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Structure and stability of decontamination foam in concentrated nitric acid and silica nanoparticles by image analysis

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ABSTRACT

The structure and stability of decontamination foam containing chemical reagents were investigated by image analysis, for silica nanoparticle (NP) concentrations varying from 0 to 5 wt.%. The decontamination foam that contained 1% v/v non-ionic surfactant and 1 M HNO_3 as the chemical reagents at extremely low pH, was stable for 60 min when the silica NP content was greater than 3 wt.%. The foam structure was analyzed in terms of bubble size distribution and bubble count to clarify the enhancing effect of silica NPs on the foam stability. Image analysis showed that as the addition amount of silica NPs increased, the total number of formulated bubbles increased while the bubble size decreased. The amount of silica NPs attached to the bubbles increased with the addition of silica NPs, indicating that silica NPs present in the liquid film around the bubbles effectively inhibit bubble breakage. In bubble structure, the thickness of liquid film in silica NPs was higher than that without silica NPs. The measurements of liquid volume in foam confirmed that the chemical reagent increased in the increase of the liquid film. The decontamination efficiency, in terms of weight loss of the corroded specimen, increased in the addition of silica NPs due to the increase of chemical reagents in thicker liquid film. This study provides a new insight into the role of silica NPs in decontamination foam.

1. Introduction

Nuclear facilities are routinely contaminated with radionuclides during operation; more specifically, corrosion oxides may be formed on the surface of large components of nuclear facilities. The uptake of radionuclides by these oxides depends on the specific operational and structural conditions of the facility (Rao et al., 1997; Song et al., 2004; Thomas, 1990). Thus, it is necessary to remove the oxide layer by chemical or physical decontamination methods. However, a large amount of secondary wastes was often generated from chemical decontamination process. In this regard, using decontamination foam, which contains chemical decontamination agents, is considered one of the alternatives to reduce secondary waste (Yoon et al., 2014a). In addition, the foam are good for decontaminating complex shapes in vertical surfaces injecting gas for filling decontamination foam.

Decontamination foam comprises a surfactant to generate the foam, a co-surfactant to reduce the total number of surfactants, and chemical reactants to dissolve the contaminants on a solid surface. Since the foam requires relatively low concentrations of chemical reactants, the contact time of these reactants in the foam must be sufficiently long for better decontamination efficiency. Previously, CEA (Commissariat a l'Energie Atomique) developed decontamination foams using bio-degradable non-ionic surfactants and viscosifiers (Faure et al., 2010). The use of xanthan gum as a viscosifier increased the foam's lifetime as well as the contact time between the chemical decontamination agents and the contaminated surface (Dame et al., 2005). In addition, silica nanoparticles (NPs) were used as viscosifying agents to enhance the foam stability (Guignot and Faure, 2010; Yoon et al., 2014a). Our previous study showed that a mixture of silica NPs and xanthan gum increased the foam stability and decontamination efficiency (Yoon et al., 2014a). In this study, only silica NPs were used, without xanthan gum as stabilizer, to reduce the use of organic materials for the treatment of secondary waste in the facility.

Owing to the unique surface properties of foam, the hydrophilic silica particles remain in the liquid film of the bubbles, thus preventing drainage of liquid in the foam (Carn et al., 2009;

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Guillermic et al., 2009). Silica NPs can be specifically hydrophobized for optimal adsorption at the liquid/gas interface; such hydrophobization leads to the formation of resistive membranes between the bubbles and prevents coarsening by reducing internal gas transfer (Abkarian et al., 2007; Binks, 2002; Martinez et al., 2008). Mesoporous silica NPs with various shapes and structures were synthesized and used as additives to efficiently enhance foam stability and decontamination (Yoon et al., 2014b,c).

Previous studies have reported that the structure of foam has a strong influence on its rheology and stability. Rand and Kraynik (1983) described the relationship between bubble size distribution and drainage rate. In their study, the enhanced stability of foams with smaller bubbles was explained by a decrease in drainage. Sarma and Khilar (1988) reported that a more uniform bubble size distribution and higher initial gas volume fraction provide more stable foams. Horozov (2008) indicated that silica NPs form dense particles around the bubbles and a three-dimensional network in the bulk aqueous phase to stabilize the liquid films.

In this study, a decontamination foam containing NPs was tested to evaluate the stability and structure of bubbles in terms of their size and number. Image analysis was performed to investigate the relationship between the structure and stability of the decontamination foam. To ensure effectiveness of the proposed decontamination foam, dissolution tests were conducted on a simulated oxide layer from the surface of carbon steel.

2. Experimental

2.1. Preparation of decontamination foam

Commercial surfactants were used without further purification for all experiments. The surfactant Elotant™ Milcoside 100 (EM100) (alkyl polyglucoside) was supplied by LG Household & Health Care, and it contained 10 alkyl chains on average. All solutions were prepared using deionized water from a Milli-Q water system. To investigate the effect of NPs on the foam stability, fumed silica NPs (M-5, Cabosil) were selected because of their high stability in acidic media, as well as low cost. The fumed silica NPs had network structures consisting of particles of 30–50 nm in diameter, as shown in the TEM image in Fig. 1. For the foam stability tests, 100 ml of deionized H_2O was mixed with a 1.0% v/v solution of EM100 and 1 M HNO_3 in a 100 ml beaker for all samples. M-5 silica NPs of 0, 0.5, 1.0, 3.0, or 5.0 wt.% were added for investigating the foam stability.

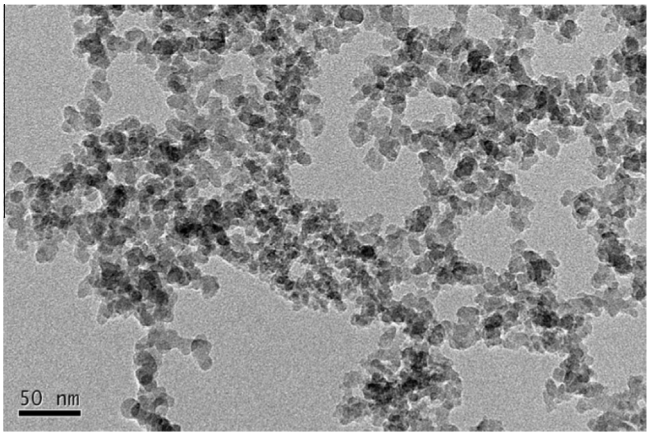

Fig. 1. TEM images of fumed silica NPs (M-5, Cabosil).

2.2. Structure and stability of decontamination foam

In all tests for foam stability and structure, the foam height and the liquid volume in the foam were measured using a Dynamic Foam Analyzer (DFA-100, KRŰSS, Germany) while simultaneously analyzing the foam structure (*e.g.*, size distribution of bubbles, and thickness of the liquid layers between the bubbles). During foaming, compressed air was passed through a sintered glass frit at the bottom of a cylindrical glass vessel (40 mm inner diameter) containing the solution of decontamination foam. The diameter and thickness of the glass frit were 44 and 4 mm, respectively. The pore size of the frit was 16–40 μm. The initial liquid volume was 60 ml; the gas flow was 0.2 l/min and was stopped after about 60 s of foaming. The foam height and liquid height were measured using the LED panel and photon detector located in the front and back of the column (Fig. 2) and was monitored continuously by measuring the light transmission through the glass column (Bilke-Krause et al., 2010; Oetjen et al., 2014). The total foam height was 200 mm (about 250 ml) for all measurements.

A camera with a scanning area of 10.5 × 7.5 mm was positioned 55 mm above the glass frit, for observing the bubble size distribution. As per the principle of total reflection, a prism allows 2D structure analysis of bubbles placed along the path of light. Because glass and liquid have comparable refractive indices, the light incident on foam lamella is partially diffracted and thus transmitted into the foam. In contrast, glass and air have different refractive indexes, and hence, the light hitting the gas bubbles is completely reflected. The resulting high-contrast images were analyzed using Foam Analysis Software, with the bubble size distribution being recorded every 2 s (Oetjen et al., 2014). The liquid volume was measured by DFA for drained liquid over time after foam generation. The liquid volume in foam was the amount of remained in liquid film. The amount of liquid volume in foam was determined by subtraction of liquid volume from initial liquid volume and calculated as follows:

$$\text{Liquid volume in foam } (\%) = \frac{\text{Initial liquid volume} - \text{liquid volume}}{\text{Initial liquid volume}} \times 100$$

Subsequent to the foam stability tests, the amount of silica NPs around the bubbles was quantified. To capture the buoyant NPs, the water already drained on the bottom was discarded. The remaining dried foam was washed three times with DI water, and then centrifuged three times to remove the surfactants adhered to the NPs. The obtained particles were dried and weighed.

2.3. Decontamination tests on the corroded specimen using decontamination foam

A study on weight loss of the corroded layers formed on the surface of a carbon steel specimen was carried out using decontamination foam with various concentrations of silica NPs. Carbon steel specimens with dimensions of 40 × 40 × 2 mm were used to test the extent of weight loss. The specimens were polished with abrasive paper and washed with water and ethyl alcohol. The corroded layer on the specimens was grown for 5 h at 773 K in a muffle furnace. In SEM–EDS analysis, the main element on the surface of carbon steel was 78.7% Fe, 14.8% C, and 6.5% O. After thermal treatment in air atmosphere, the corrosion layer was formed by the oxidation of carbon steel and its composition was 53.4% O, 38.1% Fe, and 8.5% C (Fig. 3). As seen in the figure, the corrosion layer has a structure of flake or scale-like.

Decontamination foams were prepared with 100 ml of 1 M HNO_3 in a 1.0% v/v EM100 surfactant solution containing varying