnetic force [3]. A cylinder filled with a magnetizable separation matrix is introduced into an external magnetic field. The matrix is composed of a loose packing of rough steel wool and its wires bundle the external magnetic field in their surroundings and produce areas on the surface, which strongly attract the particles to be separated (Figure 1). During operation, the sewage to be cleaned flows through the magnetic separator at high velocity. Finally, the suspended particles in the sewage stream are removed almost completely. This technology has been employed industrially for the separation of finest metal particles from waste water of steel and metallurgical industries such as minerals beneficiation and waste treatment in steam heating plants.

Permanent Magnetic Filtration

1) Design for the Practical Permanent Magnetic Filter (PMF)

Permanent magnets have several advantages over conventional electromagnets. The fundamental advantage is that they can provide a relatively strong magnetic field over an extended spatial region for an indefinite period of time 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 pattern owing to the rapid growth of manufacturing technique. They are also relatively inexpensive on a per unit basis depending on the material used. It is therefore devised the Permanent Magnet Filter (PMF), which can separate the metallic particles from the main stream, applied to the principle of OGMS, to remove the corrosion products in the coolant stream.

The PMF devised in this study is composed of two main parts: a separator and a driving motor. The separator consists of inner and outer assemblies, fluid channel, and a container surrounding the outer magnet assembly. The fluid channel is located between the inner and outer permanent assemblies. The rotation of the permanent magnet assemblies and the shifted arrangement of permanent magnets generate the alternating magnetic field in the fluid channel. In order to maximize the magnetic field, magnetic circuits are constructed with irons and each magnet faces an opposite polar magnet respectively. As corrosion particles in the fluid pass through the channel between the magnet assemblies, they are easily magnetized due to the strong magnetic field and then move in the direction of 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 separated from the fluid stream at the boundary wall of the vessel. Figure 2 illustrates the design of the PMF and its principle of operation.

2) Results of Experiments with the Permanent Magnetic Filter (PMF)

It was conducted several experiments with varying the various parameters including the class of particles, flow rates, the rotating velocities of magnet assembly, and the particle concentration. Experiments were usually carried out in the condition of room temperature and atmospheric pressure since, in this study, the PMF applied to the open-loop type was devised for a 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 as corrosion products simulants. The former two simulants are made in Kojundo Chemical Laboratory Co. Ltd, Japan, and the latter two simulants were supplied by Aldrich Chemical Company, USA.

Experiments with the PMF indicate relatively good separation efficiency more than 80% for all particles except hematite since it is an antiferromagnetic material with weak magnetic property. Some differences in separation efficiency are attributed to the disparities in magnetic susceptibility of those particles, especially hematite. As expected, the filter performance depends on mainly the flow rate and particle size. The experiment results show that the PMF can achieve quite high separation efficiency as the flow rate of fluid stream is lower and the particle size is bigger [3, 7]. Separation efficiency of the PMF is more than 90% for above 5 micrometers particles. Thus, it is necessary to design a cohesive device using electromagnetic field to flocculate particles into larger aggregates about 5 micrometers. In addition, the rotating velocity of magnet assemblies also could be a parameter to effect on the efficiency of magnetic filter; however, there was a little influence on separation efficiency and its effect is not steady and constant.

Electromagnetic Filtration

1) Design for the Practical Cohesive Device with Electromagnetic filtration

Electromagnetic filtration technology has reached the level of development where 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 other major water treatment equipment. Operation and maintenance of the device will impose no unusual constraints on overall plant operations. A major advantage is that the EMF can be located at higher temperature positions in the circuits than more conventional means of feedwater filtration [1]. In addition, the EMF offers more flexibility in design of 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 this cohesive device is to cause the very fine suspended magnetic particles to cohere as larger aggregates which could then easily be separated in the PMF.

The cohesive device designed in this study consists of three main parts: a vessel with inlet and outlet pipe connections, fitted inside with a solenoid to generate the background magnetic field and a structure to cool the magnet coils, the magnetizable matrix assembly, and the control panel. The whole is supported concentrically to a system of an electromagnetic solenoid (Figure 3). The vessel is made of nonmagnetic stainless steel to avoid short-circuiting the magnetic field. Since the magnetizing current also generates considerable heat and additional heat derived from the vessel itself during operation, the vessel furnished with cooling system; the electromagnetic solenoid is cooled directly by surrounding oil 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. Basically, without the matrix or with nonferromagnetic matrix, the magnetic field would be uniform in intensity throughout the volume concentric to the magnets, and no agglomeration effect would obtain. The matrix assembly adopts the ball type of fine steel matrix and its case is made of nonmetal 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. It is also equipped with on-off timer, the programming device, which automatically controls the sequence of run-stop operations.

The principle of this cohesive device operation is simple. As corrosion particles in the fluid pass through the matrix assembly along the pipe line, they are magnetized by the background magnetic field generated by electromagnetic solenoids and then small magnetized particles flocculate into lager aggregates while the power supply timer is on. After a specific interval of power supply, the timer turns off and the magnetized corrosion aggregates flow out from the matrix assembly (Figure 4).

2) Results of Experiments with the Cohesive Device

It was performed several experiments according to the variables: the class of particles, flow rates, the particle concentration, and the power-on minutes. Experiments were normally carried out in the same condition of experiments with the PMF, which was of room temperature and atmospheric pressure. The operating background fields range from 5000 Gauss to 2500 Gauss, although lower fields have been investigated, and typical applied magnetic field in this study is 5000 Gauss. Before the experiments, the size of each particle was measured and it was found that most particles were within the range of 2 or 3㎛ size.

Experiments with the cohesive device display comparatively good performance that most aggregates of particles are within the scope of 4 or 5㎛ size. At 10ppm of particle concentration, most particles indicated that their aggregates were approximately 5 micrometers except magnetite which showed 8 micrometers due to the high magnetic susceptibility of material. However, as the particle concentration was lower, the size of aggregates was smaller, which was 5.04㎛ at 10ppm, 4.83㎛ at 5ppm, 3.66㎛ at 1ppm, and 3.37㎛ at 100ppb for nickel ferrite. By the way, the flow rate did not contribute to the agglomeration of particles and there was no difference of particle size between high and low flow rates. Contrary to the flow rate, the power-on minutes played a key role in the flocculation of particles. The results indicate that the size of aggregates are 3.42㎛ after 1 minute, 3.90㎛ after 5 minutes, 4.72㎛ minutes after 10 minutes, and 5.47㎛ after 20 minutes of power-on. These results are plotted in Figure 5 and 6.

Discussion

As expected, experiments with the cohesive device using electromagnetic field show relatively good performance of the flocculation of particles with intended particles size. During subsequent normal operation in a nuclear power plant, the concentration of corrosion products at the primary coolant system is approximately 100ppb and it goes up about 20ppm during the downtime. Even though there was an effect on flocculation of particles at very low concentration of particle, such as 100ppb, it was hardly performed the experiment in such a low concentration since there were so many factors resulting in incorrect values. In this study, it was carried out the several experiments to get reasonable values and the incorrect values and errors were eliminated.

As stated above, the applied magnetic field in this study is 5000 Gauss and this is much enough to magnetize the corrosion particles and to flocculate into agglomeration. However, since this magnetic field is relatively strong, there is still magnetism in the matrix after power supply timer is off and it sometimes interrupts the corrosion particles to flow out from the matrix assembly. These residual aggregates also are obstacles to flocculation of particles and it should be cleaned before and after the operation of cohesive device.

Conclusion

According to the increasing demand for higher safety requirements in a nuclear power plant, the magnetic filter with an arrangement of permanent and electric magnets is designed and manufactured to reduce the occupational radiation exposure (ORE). The ability of the permanent magnet filter (PMF) to remove the corrosion products with more than 80% efficiency was demonstrated in preliminary experiments with the PMF. The removal efficiency is higher as the flow rate of fluid stream is lower and particle size is bigger. Although there is a little influence of the rotating velocity of magnet assemblies on the removal efficiency, the flow rate and the particle size are main parameters determining the removal efficiency. In addition, the removal efficiency of the PMF is more than 90% for above 5 micrometers particles.

There is a strong need for improving the removal efficiency of corrosion products through the way increasing the particle size. The cohesive device using electromagnetic field, thus, is developed and its general performance was relatively good. After passing through the cohesive device, most corrosion particles within the range of 2 or 3㎛ size become the aggregates within the scope of 4 or 5㎛ size. As unexpected, the flow rate did not play an important role in agglomeration of particles and flocculation mainly depended on the particle concentration and the power-on minutes of device. As the particle concentration is higher and the power-on minutes is longer, there are more aggregates and its size is bigger.

It is consequently expected that the combination of magnetic filter including the PMF and the cohesive device will be an effective way for the removal of corrosion products with more than 90% efficiency.

Acknowledgement

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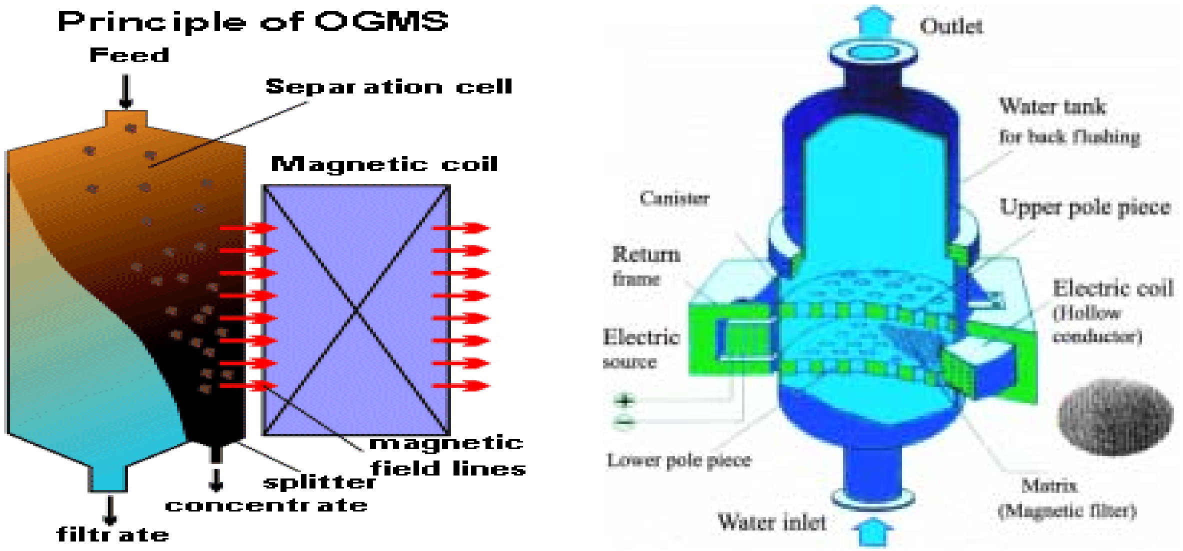


Figure 1 - Principle of OGMS and HGMS

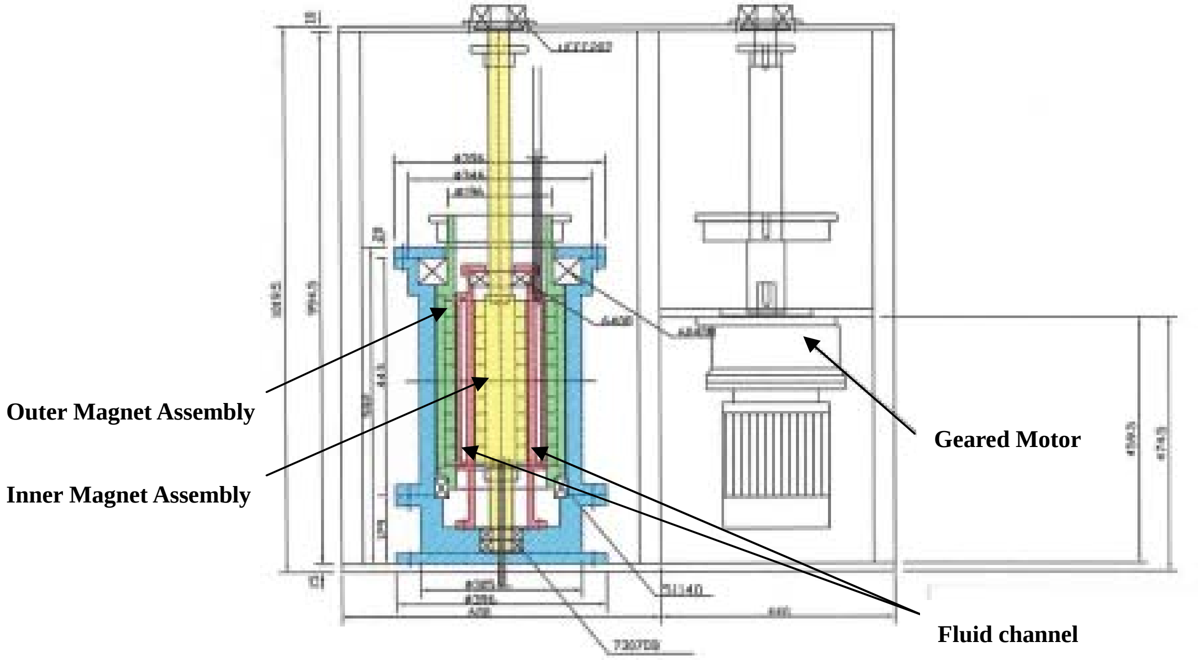


Figure 2 – Design of the Permanent Magnetic Filter (PMF)

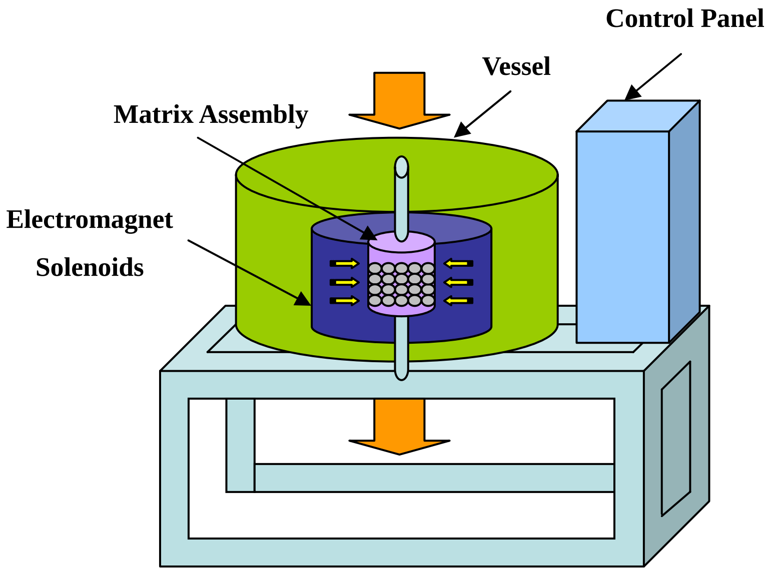


Figure 3 - Schematic Diagram of the Cohesive Device

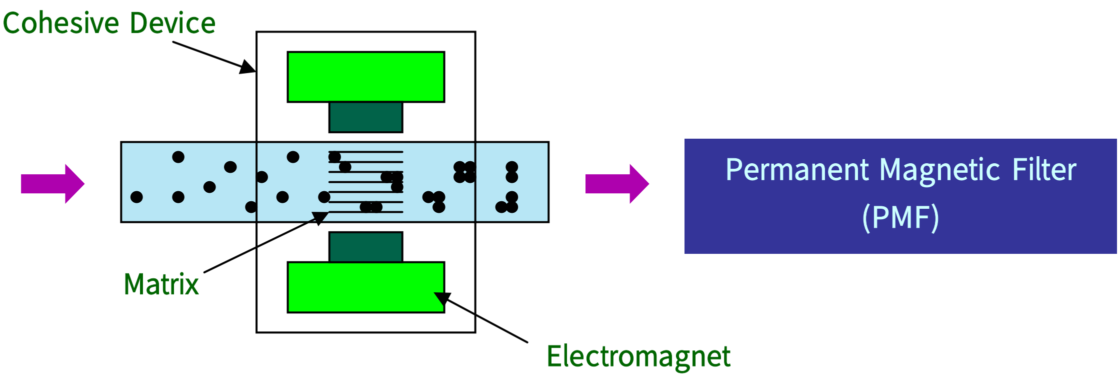
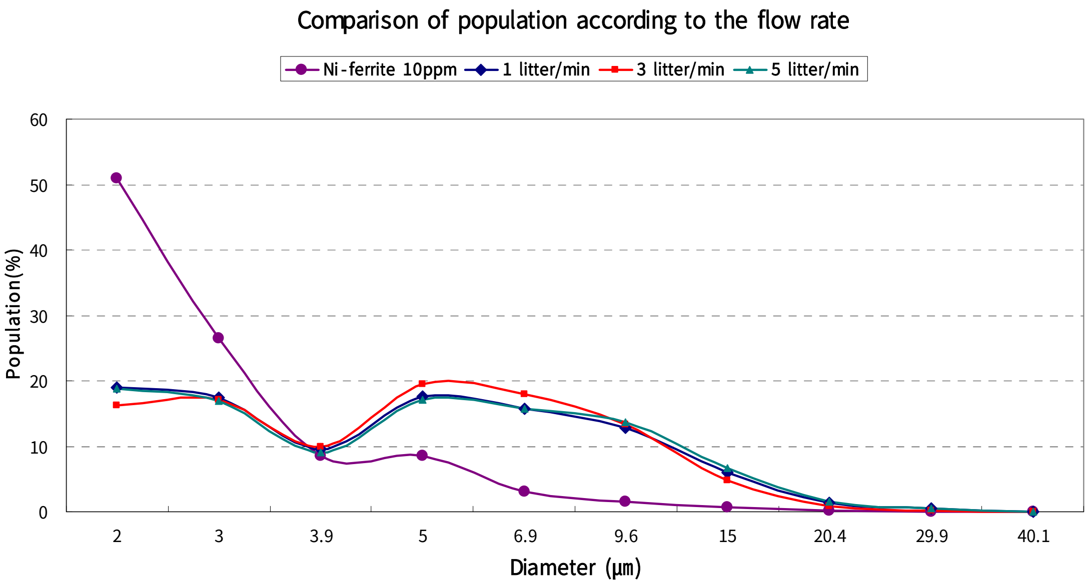
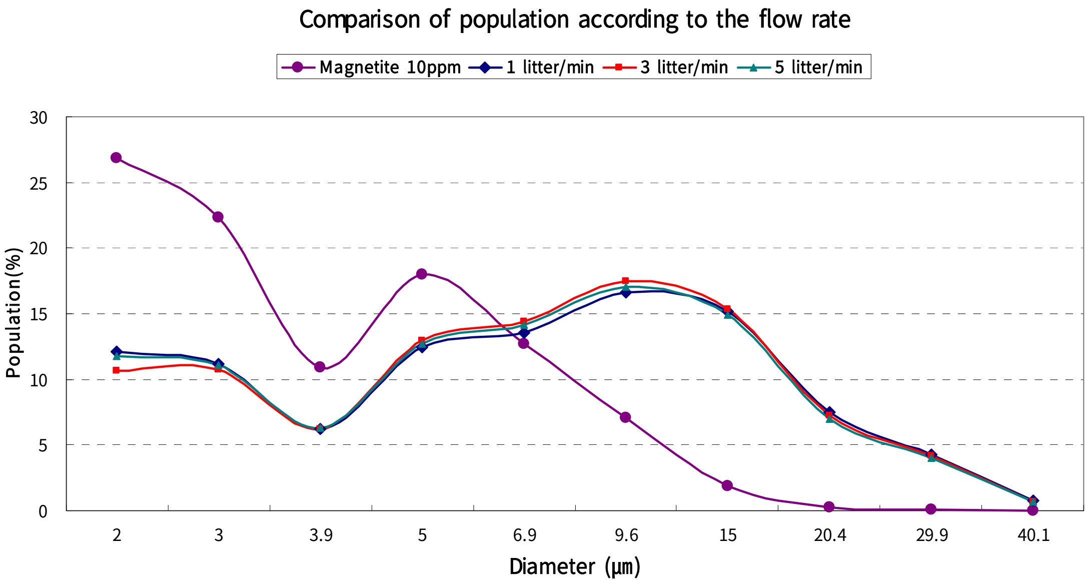


Figure 4 - Operation of the Cohesive Device

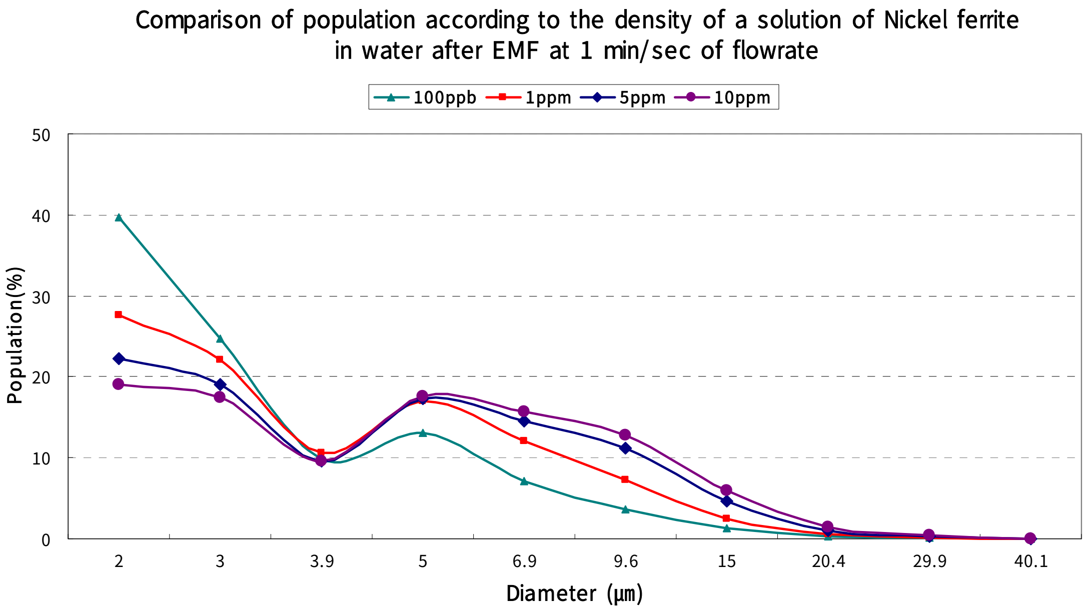


Nickel ferrite 10ppm



Magnetite 10ppm

Figure 5 – Results of Experiments with the Cohesive Device



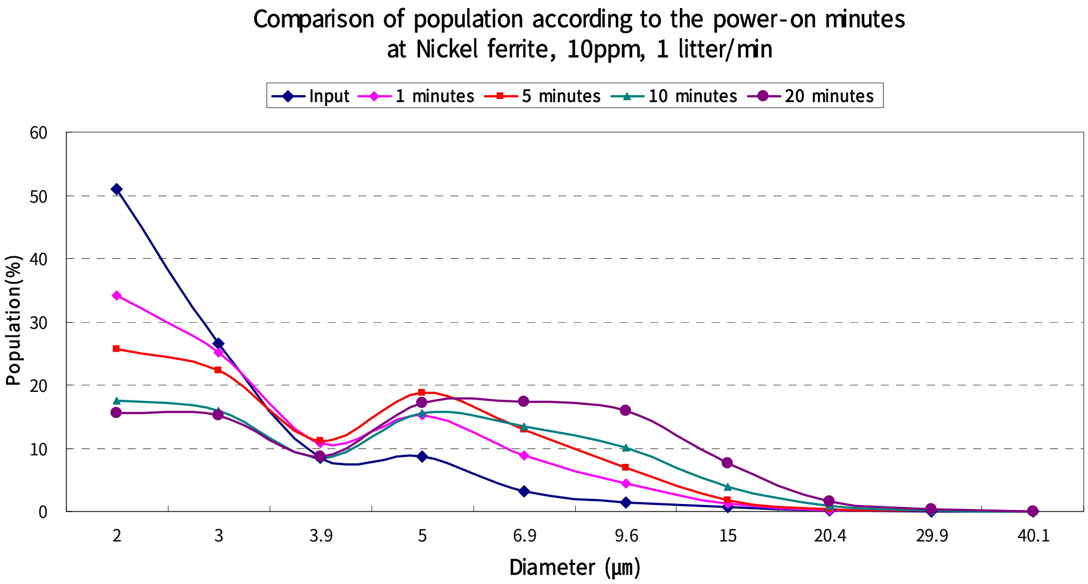


Figure 6 – Results of Experiments with the Cohesive Device