Preventing nuclear fuel material adhesion on glove box components using nanoparticle coating

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Abstract

Minimizing the retention of nuclear fuel materials in glove box components and curtailing the external exposure dose are desirable. Therefore, plutonium and uranium mixed oxide (MOX) powder adhesion-prevention technology involving nanoparticle coating of the acrylic panels of the glove box is developed. Surface analysis using atomic force microscopy showed that root mean square roughness value of the nanoparticle-coated acrylic test piece surface (75.8 nm) was higher than that of the noncoated surface (2.14 nm). The nanoparticle coating reduced the van der Waals force between alumina particles and the test piece surface through the formation of nanosized rugged surfaces. The coating reduced the minimum adhesion force (normalized by the particle diameter) between the uranium dioxide particle and the acrylic test piece surface. For the smallest particle (diameter: ~5 μm) associated with desorption, this minimum adhesion force decreased to ~5%. The nanoparticle coating also lowered the average adhesion mass per unit area of the MOX powder on the acrylic test piece to ~10%. The expectation is that this method will reduce the retention of nuclear fuel materials in the box, lower the external exposure dose, and improve the visibility of the acrylic panels.

Keywords: Nanoparticle coating, Acrylic panel, Alumina (Al₂O₃), Uranium dioxide (UO₂), Plutonium and uranium mixed oxide (MOX), AFM, Centrifugal method, Adhesion

1. Introduction

In the plutonium and uranium mixed oxide (MOX) fuel fabrication process, a MOX powder is handled in a glove box. This box consists of acrylic viewing panels or polycarbonate viewing panels on the sides, stainless steel on the outer frame, and chlorosulfonated polyethylene rubber gloves. The MOX powder is prepared using a microwave heating method in Japan, and is characterized by excellent fineness, high activity, and high sinterability (Koizumi et al., 1983; Segawa et al., 2016). However, for submicron average particle sizes of the powder, the powder is scattered during handling, and the influence of the van der Waals force increases. This facilitates adhesion of the powder to the inner surface of the glove box components, causing retention in the box. The α-rays from the powder produce colors that lead to visibility deterioration of the acrylic panels composing the glove box, while the gamma- and neutron-rays result in an external exposure dose. Therefore, preventing MOX powder adhesion to the glove box components is necessary.

Recently, studies have reported that a water-repellent coating can be created by applying nanoparticles to the surfaces of glass and polymer (Suzuki, 2007; Conti et al., 2018; Widati et al., 2019). In addition, studies suggest that a nanoparticle coating reduces, to approximately one-seventh, the adhesion force of alumina (Al₂O₃) particles to glass substrates, thereby providing a high adhesion-prevention effect (Suzuki et al., 2013). The nanoparticle coating also reduces the van der Waals...
force of adhesion due to the formation of nanoscale asperities on the surfaces of substrates and particles (Suzuki et al., 2013; Yang et al., 2005; Deng et al., 2017). The nanoparticles aggregating on the acrylic surface are smaller than the wavelength of visible light, and hence the transparency of the acrylic panel remains undiminished with the use of coatings involving these particles. Thus, we investigated the prevention of MOX powder adhesion on acrylic panels using a nanoparticle coating.

The amount of powder adhering to the acrylic panel is related to the adhesion force, particle size, particle mass, and particle morphology. The particle characteristics of general ceramics such as alumina powder differ from those of the UO$_2$ and MOX powders. Therefore, investigating the means of preventing powder adhesion to the nanoparticle-coated acrylic test piece requires evaluation using these powders.

In this study, the influence of nanoparticle coating on powder adhesion was first assessed using Al$_2$O$_3$ powder, and then further investigated using the UO$_2$ powder. Subsequently, the average adhesion mass per unit area was evaluated using the MOX powder.

2. Materials and Methods
2.1 Materials
2.1.1 Adhering powder
Al$_2$O$_3$ powder was used as a simulant of the UO$_2$ and MOX powders used in the fuel fabrication process. Spherical fused Al$_2$O$_3$ particles (Micron Co. Ltd.) with an average particle diameter of approximately 10 μm were dried at 150°C for 12 or more hours in a desiccator. The UO$_2$ powder was obtained via calcination and reduction of uranium trioxide prepared by microwave heating a uranyl nitrate aqueous solution. An SEM image of the UO$_2$ powder is shown in Figure 1. The average particle size of the powder was ~0.5 μm, as shown in Figure 2. Furthermore, the MOX powder (average particle diameter: ~0.5 μm) was obtained via calcination and reduction of PuO$_2$–UO$_2$ prepared by microwave heating a mixed aqueous solution of uranyl nitrate and plutonium nitrate (Koizumi et al., 1983; Takeuchi et al., 2009).

![SEM image of UO$_2$ powder.](image1.png)

**Fig. 1 SEM image of UO$_2$ powder.**

![Particle size distribution of UO$_2$ powder.](image2.png)

**Fig. 2 Particle size distribution of UO$_2$ powder.**

2.1.2 Test piece
Test pieces were made by cutting the acrylic polymer (polymethyl methacrylate) plate (Acrylite MR-200W®, Mitsubishi Rayon Co., Ltd.) which was fabricated by the continuous casting method as same as the acrylic viewing panels of the glove box. Two sizes of test pieces with 10 × 25 mm and 20 × 50 mm were employed.

The continuous casting method has the advantages of casting method and extrusion method, and is suitable for the production of large-scale and smooth acrylic plates used for the glove box. In the continuous casting method, acrylic raw material is poured between the upper and lower stainless steel belts, and an acrylic plate is produced by casting polymerization. However, in the continuous casting method, when the acrylic plate is fed from the stainless steel belt, linear fine grooves are formed on the surface of the acrylic plate in the run direction.

2.1.3 Nanoparticles
A silica nanoparticle suspension (Glaco miller coat ZERO®, SOFT99 Co., Ltd.) containing spherical silica nanoparticles (primary diameter: ~7 nm) dispersed in isopropyl alcohol, was used as the coating material. Since Glaco is suitable for coating on the hydrophilic glass substrate, the nanoparticles can be applied uniformly on the glass surface.
Whereas acrylic test piece is hydrophobic and may be dissolved in the isopropyl alcohol with high volatility contained in Glaco, and hence the uniformity of the nanoparticle coating on acrylic surface is lower than that on the glass surface.

2.2 Methods
2.2.1 Nanoparticle coating preparation

Acrylic test pieces were coated with the nanoparticle suspension and dried at 70°C for at least 12 h in a desiccator. The differences between the surface states of the acrylic test pieces without and with nanoparticle coating were investigated. Images, values of the root mean square roughness (RMS), and average surface roughness (Ra) of the acrylic test piece surfaces noncoated and coated with nanoparticles were measured via atomic force microscopy (AFM; SPM400, SII Nano Technology Inc.). The AFM images were collected using two types of cantilevers. That is, one with a spring constant of 0.02 N/m (SN-AF01-S-NT, SII Nano Technology Inc.) for the contact mode and one with a spring constant of 42 N/m (SI-DF40S, Hitachi High-Tech Corp.) for the dynamic force microscope (DFM) mode, respectively. The contact mode is suitable for measuring hard surfaces, and the cantilever is brought into contact with a sample surface (Arutunow et al., 2013, Vasić et al., 2020). The DFM mode is used for high-resolution observation of a sample with a markedly uneven surface, and the cantilever oscillates at a specific frequency (Jin et al., 2009; Riau et al., 2015).

Table 1 shows the RMS and Ra values. Similarly, Figures 3 and 4 show AFM concavo-convex images obtained for the surfaces of acrylic test pieces without and with a nanoparticle coating. As shown in Table 1, the RMS and Ra values of a test piece were 2.14 and 1.70 nm, respectively. An RMS value of 1.5 ± 0.3 nm (Riau et al., 2015), which was almost the same as the one in this study, has been reported for an acrylic surface. However, after the nanoparticle coating, the thickness of the acrylic surface increased to several-hundred nanometers and the ruggedness of the surface also increased (Figure 4(c)). The RMS and Ra values increased to 75.8 and 52.3 nm, respectively, as shown in Table 1. Surface height and the RMS values of 70–200 and 95.6 nm, respectively, have been reported for a glass surface coated with silica nanoparticles, which were prepared by the sol-gel process (Widati et al., 2019). The surface roughness of a nanoparticle coating on a substrate can be affected by the nanoparticle characteristics, surface state of the substrate, and deposition characteristics of the nanoparticles on the substrate.

Table 1 Surface roughness of the acrylic test pieces noncoated and coated with nanoparticles.

<table>
<thead>
<tr>
<th>Description</th>
<th>RMS [nm]</th>
<th>Ra [nm]</th>
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<tbody>
<tr>
<td>Acrylic surface noncoated with nanoparticles</td>
<td>2.14</td>
<td>1.70</td>
</tr>
<tr>
<td>Acrylic surface coated with nanoparticles</td>
<td>75.8</td>
<td>52.3</td>
</tr>
</tbody>
</table>

Fig. 3 Surface roughness images obtained via contact-mode AFM performed on the surface of acrylic test pieces without a nanoparticle coating; (a) 3-D image, (b) 2-D image, and (c) surface roughness.

Fig. 4 Surface roughness images obtained by performing AFM (in the DFM mode) on the surface of acrylic test pieces with a nanoparticle coating; (a) 3-D image, (b) 2-D image, and (c) surface roughness.
2.2.2 Al₂O₃ powder and UO₂ powder adhering method

To assess differences in the adhesion force between various powders and the acrylic test pieces with and without a nanoparticle coating, the powders were exposed to the test piece using the apparatus shown in Figure 5.

The apparatus comprises a sieve, funnel, and rubber tube (Suzuki et al., 2013). Each powder was placed on the mesh of the apparatus and blasted by an air flow from under the mesh. The air flow distributed the powder through the rubber tube, causing adhesion to the test piece. Subsequently, the test pieces were turned to remove particles with adhesion forces below gravity. The forces for adhesion of the Al₂O₃ and UO₂ powders to the acrylic test piece were measured using centrifugal equipment.

Fig. 5 Schematic of the apparatus for adhering of Al₂O₃ or UO₂ powders. The sieve, funnel, and rubber tube are shown.

2.2.3 MOX powder adhering method

The applicability of the nanoparticle coating to the acrylic panel of the glove box in the nuclear fuel fabrication facility was evaluated using the MOX powder employed in the actual process. Owing to the limitation of the equipment installed in the box, the measurement of adhesion force by the centrifuge was impossible. Therefore, the adhesion level of the MOX powder to the acrylic test pieces with and without the nanoparticle coating was evaluated.

The adhering apparatus is shown in Figure 6. The MOX powder was sieved through a 160 μm mesh sieve, and exposed to acrylic test pieces with and without a nanoparticle coating. Subsequently, the MOX powder was uniformly adhered to the acrylic test pieces with a jig, which was partially cut and the acrylic test pieces were kept steady for 5 min. Thereafter, the test pieces were turned to remove particles via gravity force.

Fig. 6 Illustration of the adhering apparatus for the MOX powder involving a 160 μm mesh sieve.
2.2.4 Centrifugal method for measuring the adhesion force

The centrifugal method is widely used as a simple and practical method for measuring the adhesion force between particles and substrates (Suzuki et al., 2013; Booth et al., 1987; Podczeck et al., 1996, Podczeck, 1999; Thomas et al., 2015; Thomas et al., 2017). The adhesion force between the particle and surface of a test piece was measured using the centrifugal equipment shown in Figure 7. Owing to the centrifugal force generated by the rotation of the centrifuge, a force was applied to the particles adhering to the surface of a test piece. The force was applied in the direction in which particles were released from the test piece. The outer wall was coated with grease to prevent readhesion of particles desorbed from a test piece due to the centrifugal force. Moreover, the rotation speed of the centrifuge was increased to 5000 rpm, with each rotation speed maintained for 1 min. The particles remaining on the surface of each test piece were then observed under a microscope.

\( F = m r \omega^2 = \frac{\pi}{6} X^3 \rho D r \left( \frac{2\pi N}{60} \right)^2 \)  \hspace{1cm} (1)

Where, \( m \) is the particle mass calculated from the particle diameter \( X \) and the particle density \( \rho_D \), \( r \) is the revolution radius from the center of the rotating part of the centrifuge to the surface of the acrylic test piece, and \( \omega \) is the angular velocity, which was obtained from the revolution speed \( N \).
2.2.5 Adhesion quantity measurement

The MOX powder masses adhering to the acrylic test pieces with and without nanoparticle coating were measured using an electronic balance (ME54E, Mettler Toledo Co., Ltd.). All experiments were performed with \( n = 4 \). In addition, the average adhesion mass per unit area of the MOX powder was calculated.

3. Results and Discussion

3.1 \( \text{Al}_2\text{O}_3 \) powder results

To eliminate dependence on the particle size (Suzuki et al., 2013; Yang et al., 2005), the van der Waals force \( F_V \) is normalized as follows:

\[
\frac{F_V}{X} = \frac{A}{12z^2} \tag{2}
\]

Where, \( A \) is the Hamaker constant, \( X \) is the particle diameter, and \( z \) is the gap (generally defined as \( z = 0.4 \) nm). The Hamaker constant between the \( \text{Al}_2\text{O}_3 \) and acryl is estimated as follows:

\[
A_{\text{Al}_2\text{O}_3/\text{Acryl}} = \sqrt{A_{\text{Al}_2\text{O}_3}A_{\text{Acryl}}} \tag{3}
\]

The Hamaker constant of \( \text{Al}_2\text{O}_3 \) \((A_{\text{Al}_2\text{O}_3})\) is \( 15.5 \times 10^{-20} \) J (Shaw, 1980), whereas the corresponding value for acryl \((A_{\text{Acryl}})\) is \( 6.3 \times 10^{-20} \) J (Visser, 1972).

The normalized adhesion forces between the \( \text{Al}_2\text{O}_3 \) particles and acrylic test pieces are shown in Figure 8. For the surface noncoated with nanoparticles, the results reveal an essentially constant normalized adhesion force that is independent of the particle diameter, and the adhesion forces of the \( \text{Al}_2\text{O}_3 \) particles to acrylic test pieces are almost the same as those to the glass test pieces. In contrast, for the surface coated with nanoparticles, the maximum adhesion force of the \( \text{Al}_2\text{O}_3 \) particles to acrylic test pieces is lower than that for the surface noncoated with nanoparticles, and the lower limit of the adhesion force of the \( \text{Al}_2\text{O}_3 \) particles to acrylic test pieces is nearly the same as that to the glass test piece. The large variation in the normalized adhesion forces is assumed to be caused by the non-uniformity of the nanoparticle coating on acrylic test pieces.

The measured value of the normalized adhesion force acting on the acrylic test piece (~5 nN/\( \mu \)m) is considerably lower than the theoretical value (~50 nN/\( \mu \)m). Nanoscale ruggedness is likely even on the surface of the smooth-looking acrylic test piece (Figure 3(c)). For the surface coated with nanoparticles, the acrylic test piece and the glass test piece are characterized by similar normalized adhesion forces for the \( \text{Al}_2\text{O}_3 \) particles. The surface of the acrylic test piece produced a low RMS and appears smooth; hence, a uniform nanoparticle coating is possible, as in the case of the glass test piece. The decrease in the adhesion force is attributed to a diminishing van der Waals force resulting from increasing surface unevenness of the coating.

![Fig. 8 Relationship between the Al₂O₃ particle size and the normalized adhesion force associated with the particle size.](image-url)
3.2 UO₂ powder results

The released particles are identified as shown in Figure 9, and the normalized adhesion force of the UO₂ particles are shown in Figure 10. For the surface noncoated with nanoparticles, the normalized adhesion forces are relatively higher than that of the Al₂O₃ particles considered in Figure 8. Furthermore, the UO₂ particles are agglomerated, forming a large contact area with the surface of the acrylic test piece. This suggests that the normalized adhesion force is larger than that of the Al₂O₃ particles. In addition, a large variation in the normalized adhesion forces is observed due to the non-uniformity of the nanoparticle coating on acrylic test pieces as well as the results using the Al₂O₃ particles.

For the surface coated with nanoparticles, the normalized adhesion forces of the UO₂ particles are almost identical to those of the Al₂O₃ particles. The UO₂ particles desorbed at 3000 rpm or less were low on the noncoated surface, with most particles released at 4000 rpm or higher. A minimum normalized adhesion force of ≤3.8 nN is estimated for the particle with the smallest diameter (~5 μm). In contrast, a minimum force of ≤0.2 nN is estimated for the nanoparticle-coated surface, where particles desorbed from 1000 rpm. These results reveal that the nanoparticle coating reduces the minimum normalized adhesion force between the particles and surface of the acrylic test piece to ~5%.

![Fig. 9 UO₂ particles desorbed from the surface of acrylic test piece with a nanoparticle coating generated under a centrifugal force of (a) 3000 rpm and (b) 4000 rpm.](image)

![Fig. 10 Relationship between the UO₂ particle size and the normalized adhesion force associated with the particle size.](image)

3.3 MOX powder results

The average adhesion masses per unit area of the MOX powders adhered to the acrylic test pieces with and without the nanoparticle coating are shown in Figure 11. The maximum and minimum average adhesion masses per unit area are indicated by the horizontal lines above the test piece axis.
The average adhesion mass per unit area of the MOX powder is ~2.0 g/m² for the acrylic test piece without a nanoparticle coating, decreasing to ~0.18 g/m² for the test piece with the coating. The large variations between the maximum and minimum values of the adhesion masses may have resulted from the sieving test method. The nanoparticle coating reduces the average adhesion mass per unit area of the MOX powder adhesion on the test piece surface to ~10%. This emerges as a useful method for minimizing MOX-powder retention in the glove box, lowering the external exposure dose, and improving the visibility of the acrylic panels.

![Fig. 11 Average adhesion masses per unit area on acrylic test pieces with and without nanoparticle coating of the MOX powder.](image)

### 4. Conclusions

The effect of preventing powder adhesion on a fine uneven surface formed on an acrylic test piece by applying a nanoparticle coating is investigated. Images obtained via AFM revealed that the average surface roughness value of the nanoparticle-coated test piece is higher than that of the noncoated surface. The nanoparticle coating reduces the adhesion forces of the Al₂O₃ or UO₂ particles by reducing the van der Waals force resulting from the increase in surface ruggedness. The coating also reduces the average adhesion mass per unit area of the MOX powder adhesion on the acrylic test piece surface. Powder adhesion is prevented by the nanoparticle coating on the test piece surface of the glove box components. Accordingly, the expectation is that the coating minimizes the retention of the nuclear fuel materials in the glove box, lowers the external exposure dose, and improves visibility of the acrylic panels.

### References


