The separation of silica nanoparticle by cetyltrimethylammonium bromide from decontamination foam waste

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1. Introduction

Silica nanoparticle has been widely used in various field such as catalyst, biomolecule separations, chromatographic supports, miniaturization of electronic devices, and semiconductor nanostructures[1,2]. Recently, silica nanoparticle could be used as foam stabilizers for the decontamination foam[3]. Decontamination foam has been considered as a potential application for the cleaning of radioactive contaminant in the field of metallic walls, overhead surfaces, and complex components[4]. Moreover, foam decontamination could generate the low secondary waste amount owing to its volume expansion. In order to increase the decontamination efficiency, it is essential to improve the foam stability with low amount of chemical decontamination agent. Yoon et al. reported that the silica nanoparticle containing surfactant increased the foam stability compared to only surfactant solution[3]. Nanoparticle has been used with surfactant, which they adsorb at fluid/fluid interface, to stabilize emulsions or bubbles in foams[5-7]. Despite of improving foam stability, they still used the surfactant, silica nanoparticle (1 wt%), and viscosifier. In addition, it is difficult to separate silica nanoparticle from decontamination solution. Because nanoparticles differ from classical solid particles due to smaller particle size and their specific properties. Thus, the separation method for nanoparticle should be also developed with high recovery rates.

2. Methods and Results

For the silica nanoparticle separation, the CTAB was also used as additive to aggregate the silica nanoparticles in the SiO₂ solution, and its separation efficiency was investigated. The desired concentration of CTAB (powder) was added into the SiO₂ solution in the range of 0.05 mM to 5 mM. The SiO₂ amount in the solution was 1 wt%. The SiO₂ solution included CTAB was stirred at 400 rpm for 60 min. The SiO₂ solution was centrifuged at 3500 rpm for 30 min. After centrifugation, the top clear solution in the centrifuge tube was used to measure the transmittance of solution by Turbiscan (Formulaction, France). Turbiscan detected the backscattering and transmission of monochromatic light (λ =880 nm) through the sample. Measurement was conducted for 60 min. The results showed the change of transmittance of light versus time. Inductively coupled plasma atomic emission spectroscopy (ICP-AES, OPTIMA) was used to measure the SiO₂ content in the solution after centrifugation. The particle size of SiO₂ after adding CTAB was also measured using the particle size analyzer. All the measurements were carried out at room temperature.

3. Result & discussion



Fig. 1. Transmittance and Recovery rates of silica nanoparticles after centrifugation without CTAB.

CTAB can be used to separate the SiO₂ particles as additive for the aggregation of silica nanoparticles. Generally, silica nanoparticle shows a negative charge on the particle surface and the zeta potential ζ remains lower than 30 mV leading to their stability[8]. Liu et al. reported the aggregation of silica nanoparticles by cationic surfactant CTAB and proposed the coagulation mechanism of silica nanoparticle with CTAB. We used the CTAB as additive for separation of silica nanoparticle after foam stability measurement and the effect of CTAB for the SiO₂ separation. Fig. 1 shows the transmittance of SiO₂ solution after centrifugation without CTAB. The interpretation of the transmission profiles was based on the change in the light transmission caused by change of SiO₂ contents. As shown in Fig 1a, the transmittance of 1wt% SiO₂ solution without CTAB linearly increased according to the centrifugation time. However, no difference of transmittance increasing time was observed after 50 min, which means SiO₂ nanoparticles are still suspended in the solution. We also calculated the recovery rate to determine the separation efficiency. The reference was based on the transmittance of distilled water. About 83% of recovery rate of SiO₂ solution was achieved after 60 min by centrifugation. Fig. 2 shows the transmittance of SiO₂ solution with a series of CTAB containing different CTAB amount. The SiO₂ solution

was centrifuged for 30 min. When the CTAB added in the SiO_2 solution, the transmittance become more increase and similar with that of water even at small concentration of CTAB (0.1 mM).



nanoparticles after centrifugation with CTAB.

It is indicating that efficient agglomeration of SiO₂ particles can be obtained because of interaction SiO₂ and CTAB. The recovery rate of SiO₂ with CTAB was also similar with that of water (except for 0.05 mM of CTAB). From Fig. 2, the concentration of $CTAB \ge 0.1$ mM could effectively agglomerate the SiO₂ particles and very few silica contents remain in the solution. The Fig. 3 shows the particle size and SiO₂ contents according to the addition of CTAB amount. All measurements were carried out after reaching the equilibrium. At low concentration of CTAB (0.1 mM), no increasing particle size of SiO₂ was observed and SiO₂ contents was also high, indicating that it is low to induce the efficient aggregation. However, the particle size of SiO₂ was dramatically increased with CTAB concentration (0.5 mM), and SiO₂ contents was also decreased after centrifugation. The particle size and contents of SiO₂ are dependent on the CTAB concentration. It is reported that the charge neutralization seems to be main reason for nano silica aggregation, but hydrophobic effect from hydrocarbon tails of CTAB may contribute to the aggregation as well[8]. Liu et al. proposed the aggregation mechanism of silica nanoparticle; when the low concentration of CTAB, little CTAB can be absorbed on the silica surface. As increasing the CTAB, negative surface charge is reduced and agglomeration may occur until charge neutralization is achieved due to the hydrophobic interaction. Finally, when the CTAB concentration is too high, a bilayer of CTAB could envelop the particle surface and thus re-stabilize the silica nanoparticle suspensions[8].

3. Conclusions

The addition of CTAB induced the flocculation of SiO_2 particles and increase the hydrophobic interaction between SiO_2 particle and hydrocarbon tail of surfactant, which in turn improved the foam stability. The flocculation of silica nanoparticle added by CTAB could be quickly achieved for only 30 min. The particle size of SiO_2 was larger as CTAB amount increased, and SiO_2 contents in the top solution were decreased after centrifugation.



Fig. 3. The particle size and SiO_2 contents after separation according to the CTAB concentration.

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REFERENCES

[1] R. Hoppe, A. Ortlam, J. Rathousky, G. Schulz-Ekloff, A. Zukal, Syhtnesis of titanium-containing MCM-41 mesoporous molecular sieves in the presence of zinc phthalocyanine and rhodamine B, Microporous Mater. 8 (1997) 267-273

[2] D. Chomski, O. Dag, A. Kuperman, N. Coombs, G. A. Ozin, New forms of luminescent silicon: silicon-silica composite mesostructures, 2 (1996) 8-13.

[3] I.H. Yoon, C.H. Jung, S.B. Yoon, S.Y. Park, J.K. Moon, W.K. Choi, Effect of silica nanoparticles on the stability of decontamination foam and their application for oxide dissolution of cored specimens, Ann. Nucl. Energy, 73 (2014) 168-174.

[4] L. Nunez, L. Kaminski, Foam and gel methods for the decontamination of metallic surface, US patent, 7 (2007) 166,758.

[5] C.Y. Hu, S. L. Lo, C. M. Li, W. H. Kuan, Treating chemical mechanical polishing (CMP) waste water by electrocoagulation-flotation process by surfactant, J. Hazrd Mater., A120 (2005) 15-20.

[6] CY. Lien, J.C. Liu, Treatment of polishing wastewater from semiconductor manufactural by dispersed air flotation 132 (2006) 51-57.

[7] B.P. Binks, Particles as surfactants-similarities and differences, Curr. Opin. Colloid Interface Sci., 7 (2002) 21-41.
[8] Y. Liu, M. Tourbin, S. Lachaize, P. Guiraud, Silica nanoparticles separation from water: Aggregation by cetyltrimethylammonium bromide (CTAB), Chemosphere, 92 (2013) 681-687.