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Foam decontamination containing silica nanoparticles of various structures

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1. Introduction

A large amount of liquid waste is generated from the decontamination that occurs when dismantling nuclear facilities. A process is needed to decrease the amounts of chemical reagents and secondary waste produced during the decontamination process. Decontamination foam is a non-stable, two-phase fluid with aqueous and gas phases representing not more than 10% and 90% of the total volume, respectively. This formulation can significantly decrease the amounts of chemical reagents and secondary waste [1].

The advantage of decontamination foam is its potentially wide application for metallic walls, overhead surfaces, and the elements of complex components and facilities. In addition, the application of foam allows for remote decontamination processing using only an injection nozzle and the equipment to generate the decontamination foam, which reduces operator exposure to high radioactivity [2].

Solid colloidal particles increase the foam stability in the foam formulation. These particles can be specifically hydrophobized for optimal adsorption at the liquid/gas interface, which creates armor for the bubbles and prevents coalescence by reducing the internal gas transfer [3-5]. Conversely, hydrophilic particles remain confined in the liquid phase, and to enhance the foam stability. In addition, the silica nanoparticles (NPs) were synthesized with various structures and used for the stabilizer of decontamination foam [6-7]. In this study, we aimed to synthesize silica nanoparticles (NPs) with various structures such as porous, core-shell, and non-porous using methods proposed in previous literatures. We also investigated the effect of silica NPs with various structures for the foam stability and oxide dissolution rate with chemical reagents.

2. Methods and Results

The non-porous silica NPs with size of ca. 200 nm in diameter were synthesized by combining the modified Stöber method [8] and silica seed growth [9]. In typical synthesis, the silica seed solution (24 ml) prepared after the stirring for 24 h at ambient temperature with molar ratio of TEOS/NH_4OH/H_2O/EtOH = 1.00/1.60/13.88/152.78 was suspended in mixture solution of absolute ethanol (1000 ml), deionized water (80 ml), and 28 wt. % NH_4OH (40 ml). After the stirring for 30 min, TEOS (40 ml) was added into the above reaction mixture and it continued stirring for 6 h under the same condition. Finally, the mixture had the addition of TEOS (80 ml) and was stirred for 6 h more.

In the synthesis of the porous silica NPs with various sizes, a mixture solution consisting of cetyltrimethylammonium bromide (CTABr; 2.4 g), 28 wt.% NH_4OH (7.5-30 ml), and DI H_2O (900 ml) was vigorously stirred for 30 min until reaching a clear solution. The mixture was added by TEOS (12.9 ml) and kept stirring for 60 min. Afterward the reaction mixture was aged at 343 K overnight.

The foam stability was measured using a Foamscan instrument. The foam is 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 where 60 ml of the foaming aqueous solution at ambient temperature is placed. In all experiments, the foam was allowed to reach a volume of 200 ml. The bubbling was then stopped, and the evolution of the foam was analyzed using conductivity and optical measurements. The silica NPs with various structures such as porous, core-shell, and non-porous of 1 wt.% were added to a 1% Elotant^{TM} Milcoside 100 (EM 100) surfactant solution to investigate the foam stability compared with only 1% surfactant.

In oxide dissolution test, 304 stainless steel specimens having 20 mm × 20 mm × 2 mm in dimension were used. The corroded layer of specimens was grown in a semi-loop. Non-porous, core-shell and porous silica NPs of 1 wt.% were added to a 30 ml of 1% EM 100 surfactant solution containing 1M HNO_3 and compared to the foam stabilized only with Elotant^{TM} Milcoside 100 (EM 100) . About 100 ml of decontamination foam was prepared by shaking the 30 ml of decontamination foam for 10 sec. The concentration of dissolved iron from the specimens was measured by Flame-Atomic Absorption Spectroscopy (AAS).

3. Results and Discussion

Fig. 1 shows TEM images of the non-porous, core-shell with a silica core and porous shell, and porous silica NPs. As shown in Fig. 1, the non-porous (Fig. 1a) and core-shell (Fig. 1b) silica NPs exhibited a monodispersive size of ca. 200 nm in diameter and uniform porous shell of ca. 20 nm in thickness, respectively. The pore structures of the porous silica NPs exhibited a size of ca. 50 nm in diameter and the existence of an ordered hexagonal array from the result of the TEM and XRD (data not shown) analysis. From the results of N_2 sorption isotherm, BET surface area, total pore volume, and average pore size of the core-shell and porous silica NPs exhibited 251 and 808 m^2/g, 0.22 and 0.84 cm^3/g, and 3.49 and 4.18 nm, respectively.

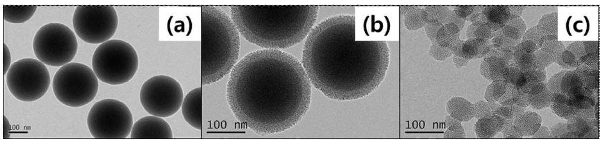

Fig. 1. TEM images of (a) non-porous, (b) core-shell, and (c) porous silica nanoparticles.

The addition of 1 wt.% porous silica NPs improves the life-time in liquid volume to fall to one-tenth as foam stability, compared to the foam stabilized only with 1% EM 100 (Fig. 2), indicating that the use of NPs increases the foam stability. The size of core-shell silica NPs was similar with that of non-porous silica NPs. However, the liquid volume in foam of core-shell silica NPs was higher than that of non-porous silica NPs owing to the low density and high porosity of the porous shell. The use of the porous silica NPs with lower density and smaller size showed an enhancement of the foam stability owing to their structural difference such as relatively high specific surface area, large pore volume, and porosity.

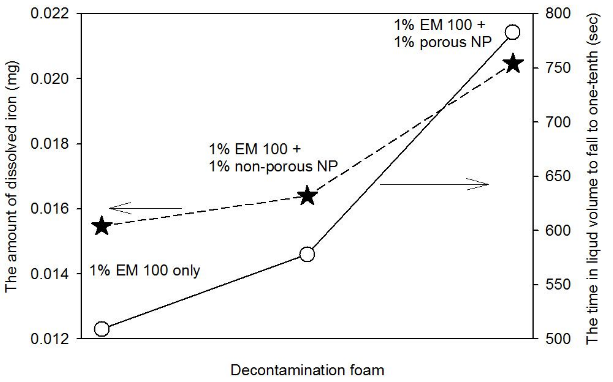

Fig.2. The concentration of dissolved iron using bottle tests of decontamination foam and the life-time in liquid volume to fall to one-tenth.

In oxide dissolution test, 1 wt.% porous and non-porous silica NPs were added to the decontamination foam containing 1M HNO_3. The mixture of porous NPs in surfactant improves oxide dissolution rate by a factor of 2 of dissolved iron compared with surfactant only. The foam stability was related the concentration of dissolved iron. These results indicated that the increase of stability in the foam accommodated the dissolution of iron because of enhancing the contact time of chemical reagents to the corroded specimen.

4. Conclusions

This study showed the effect of the silica NPs with various structures on the decontamination foam. The result indicates that porous NPs have a significant effect on the foam stability and oxide dissolution rate because of lower density and smaller size owing to high specific surface area, large pore volume, and porosity.

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