Experimental study on the effects of fine bubbles on polydisperse submicron aerosol removal efficiency during pool scrubbing

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Received: 13 December 2019; Revised: 26 April 2020; Accepted: 25 August 2020

Abstract
Radioactive aerosols are strongly diffusive and migratory and thus have presented one of the greatest challenges during the decommissioning of the Fukushima Daiichi Nuclear Power Plant (NPP). Although cutting through debris underwater can suppress the generation of radioactive aerosols from pool scrubbing to some extent, the removal efficiency of bubble columns can be influenced by many factors. In this study, fine bubbles (microbubbles and nanobubbles) with large specific surface areas were introduced into a simple scrubber; nanobubbles, in particular, are known to have long residence times in water. The effects of fine bubbles on the aerosol removal efficiency during pool scrubbing were studied for TiO$_2$ (around 100 nm) and ZrO$_2$ (around 100 nm) aerosols. Due to the fact that TiO$_2$ (4.23g/cm$^3$) has similar density with CsOH (3.68g/cm$^3$) and CsI (4.51g/cm$^3$). On the other hand, ZrO$_2$ was found in the fuel debris (Zirconium-Water Reaction). To clarify the effects of fine bubbles, three kinds of water were prepared (i.e., distilled water, nanobubble water, and microbubble water). As a result, the removal efficiency of fine bubbles for TiO$_2$ aerosols decreased, while that observed for ZrO$_2$ aerosols improved in some cases. The improved removal efficiency achieved using fine bubbles may provide a new method for suppressing the generation of radioactive aerosols in the decommissioning of the Fukushima Daiichi NPP.

Keywords : Radioactive aerosol, Debris, Pool scrubbing, Fine bubble, Removal efficiency, Fukushima Daiiichi

1. Introduction

Radioactive aerosols, which are strongly diffusive and migratory, are among the most difficult problems impeding the decommissioning of Fukushima Daiichi Nuclear Power Plant (NPP). While cutting debris underwater can somewhat suppress the submicron radioactive aerosols (Onodera et al., 1991) generated due to pool scrubbing (Peillon et al., 2017), the removal efficiency of bubble columns may be influenced by many factors. In the theory of aerosol absorption by bubbling described by Fuchs (1964) and Ghiaasiaan (1997), the internal circulation of gas inside a rising gas bubble is equivalent to Hill’s vortex and the absorption of aerosols at the interfaces of the rising bubbles can be described by inertial deposition, sedimentation, and diffusion. These three processes are related to particle size, bubble size, and the velocity at which bubbles rise. Many methods have been proposed to increase the amount of aerosols removed by the water phase, including air bubble subdividing devices (Cadavid-Rodriguez, 2014) and surfactants (Koch, 2012). Additionally, it has been shown that the strength of the internal circulation within bubbles is an important parameter affecting its particle removal rate. Slower internal circulation due to the presence of surfactants at the water–bubble interface will greatly reduce the particle removal rate (Friedlander, 2000). However, by adding surfactants, many microbubbles (MBs) can be generated at the nozzle inlet (Koch, 2012) and more particles can be removed from the resultant MBs due to their small size. Therefore, to achieve the highest removal efficiency with surfactants, the trade-off between internal circulation and bubble size is unavoidable.
It is clear that the conditions at the water–bubble interface are very important for pool scrubbing. However, due to the high radioactivity and large amount of radioactive waste at the Fukushima Daiichi NPP, air bubble subdividing devices are thought to be difficult to operate and likely to increase the amount of radioactive waste, while it is assumed that surfactants will increase the wastewater treatment load. Thus, a new method is needed to decrease the aerosols released at the surface of pools. In previous studies, methods for decreasing surface tension using nanobubbles (NBs) have been explored, as fine bubbles have the potential to decrease the sizes of rising bubbles during pool scrubbing (Yasui et al., 2019). Therefore, in this study, NBs, which have long residence times and large specific surface areas, were introduced into the water around the debris before cutting in Fukushima Daiichi NPP. This could be treated as a simple scrubber (Fig. 1); MBs were also tested for comparison. The effects of fine bubbles on the aerosol removal efficiency during pool scrubbing were studied for TiO$_2$ (around 100 nm) and ZrO$_2$ (around 100 nm) aerosols. The reason for choosing TiO$_2$ particles is that TiO$_2$ (4.23g/cm$^3$) has similar density with nuclear accident relevant cesium bound compounds CsOH (3.68g/cm$^3$) and CsI (4.51g/cm$^3$). However, for ZrO$_2$ particle, it was found in the fuel debris due to the Zirconium-Water Reaction during the accident. Therefore, TiO$_2$ and ZrO$_2$ particles are believed to be able to verify the feasibility of using fine bubbles to improve the scrubbing performance for radioactive aerosols. To clarify the effects of fine bubbles, three kinds of water were prepared: distilled, NB, and MB water, and the methods employed provide a new means of enhancing the removal of radioactive aerosols from NPPs already burdened by high radioactivity and abundant radioactive waste, such as the Fukushima Daiichi NPP.

![Schematic representation of fine bubbles in a pool scrubber.](image_url)

**2. Materials and methods**

**2.1 Objectives**

The primary objectives in this study were to: (1) explore the feasibility of using fine bubbles to reduce the release of aerosols during pool scrubbing, (2) verify if the remove efficiency was improved by introducing NBs or MBs, and (3) explain the phenomena observed in the experiment.

**2.2 Properties of fine bubbles that may influence aerosol removal efficiency**

Fine bubbles have several characteristics that may allow them to improve aerosol removal efficiency. Specifically, NBs have longer residence times than MBs and thus, NBs rise more slowly in pool waters (Sun et al. 2016). With their large specific surface areas, electrical charge under neutral pH conditions, and hydrophobic surface properties, fine bubbles are able to adhere to various particles, including aerosols (Sobhy et al. 2013, Ahmadi et al. 2014). Therefore, the particles transferring at the gas-water interface could be accelerated by the NBs-particles attachment. Furthermore, the presence of NBs can reduce the velocity at which bubbles rise, thereby increasing the probability of particles transferring to the water (Fan et al. 2010). At last, the presence of NBs may reduce the surface tension of pool waters, thus causing more and smaller bubbles to form in the water column as surfactant which was proved to be helpful in pool scrubbing process (Koch, 2012). As a result, NBs are believed to be a more suitable method to suppress the release of radioactive aerosols in Fukushima Daiichi NPP decommissioning as an environmentally friendly and low-cost material.
2.3 Experiment materials

Two kinds of particles were used in the pool scrubbing experiment: TiO₂ (rutile, 50% at 100 nm; 25% at <100 nm; 25% at >100 nm; maximum: 150 nm; minimum: 25 nm) (Kawamura et al., 2016) and ZrO₂ (80% at 100 nm; 10% at <100 nm; 10% at >100 nm; maximum: 150 nm; minimum: 30 nm). An aerosol-loaded gas was generated by a Palas RBG 1000 (Palas GmbH, Germany), a piston-brush-type particle dispersion system. The size and concentration of aerosols were measured using a Welas digital 3000 (Palas GmbH), which has two sensors placed before and after the water column. Nanobubbles were generated with an ultrafine GaLF standard (IDEC Corp., Japan) and the peak diameter was located at 100–200 nm. Microbubbles were generated with an OM4-MDG-045 magnetic gear pump (AURA TEC Co., Ltd., Japan) and the peak diameter was located ~1 μm. Nanobubble water was circulated for 10 min in the generator and MB water was circulated for 30 min in the generator with air. However, MBs were simultaneously generated in the NB along with NBs. Therefore, to prepare NB water, the method recommended by Calgaroto (2015) was used because of the presence of MBs in the generated NB water. As a result, the generated NB water was abandoned for 3 h before the scrubbing experiment. The fine bubbles in the water were then measured with an Archimedes resonant mass measurement (RMM) device (Malvern Instruments, UK).

2.4 Experiment conditions

The experimental set-up is shown in Fig. 2 and the experiment conditions are listed in Table 1. Nitrogen was used as the working gas. There were two parallel lines at the gas inlet; one was employed to supply gas to the aerosol generator. After the generator, the particle-loaded gas mixed with pure nitrogen from the second line to control the aerosol concentration. The total gas flow rate was controlled at 25 ± 1 L/min.

![Fig. 2 Experimental setup](image)

**Table 1. Experimental conditions.**

| Concentration   | TiO₂ 100 nm (particles/mL) | ZrO₂ 100 nm (particles/mL) |
|-----------------|----------------------------|----------------------------|
|                 | High | Low  | High  | Low  |
| Distilled water | 9.3×10⁴ | 2.1×10⁴ | 9.2×10⁴ | 5.0×10⁴ |
| NB 10 min (3 h) | 1.1×10⁵ | 2.1×10⁴ | 1.1×10⁵ | 3.5×10⁴ |
| NB 10 min (12 h)| 1.1×10⁵ | 3.9×10⁴ | 8.0×10⁴ | 2.2×10⁵ |
| MB 30 min       | 9.7×10⁴ | 2.8×10⁴ | 1.0×10⁵ | 1.5×10⁵ |

Note: 3 h and 12 h means the experiment was performed 3 hours and 12 hours after NB generation, respectively.

Before the aerosol-loaded gas flowed through a nozzle (48×0.7 mm) into the water column, which was 20×20×100 mm in size and had a water level of 0.55 mm, 5 L/min was extracted for measurement. After the water column set, 5 L/min of gas flow was also extracted for measurement. The sampling nozzles were designed for isokinetic particle...
sampling. The bubbles inside the water column were photographed via high speed camera at 1000 fps. The total concentrations of particles at the water column inlet are listed in Table 1. These concentrations were controlled; for the high concentration case, there were ~1000 particles/mL and for low concentration case, there were <5000 particles/mL. The removal efficiency can be calculated as:

\[ \eta_i = \frac{C_{\text{in},i} - C_{\text{out},i}}{C_{\text{in},i}} \]

(1)

where \( i \) represents parameters for certain particle sizes, \( \eta_i \) is the removal efficiency for certain particle sizes, \( C_{\text{in},i} \) is the particle concentration at the inlet of the water column, and \( C_{\text{out},i} \) is the particle concentration at the outlet of the water column. However, the two sensors cannot work simultaneously and thus, switching between the sensors was necessary. The practical measurement process is shown in Table 2.

| Count | N1 | N2 | N3 | N4 | N5 | N6 |
|-------|----|----|----|----|----|----|
| Upstream | N1 | N2 | N3 | N4 | N5 | N6 |
| Downstream | n1 | n2 | n3 | n4 | n5 | N6 |

Note: N1–N6 represent concentrations measured six times upstream and n1–n5 represent concentrations measured five times downstream (Charvet et al. 2011).

The working sensor was shifting during measurement between upstream and downstream lines. For example, the measurement began from N1, which measured the aerosols at the water column inlet for 30 s and then switched to the sensor at the outlet of the water column for n1. The switching between these sensors took 30 s. Then, n1 measured the aerosol downstream for 30 s. Due to the fact that the concentration may fluctuate slightly during the measurement interval, the removal efficiency of the Ith measurement for a certain particle size, \( i \), was calculated as:

\[ \eta_{i,I} = 1 - \frac{\eta_{i,I}}{\sum_{i=1}^{n} \eta_{i,I}} \]

(2)

The sediment on the surface of container was subtracted from the inlet concentration following the methods of Cadavid-Rodriguez et al. (2014).

3. Results and discussion

3.1 Fine bubble results

![Fig. 3 Size distributions of fine bubbles in different kinds of water: NB water 1 h (red), 3 h (yellow), and 15 h (green) after generation and MB water (blue).](image-url)
The measurements of the fine bubble waters used in this study are shown in Fig. 3. Three kinds of NB water are shown according to the time after generation. One hour, 3 h, and 15 h after generation, the total concentrations were 2.53×10^4 particles/mL, 1.91×10^5 particles/mL, and 8.63×10^3 particles/mL, respectively. NBs water from 3 h after generation was used in the experiment and showed the highest bubble concentration among the three cases; the peak value was located at approximately 160 nm. The results showed that the concentration of NBs increased 1–3 h after generation and then decreased 3–15 h after generation. The initial increase was assumed to be caused by the growth of very small bubbles, while the subsequent decrease was attributed to the disappearance of NBs. In the MB water, the concentration of NBs was 3.39×10^4 particles/mL, which was similar to the NB water 1 h after generation. However, almost no bubbles >1 μm in diameter were observed, which may indicate that MBs disappeared rapidly after generation.

### 3.2 Bubbles in the water column

Figure 4 shows the gas bubbles rising in the water column during the pool scrubbing experiment. The water column was divided into two parts, corresponding to the area by the outlet nozzle and just below the water surface. Although there were 48 small holes in the nozzle, when the gas was expelled, small bubbles merged into larger ones. While rising, these larger bubbles broke into smaller ones once again. There were clearly more bubbles in the NB water than in either the distilled or MB water, which demonstrated the possibility of improving the aerosol removal efficiency by introducing smaller bubbles. However, it was assumed that NBs also existed in the MB water, yet the flow pattern in the MB water was similar to that in the distilled water, which may have been caused by the low NB concentration.

### 3.3 Generated aerosol particles

Although 100-nm TiO_2 and 100-nm ZrO_2 aerosol particles were generated, these were not monodisperse; instead, they were widely distributed due to particle aggregation, as the dispersion system could not disperse all of the particles as small as several hundred nanometers. The concentration of generated aerosols changed over time (in 30 s for one measurement), as shown in Fig. 5 and the size distribution of the generated TiO_2 and ZrO_2 aerosols are shown in Fig. 6. The refractive indices of TiO_2 and ZrO_2 were 2.4+0i and 2.2+0i, respectively.

![Fig. 4 High-speed photographs of rising bubbles in (a, d) distilled water, (b, e) MB water, and (c, f) NB water.](image-url)
It is obvious from Fig. 6 that both of the particle types exhibited two peaks in their size distributions. For TiO$_2$ particles, the higher peak was near 450 nm and the lower peak was at ~160 nm. For ZrO$_2$ particles, the higher peak was near 160 nm and the lesser peak was widely distributed from 300–600 nm. These particle distributions may also indicate that TiO$_2$ particles are more likely to aggregate than ZrO$_2$ particles during gas flow.
3.4 Pool scrubbing efficiency of different particles

The distributions of the removal efficiency of TiO$_2$ particles are shown in Fig. 7(a, b). It can be seen from this figure that both high and low particle concentrations exhibited similar trends, indicating that the particle concentration in the gas had little, if any, effect on the removal of TiO$_2$ particles. When fine bubbles were introduced into the pool scrubber, removal efficiency declined, which was opposite to the predicted response. For particles <500 nm in NB or MB water, the removal efficiency even became negative (i.e., the particle concentration increased instead of decreasing), which may have been caused by the breakup of large aggregates or by the aggregation of small particles. However, this remains unclear because particles smaller than 150 nm could not be measured in this study. For cases 12 h after bubble generation, the removal efficiency increased slightly and approached that in distilled water; this may have been caused by the decreased NB concentration after 12 h. Although the flow pattern of MB water was similar to that of distilled water, the distribution of its removal efficiency was similar to that of NB water.

The distributions of the removal efficiency of ZrO$_2$ particles are shown in Fig. 7(c, d). It can be seen that high and low particle concentrations exhibited different trends, in contrast to the experiments with TiO$_2$ particles. For high concentrations, when NBs were introduced into the pool scrubber, the removal efficiency decreased notably. This phenomenon may also have been caused by the breakup of large aggregates or by the aggregation of small particles, as smaller particles were more abundant in the gas flow when compared to the TiO$_2$ experiments. However, for low concentrations, the NB water increased the removal of small particles (<250 nm) and exhibited a similar trend to the distilled water for larger particles. Notably, the removal efficiency of NB water increased for both high and low concentrations after 12 h. Additionally, the removal efficiency of MB water was similar to that of distilled water for ZrO$_2$ particles, which is different than the pattern observes for TiO$_2$ particles.
Fig. 7 Distributions of the removal efficiency of TiO$_2$ particles for (a) high particle concentrations and (b) low concentrations; ZrO$_2$ particles for (c) high particle concentrations and (d) low concentrations.

4. Conclusions

In the pool scrubbing experiment, TiO$_2$ and ZrO$_2$ particles were chosen to represent the behavior of parts of the radioactive aerosol particles in Fukushima Daiichi NPP. Although small particles (~100 nm) and a particle dispersion system were used for aerosol generation, it remained difficult to generate a monodisperse particle-loaded gas. The particles in the gas flow ranged from 150 nm to several micrometers. From the particle distribution observed during gas flow, it was found that TiO$_2$ particles were more likely to aggregate than ZrO$_2$ particles, which may indicate that ZrO$_2$ particles are more likely to break into smaller particles due to disturbances in flow. However, the lower limit of the sensor was ~150 nm; therefore, high concentrations of ~100-nm particles may have existed in the gas flow. If so, this may have led to increased coagulation and thus an increase in the number of larger particles.

The aerosol removal efficiency of NB water should be increased by increasing the number of small rising bubbles, as well as the circulation time in the water column (Fuchs 1964, Koch 2012). However, it was observed to be decreased in our experiment except the low ZrO$_2$ particle concentration case. The decrease of removal efficiency may have been caused by that NBs on the surface of rising bubbles prevented the absorption of particles into the bubbles. Additionally, the NBs and aerosol particles on the surface of rising bubbles may have caused the internal circulation of gas to cease (Friedlander, 2000). Finally, large aggregates may break into smaller particles or small particles (<150 nm) may coagulate into larger particles during pool scrubbing, thereby increasing the number of particles (i.e., the concentration) of certain sizes.

Here, we conducted pool scrubbing experiment in distilled, MB, and NB water to investigate the effects of fine bubbles on the removal efficiency of polydisperse submicron aerosols. From the measurement of these generated...
aerosols, TiO\textsubscript{2} particles were found to be more likely to aggregate than ZrO\textsubscript{2} particles. More small rising bubbles were also observed in the NB water case, which may indicate decreased surface tension. The removal efficiency of TiO\textsubscript{2} particles was decreased by the addition of fine bubbles. The agreement of the results of both high and low particle concentration was thought to be caused by the particle properties, for example, wettability. While the removal efficiency of ZrO\textsubscript{2} particles only decreased at high concentrations but increased with the addition of fine bubbles at lower particle concentration, which may show NBs may improve the removal efficiency for ZrO\textsubscript{2} particles and the decrease in high particle concentration may come from the breakup of big aggregation or aggregation of small particles.

For the experiment results of NB water after generation for 12h, it was found the removal efficiency increased to some extent compared with NB water case. Especially, for ZrO\textsubscript{2} particles, the removal efficiency of NBs case became higher than the distilled water case, which showed the NBs increased the removal efficiency. The difference of removal efficiency in NB water case and NB water (after 12h) case was assumed to be caused by the NB concentration change.

Therefore, the removal efficiency was found to be heavily impact by fine bubbles in the water column. In addition, NBs showed the potential to improve the scrubbing performance which could be used in the work for retrieval of fuel debris during the Fukushima Daiichi NPP decommissioning, although the operation conditions (NBs concentration, gas flowrate, particle types) should by optimized.

A negative removal efficiency was observed in the experiment, which was assumed to be caused by the breakup of large aggregates or, conversely, by the aggregation of small particles, since the lower limit of the sensor was ~150 nm and the particles we used were ~100 nm. To explain the phenomena observed in this study, additional experiments are needed and should include the control of fine bubble and particle concentrations. However, analyses based on the experimental conditions highlighted here may be useful for further analyzing different assumptions applied to and mechanisms of aerosol removal.

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[DOI: 10.1299/mel.19-00655] © 2020 The Japan Society of Mechanical Engineers
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