# Effect of silica nanoparticles on the stability of decontamination foam and their application for oxide dissolution of corroded specimens

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A B S T R A C T

Foam stability was investigated by varying the amount of silica nanoparticles in a decontamination foam containing a surfactant and a stabilizer. In addition, a study on the oxide dissolution of a corroded specimen using a decontamination foam was performed to evaluate the decontamination efficiency of the foam. The decontamination foam prepared with Elotant™ Milcoside 440N (EM440N) as a non-ionic surfactant displayed the highest foam stability compared with that of foam containing other surfactants such as SDS and Triton X-100 in acidic pH. For the decontamination foam prepared using silica nanoparticles, the liquid volume in the foam was enhanced by a factor of 2 compared with that of the foam prepared using only a surfactant. Silica nanoparticles are thought to play a key role as an effective stabilizer of decontamination foam in acidic pH. The decontamination efficiency toward dissolving iron was improved by up to approximately 94% when using a decontamination foam in 1.0% EM440N consisting of the mixture of 3.0 wt.% silica nanoparticles and 0.1 wt.% xanthan gum compared with that of the chemical decontamination agent alone. This result indicates that an increase in foam stability enhanced the oxide dissolution of the decontamination foam owing to an increase in the contact time between the decontaminant foam and the corroded specimen.

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## 1. Introduction

Nuclear facilities must be decontaminated to reduce the radiation field of complex equipment during operation, while being repaired, or when under decommissioning (Riess et al., 2009). Chemical decontamination techniques have been employed to decontaminate surfaces such as components, structural materials, and equipment contaminated with radionuclides. However, the complexity of the equipment used and the location of radioactive contaminants hinder the implementation of conventional aqueous processes (Nunez and Kaminski, 2007).

Decontamination foam has a potentially wide range of applications in the cleaning of metallic walls, overhead surfaces, and elements of complex components and facilities (Nunez and Kaminski, 2007). In addition, the foam is a good material for *in situ* decontamination because it generates a low final waste volume owing to its volume expansion. Therefore, such foam substantially decreases the amount of chemical decontamination agents used during decontamination processes and the amount of secondary waste

volume generated (Causse et al., 2009). The decontamination efficiency can be enhanced by improving the contact time between the chemical decontamination agents and the contaminated surface through the introduction of stabilizers into the decontamination foam (Dame et al., 2005).

Various stabilizers reported in the literature can be grouped into the following categories: organic compounds, electrolytes, finely divided particles, polymers, biopolymers, and liquid crystals (Lai and Dixit, 1996). Decontamination foam was developed at the Commissariat a l'Energie Atomique (CEA) using biodegradable non-ionic surfactants and viscosifiers (Faure et al., 2010). The use of xanthan gum as a viscosifier increases the foam's lifetime and thus the contact time between chemical decontamination agents and contaminated walls (Dame et al., 2005). In addition, the CEA reported that silica nanoparticles act as viscosifying agents and enhance foam stability (Guignot and Faure, 2010).

Due to the unique surface properties, hydrophilic silica particles cause an increase in the viscosity of liquid films, leading to a delay in drainage and an enhancement in foam stability (Kruglyakov and Taube, 1972; Wilson, 1980). Conversely, silica nanoparticles can be specifically hydrophobized for optimal adsorption at the liquid/gas interface, which creates a type of armor for bubbles and prevents

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coarsening by reducing internal gas transfer (Binks, 2002; Abkarian et al., 2007; Martinez et al., 2008). The foam can be stabilized by mixing amphiphilic surfactants and inorganic particles, and the foam stability can be enhanced by modifying the surface of the nanoparticles from being hydrophilic to being partially hydrophobic (Binks et al., 2008; Zhang et al., 2008).

The surface in nuclear facilities may become contaminated with radioactive isotopes during operation, while being repaired, or when under decommissioning. Radionuclide uptake occurs up to the inner oxide layer and oxide/metal interface. Thus, it is necessary to remove the oxide layer using decontamination foam containing chemical decontamination agents. In this study, a corroded specimen was used to prepare simulated LWR oxide layers on an Inconel 600 surface. To ensure the effective decontamination of radioactive contaminants from a system being decontaminated, the decontamination foam was evaluated for its ability to dissolve the oxide layers of corroded specimens. However, a study on foam stability and the decontamination of corroded specimens using decontamination foam containing a non-ionic surfactant in the mixture of silica nanoparticles and a viscosifier has not been conducted.

In this work, to develop new decontamination foam, we prepared decontamination foam with highly enhanced foam stability and decontamination efficiency through a combination of various viscosifiers based on the results of previous studies and silica nanoparticles owing to their stability in acidic pH containing chemical reagents. The foam stability was evaluated for decontamination foam of various compositions using a Foamscan instrument, and the efficiency of the foam toward the decontamination of a corroded specimen was investigated.

## 2. Materials and methods

### 2.1. Preparation of the decontamination foam

In the experiments, all surfactants were used without further purification. Sodium dodecyl sulfate (SDS, purity >99%) and Triton X-100 (purity >99%) as anionic and non-ionic surfactants, respectively, were supplied by Sigma–Aldrich. A non-ionic surfactant containing decyl glucoside, Elotant™ Milcoside 440N (EM440N), was supplied by LG Household & Health Care in the Republic of Korea and contained an average of 10 alkyl chains. Various viscosifiers were tested for their ability to increase the stability of the decontamination foam: xanthan gum (Sigma), gelatin (J.T. Baker), glycerol (Sigma–Aldrich), and carboxymethylcellulose (CMC, Sigma). To investigate the effect of nanoparticles on the foam stability, fumed silica nanoparticles (M-5, Cabosil) were selected owing to their stability in acidic media and low cost. All decontamination foam formulations were prepared using deionized water obtained from a Milli-Q water system.

### 2.2. Stability test for the decontamination foam

The foaming properties (i.e., foam formation and foam stability) were determined using a Foamscan instrument (Teclis/IT Concept, Longessaigne, France). Foamscan is a commercially available instrument that can measure the foamability (i.e., the amount of foam generated over a certain period), foam stability (i.e., the decay in foam volume as a function of time), and foam drainage (i.e., the change in liquid volume in the foam as a function of time). Fig. 1 shows a schematic diagram of this equipment. For the foam stability tests, 60 ml of DI $H_2O$ was mixed with a 1.0% solution of various surfactants SDS, Triton X-100, and EM440N in a 100 ml beaker. To a 1.0% EM440N surfactant solution, 0.5, 1.0, and 3.0 wt.% M-5 silica
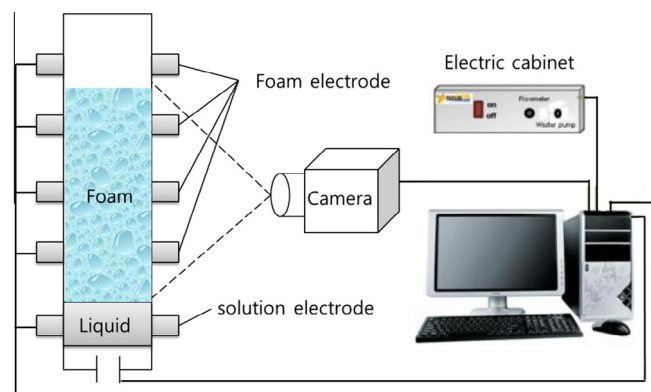


**Fig. 1.** Schematic diagram of a Foamscan instrument.

nanoparticles and 0.1 wt.% xanthan gum were added to investigate the foam stability.

The foam was generated by blowing nitrogen gas at a flow rate of 200 ml/min through a porous glass filter at the bottom of a glass tube in which 60 ml of the prepared decontamination foam (25 ± 1 °C) was added. After the foam volume reached 200 ml, bubbling stopped, and the foam was analyzed by monitoring the change in conductivity and optical properties of the foam using a CCD camera.

The liquid volume in the foam, the foam homogeneity, and the liquid drainage from the foam were measured based on a change in the foam's conductivity using electrodes positioned in different locations of a glass column. The liquid volume in the foam was calculated by subtracting the liquid volume (placed at the bottom of the column) in the cell from the initial liquid volume (60 ml) by measuring the change in conductivity (Carey and Stubenrauch, 2009, 2010).

The viscosity of decontamination foam is very useful for determining the foam's stability. The viscosity properties of the various foam formulations were measured using a rheometer (Brookfield Eng. & Lab. Inc., LVDV-III ULTRA) and Rheocalc 32 software at a shear rate of 66/s for 2 min (120 points).

### 2.3. Oxide dissolution of the corroded specimen using the decontamination foam

A study of the oxide dissolution of corroded layers formed on the surface of an Inconel specimen was carried out using various decontamination foam formulations. Inconel specimens with dimensions of 20 × 20 × 2 mm were used to test the extent of oxide dissolution. Inconel 600 is composed of Ni (75.95 wt.%), Cr (15.17 wt.%), Fe (8.19 wt.%), and other elements (0.69 wt.%). The specimens were polished with abrasive paper and washed with water and ethyl alcohol. The corroded layer on the specimens was grown for one month in a semi-loop filled with a solution composed of 650 ppm $H_3BO_3$ and 2.2 ppm LiOH and maintained at 350 °C at 2500 psi, simulating the primary water in LWR. The corroded Inconel specimens were analyzed by transmission electron microscopy (TEM) and selected area electron diffraction (SAED). The analyses showed that the oxide layers were grown and deposited on the surface and that the resultant chemical composition of the oxide layers was $NiFe_2O_4$ (Park et al., 2012). Based on these results, the degree of oxide dissolution was evaluated by the concentration of dissolved iron in the foam liquid.

To compare with the decontamination foam prepared in this work, a chemical decontamination agent solution was prepared with 30 ml of 1 M $HNO_3$ for a control test. In addition, three decontamination foams were prepared with 30 ml of 1 M $HNO_3$ for the