

Performance of Foam Decontamination for Fixed Radioactive Contamination

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1. Introduction

Decontamination is one of the radiation protections to minimize the worker's exposure during the work of maintenance and/or decommissioning of nuclear facilities. The foam decontamination process is well known as one of the chemical decontamination processes for removing radioactive particles on the surface, and also highly applicable to the surface contaminated with an oily constituents since the decontamination agent itself contains a surfactant. In addition, the foam decontamination process has advantages such as increasing easiness and safety in decontamination operation and generating a small amount of secondary waste, but has a disadvantage of relatively low decontamination efficiency. Particularly, when the foam decontamination process is used to remove fixed contaminants such as radioactive oxide films in which radionuclides are incorporated. Attempts have been made to improve the low decontamination performance of the foam decontamination process. One of them is to increase the contact time between the decontamination foams and the surface to be decontaminated. Another way to improve decontamination performance is to introduce a suitable chemical decontamination agent into the foam to remove the contaminants on the surface of the decontamination object.

In this study, we investigated the removal performance of NiFe_2O_4 and FeCr_2O_4 films formed on the surface of 304 stainless steel as fixed contamination using decontamination foams containing acidic and oxidative chemical decontamination agents. In addition, the performance of the decontamination foam was evaluated on radioactive specimens contaminated in the primary system of the nuclear power plant (NPP).

2. Materials and Methods

2.1 Preparation of Decontamination Foams

The decontamination foams used in this study were prepared by combining surfactants, silica nanoparticles and chemical reagents such as inorganic acids containing HNO_3 , HF and H_2SO_4 , and oxidizing agent containing Ce(IV) .

In order to evaluate the removal performance of the simulated specimens coated with oxide film and the decontamination efficiency of radioactive specimens drawn from the NPP, two kinds of decontamination

foams such as acidic foam (AF) and oxidative foam (OF) were prepared as summarized in Table 1.

Table I: Composition of Various Decontamination Foams

Foam Type	Chemical Composition	pH
AF-1	1 wt% EM100 ¹⁾ + 1 wt% M-5 ²⁾ + 2M HNO_3	≈ 0.7
AF-2	1 wt% EM100 + 0.5M HF + 0.5M HNO_3	≈ 1.2
AF-3	1 wt% EM100 + 0.2M HF	3 ³⁾
AF-4	1 wt% EM100 + 0.5M HF	3 ³⁾
OF-1	1 wt% EM100 + 1 wt% M-5 + 0.05M Ce(IV) + 0.5M H_2SO_4	< 0.2
OF-2	1 wt% EM100 + 1 wt% M-5 + 0.1M Ce(IV) + 0.5M H_2SO_4	< 0.2
OF-3	1 wt% EM100 + 1 wt% M-5 + 0.2M Ce(IV) + 0.5M H_2SO_4	< 0.2

1) Nonionic surfactant, ELOTANT Milcoside 100, C08-10 alkyl polyglucoside

2) CAB-O-SIL[®] M-5 Fumed Silica Nanoparticle

3) pH adjusted by adding H_2SO_4

2.2 Performance Tests with Decontamination Foams

Prior to the decontamination tests for radioactive specimen, the experiments on the removal of oxide from the specimens coated with simulated oxide film were carried out to verify the performance of decontamination foams on the dissolution of oxide film. To this end, two types of simulated oxide specimens coated with NiFe_2O_4 and FeCr_2O_4 were prepared. The oxide removal performance test was carried out in an experimental apparatus consisting of a decontamination foam generator and a foam filling column.

Based on the results of oxide removal test on the simulated specimens, the performance tests were carried out to verify the applicability of decontamination foams to the Inconel 690 sealing assembly used to block damaged heat transfer tube in steam generator of Gori Unit 3 NPP using acidic and oxidative decontamination foams.

3. Results and Discussion

3.1 Oxide Removal Performance

For the removal of NiFe_2O_4 film using various acidic decontamination foams, NiFe_2O_4 film was completely dissolved within 2 hours by strong acidic decontamination foam, AF-2 mixed with 0.5 M nitric acid and 0.5 M hydrofluoric acid, while only ca. 3% of the oxide film was dissolved during the same time period by strong acidic decontamination foam, AF-1 containing only 2 M nitric acid. Under the milder

conditions of acidic decontamination foam, NiFe_2O_4 film hardly dissolved in the acidic decontamination foam, AF-3, containing 0.2 M HF with pH 3 adjusted by sulfuric acid. On the contrary, the oxide film completely dissolved within 3 hours in the acidic decontamination foam, AF-4, containing 0.5 M HF with pH 3 adjusted by sulfuric acid.

On the other hand, FeCr_2O_4 film formed on the STS 304 specimen was completely removed within 3 hours by oxidative decontamination foam, OF-3 containing 0.2 M Ce(IV) as an oxidant. However, FeCr_2O_4 film was hardly removed by the acidic decontamination foam, AF-1 containing 2 M HNO_3 or AF-2 mixed with 0.5 M HNO_3 and 0.5 M HF. From these results, it was found that the oxide film containing chromium can be easily removed by the oxidative decontamination method, while it was difficult to remove only by dissolving with a strong acid.

3.2 Decontamination Performance

Decontamination performance of acidic and oxidative decontamination foams on the Inconel 600 sealing assembly drawn from NPP was investigated to evaluate on-site applicability through hot demonstration of foam decontamination technology. To this end, an acidic decontamination foam, AF-4 consisting of 0.5M HF and 1wt% EM-100 (pH 3 adjusted by H_2SO_4) and an oxidative decontamination foam, OF-3 consisting of 0.2M Ce (IV) , 1wt% EM-100 and 1 wt% M-5 are applied.

As a result of the decontamination of the sealing assembly using an oxidative decontamination foam, the decontamination factor increased until the first 2 hours, and then tended to be slower, and a decontamination factor of about 2 was obtained through decontamination for 5 hours. The result using acidic decontamination foam was also similar to that using oxidative decontamination foam. The decontamination factor increased slowly after the initial 1 hour, and about 1.8 was obtained through acidic decontamination for 3 hours. It showed a difference in decontamination efficiency to the extent that it was impossible to compare the decontamination behavior of the simulated oxide specimen. It is known that the decontamination factor for the Inconel heat transfer tube is not very large even by the application of commercialized chemical decontamination technologies applicable to the primary system of NPPs. It is presumed that the reason why the decontamination performance for the Inconel 690 sealing assembly did not meet the expectations is that the properties of the oxide film formed on the sealing assembly are different from the simulated oxide film. That is, the oxide film formed on the sealing assembly is a $\text{Fe}_x\text{Ni}_y\text{Cr}_z\text{O}_4$ spinel structure, which is not an easy form to remove by only a single-stage foam decontamination process that applies either acidic or oxidative decontamination foam.

Therefore, as a countermeasure to this cause, a multi-stage foam decontamination process was applied in which the acidic decontamination foam was applied in the first step and then the oxidative decontamination foam was applied in the second step. After the two-stage foam decontamination was completed, the decontamination performance and the appearance of the sealing assembly is shown in Fig. 1. The decontamination factor of about 5.7 was obtained by repeated decontamination with acidic and oxidative decontamination foams for a total of 5 hours.

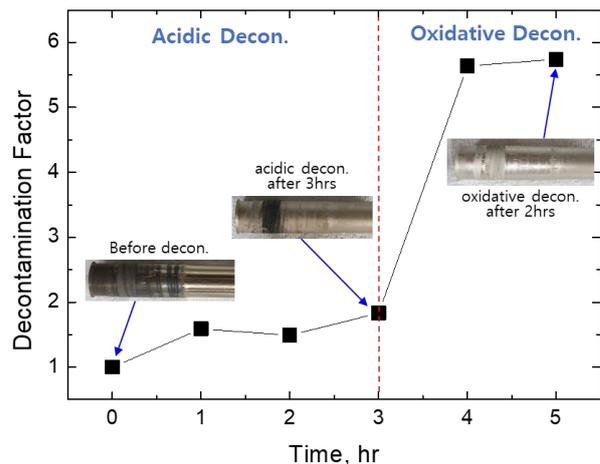


Fig. 1. Multi-stage decontamination performance for Inconel 690 sealing assembly.

4. Conclusions

Through the removal tests of simulated oxide specimens coated with NiFe_2O_4 and FeCr_2O_4 film using the decontamination foams, the effectiveness of the acidic or oxidative decontamination foam was confirmed depending on the characteristics of the oxide film. An applicability of multi-stage foam decontamination process to the Inconel 690 sealing assembly as fixed radioactive contamination was verified through the performance test result of a significant improvement in the decontamination efficiency upon application of first-step acidic decontamination foam followed by second-step oxidative decontamination foam.

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