Development of Simulated CRUD Production Method for Abrasive Decontamination Performance Evaluation

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1. Introduction

A CRUD (Chalk River Unified Deposits) formed on the metal surface of the primary side of a nuclear power plant is a material formed by depositing metal corrosion products and other particles on the metal surface during the operation of the power plant, and it is known that CRUD is mainly formed on the surface of the primary system metal under the operating conditions of a pressurized water reactor (PWR). The chemical component of the oxide deposited on the inner surface of the device of the primary system is mainly known as nickel-ferite. The oxide grown on the surface of the base material is mainly an oxide close to the base material component, and the chromium component gradually increases as it approaches the inside of the base material. The main reason for the increase in the radiation dose rate inside the primary system of nuclear power plants is the accumulation of radionuclides within the corrosion products formed on the surface of the inner material of the system. These radionuclides exist not only in the deposited oxide but also in the growth oxide layer inside the PWR, and radionuclides in the deposited oxide are formed on the surface of nuclear fuel and moved to the system surface outside the core by various mass transfer mechanisms to be deposited. Radionuclides such as ^{55}Fe and ^{60}Co are distributed in the corrosion products.

Decontamination generally means removing these oxide layer and radioactive substances accumulated in the oxide layer together with the oxide layer. Due to the problem that it is difficult to directly experiment with radioactive CRUD in the laboratory, research has been conducted through simulation of CRUD generated in the nuclear power plant. The technique used to manufacture the simulated oxide layer specimen was mainly an autoclave and coating technique, and the autoclave method takes a lot of time and cost to manufacture the simulated oxide layer specimen with a large area. The coating technique was mainly used to manufacture a magnetite oxide layer, which is a simulated oxide layer on the material surface of the secondary side of the nuclear power plant. In this study, the study was conducted to verify the decontamination performance of the abrasive blasting facility by developing a method of fabricating a primary oxide layer simulated specimen that can be applied to the decontamination performance evaluation of the abrasive blasting facility.

2. Methods and Results

In this section, the main characteristics of CRUD required for evaluating abrasive blasting performance and the results of the development of a simulated CRUD production method to be developed by reflecting these characteristics were described.

2.1 Characteristics of CRUD

The CRUD generated at the primary side of the nuclear power plant appears differently depending on the corrosion environment at the primary side of the nuclear power plant. In the case of a pressurized light water reactor (PWR) and a heavy water reactor, there is a difference in the composition of the crud. The primary CRUD on the nuclear power plant consists of two layers, a deposited layer and a grown oxide layer, and in the case of a PWR, the composition of the outermost deposited layer of the CRUD is composed of magnetite and nickel ferrite. The components of the inner layer of the CRUD in PWR are known as $FeCr_2O_4$, $NiCr_2O_4$, and Cr_2O_3. In the case of an oxide layer, depending on the composition of the base material, there is a slight difference in the oxide layer characteristics.

Various research literatures report on the oxide layer composition, physical structure, and crud thickness of stainless and alloy steel. The CRUD oxide layer is greatly influenced by the base material, coolant, chemical substances and operating conditions. An oxide layer suitable for the abrasive blasting performance test should be produced including the following items.

Characteristics of CRUD
- Compositions: CRUD mainly consists of metal oxides : Key compositionss : Fe_3O_4, NiO, Cr_2O_3
- Physical properties : Density: relatively low but firmly attached to metal surfaces.

The main characteristics of the inner and outer layers of the CRUD are summarized as follows.
- inner layer

- High chromium content and low porosity
- The closer it is to the base metal, the closer it is to Cr_2O_3
- Thickness : ~5 ㎛

• outer layer
- It exhibits relatively high porosity compared to the inner layer, and nickel ferrite
- Thickness : ~10 ㎛

Table 1: Properties of Cruds on Stainless Steel

	Density[g/cm³]	Hardness[Hv]	Porosity[%]
Fe_3O_4	~ 5.17	400	~ 40
$NiFe_2O_4$	~ 5.38	600	
Cr_2O_3	~5.22	1100	~10

2.2 Production Techniques of Simulated CRUD

In many studies, studies have been conducted on the formation CRUD on the inner surface of pipes and fuel surfaces of nuclear power plants' primary system. Many technologies for artificially generating CRUD have been studied, and the main technologies commonly used are as follows.

2.2.1. Autoclave

In general, an autoclave experimental facility is a Loop system or a closed system and consists of a reactor in a high temperature/high pressure environment and an auxiliary system capable of simulating a power plant's hydro-chemical environment so that the primary operating environment of a nuclear power plant can be simulated. Figure 1 is the autoclave experimental facility of the Korea Atomic Energy Research Institute. In the case of simulation of the primary side CRUD, a method of generating a CRUD in an autoclave environment under the same conditions as the primary system can be used, but in this case, facility installation cost and a long time sample manufacturing time are required. In addition, there is a disadvantage that it is difficult to make a large area of simulated CRUD specimen, and there is a limitation in making the thickness of the simulated CRUD thickness.

2.2.2. Thermal Spray Coating Technology

The simulated CRUD production technology using the thermal spray coating technique is used to artificially simulate and reproduce generated in actual operating environments in nuclear power plant, power generation facilities, aerospace, semiconductors, etc. It can be used to study corrosion, oxidation, and abrasion that can occur during actual operation or to evaluate surface behavior. This technology forms a coating layer by melting and heating powders such as metals, alloys, and ceramics by a high-temperature gas flow (plasma, HVOF, arc, etc.) and then spraying and stacking them on the substrate at high speed.

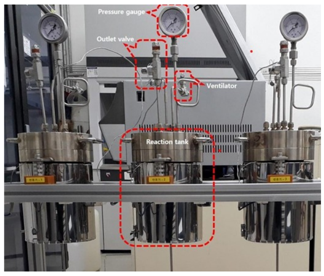


Fig. 1. KAERI autoclave facility

2.3 Development of a Production Method for Simulated CRUD

2.3.1. Development of a Production Method for simulated CRUD

In order to manufacture a simulated CRUD specimen through physical coating, the chromium component of the inner layer and the magnetite (or nickel ferrite) component of the outer layer are required. The thickness of the simulated CRUD must be at least 25µm required by the RFP of the task, and to produce a simulated CRUD thickness satisfying it, we produced three kinds of simulated CRUD specimens consisting of two layers of chromium oxide (~10µm) and magnetite (or nickel ferrite, ~15µm), a single chromium oxide layer (~30µm), and a single magnetite (or nickel ferrite, ~30µm) layer.

Simulated CRUD for evaluating the abrasive blasting performance was produced by applying the atmospheric pressure plasma spraying (APS) method. Plasma spraying is a method of fusion of powder-type materials using high temperature generated by plasma gas as a heat source among various spraying methods, and the schematic diagram of plasma spraying is shown in Figure 2. Spray powder is injected into the flame formed by plasma gas to collide with and fuse the base material.

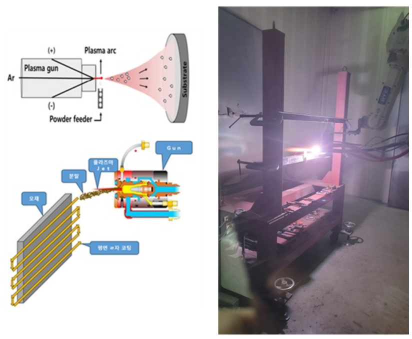


Fig. 2. Plasma thermal spray coating

(1) Selection of coating materials

In the case of thermal spraying for application to atmospheric pressure plasma thermal spraying, it is important to have particles of an appropriate size capable of securing fluidity. In the case of thermal spraying powder having a small dimension of less than 1μm, there is a problem that coating by plasma flame is not performed well. Therefore, in order to generate an simulated CRUD having an appropriate thickness, it must have particles of an appropriate size capable of having sufficient fluidity in plasma gas. In the case of magnetite used for the coating, 1-2μm particles were sintered into a size of 50μm to produce a spherical aggregate, and plasma spray coating was performed.

① Fe_3O_4 powder

Fine powder is manufactured into spherical powder of appropriate size using ceramic powder manufacturing process (Agglomeration and sintered)

② Cr_2O_3 powder

Purchased powder size 15-45μm

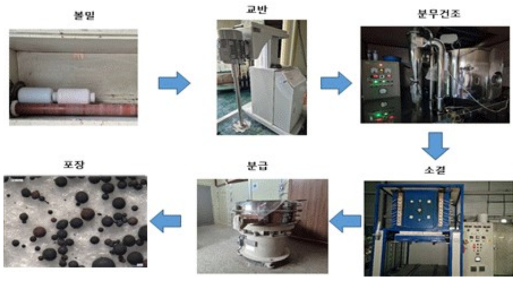


Fig. 3. Fe_3O_4 powder manufacturing process

(2) APS coating process optimization

By adjusting the APS process variables, the optimal process conditions that can satisfy the target thickness and porosity of the simulated CRUD were derived.

- Plasma coating parameters: Electricity output [KW], Current [A], coating distance, primary gas flow rate, powder particle size, number of coatings

2.3.2. Analysis of simulated CRUD

In order to examine the validity of the simulated CRUD applied with the developed thermal spray coating, the CRUD thickness and porosity were measured using an optical microscope as shown in Figure 4, and it was confirmed that a CRUD thickness, 20 to 35μm was formed as shown in Table 2. In addition, as a result of the porosity measurement, it was confirmed that the porosity of Cr_2O_3 was 19% on average, the porosity of Fe_3O_4 was 27% on average.

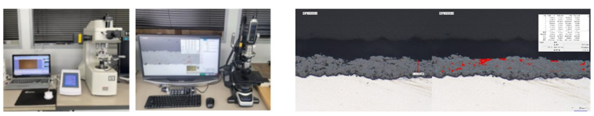


Fig. 4. Analysis equipment and Simulated CRUD

Table 2 : Properties of CRUD on Stainless Steel

CRUD layer	Thickness[㎛]	Porosity[%]
Cr_2O_3 (single layer)	28	19
Fe_3O_4 (single layer)	27	27
Cr_2O_3 / Fe_3O_4 (double layer)	35	26

Figure 5 is the result of analyzing the thickness of the simulated CRUD specimen using SEM at KAERI, and Figure 6 is the analysis of the composition of the Fe_3O_4 simulated CRUD using EDS. It was confirmed that the thickness and composition of the simulated CRUD are similar to the CRUD formed on the primary side of the actual nuclear power plant.

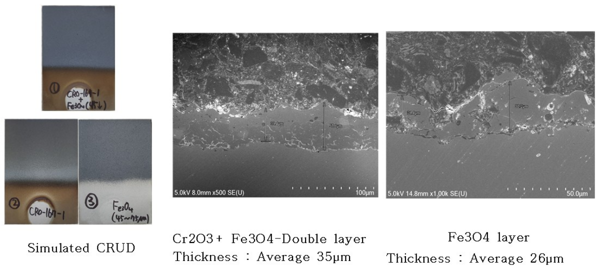
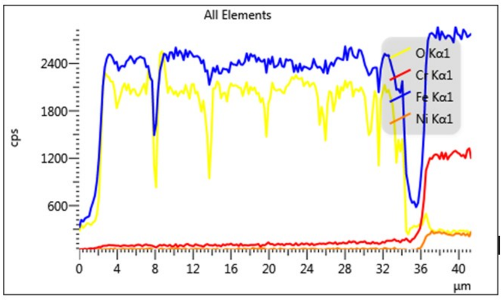


Fig. 5. Thickness of the simulated CRUD



Statistic	O	Cr	Fe	Ni
Max	45.78	2.19	51.27	0.76
Min	45.78	2.19	51.27	0.76
Average	45.78	2.19	51.27	0.76

Fig. 6. Composition of the Fe_3O_4 simulated CRUD

3. Conclusions

In this study, a method of manufacturing the simulated CRUD required for decontamination research to a thickness level of 25μm or more, which is the thickness level of the target CRUD, was developed by applying the atmospheric pressure plasma spraying technique. Through comparison with the simulated CRUD specimen prepared through an autoclave, the adequacy of manufacturing a simulated CRUD specimen using the atmospheric pressure plasma spraying technique was confirmed.

(1) By applying the atmospheric pressure plasma spraying technique, a method of manufacturing by coating the chromium oxide inner layer with an average thickness of 19μm and the magnetite of the outer layer with an average thickness of 10μm was established. In addition, a method of manufacturing the chromium oxide single layer with an average thickness of 28μm and the magnetite single layer with an average thickness of 29μm was established.

(2) The structural similarity between the simulated CRUD specimen manufactured through the atmospheric pressure plasma spraying technique and the autoclave manufactured simulated CRUD specimen was confirmed to be applicable to the abrasive blasting performance verification of abrasive blasting facility.

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