Cleaning Materials and Methods for Effective Removal of Indoor Radioactive Contamination

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To support the safe return of residents after the TEPCO Fukushima Daiichi Nuclear Power Plant accident, cleaning methods and materials for the removal of radiocesium (¹³⁷Cs) from household surfaces were compared. A spot contaminated with ¹³⁷Cs on a vinyl floor sheet or a glass plate was wiped in different moisture conditions with different cleaning materials, including a paper sheet, cellulose sponge, polyester sheet, and a polyester-polyamide sheet. Radioactive solid particles on a vinyl sheet were wiped with miniature mops made of cellulose sponge, polyester microfiber, or cotton yarn. There was little difference in the removal of radioactivity among cleaning materials when the contaminated spot was wiped in wet conditions. The removal of contaminated particles depended on the structure of the mop. The use of an abrasive and a detergent worked well for the vinyl sheet and the glass plate, respectively. These observations suggested that, in appropriate conditions, effective decontamination was achievable by regular indoor cleaning with commercially available cleaning devices.

Key Words: indoor decontamination, radiocesium, cleaning material, wipe test model, miniature mop model

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1. Introduction

Nearly nine years have passed since the radiological accident at the TEPCO Fukushima Daiichi Nuclear Power Plant (FDNPP). Decontamination of radioactivity in residential areas has been extensively carried out to reduce the ambient dose rate to 0.23 μSv/h or less, which is assumed to be equivalent to an effective dose of 1 mSv/yr for residents living an ordinary life¹,². Out of 604 monitoring stations located in the Fukushima prefecture, as of August 1 2019, 493 (81.6%) stations reported ambient dose rates lower than 0.2 μSv/h, and the remaining 60 stations (9.9%) reported rates lower than 0.5 μSv/h³,⁴. Along with the reduction of environmental radiation, the evacuation order to residents has been withdrawn in many regions. As of April 2017, the evacuation order zone area was 370 km², which was equivalent to 32.2% of the initial 2013 evacuation-designated zone area (1,150 km²). The number of people who originally lived in evacuation order zones has also decreased from 81,000 in 2013 to 24,000 in 2018⁵. However, the return of residents to their hometowns has not been progressing well and the number of evacuees remains at approximately 40,000 for various reasons⁶. Previous reports have indicated that one of the major concerns of returning residents (returnees) is the health effect of radiation in their living environment⁷–⁹.

As traditional Japanese and old wooden houses have poor airtightness, indoor contamination may easily occur through ventilation. It was reported that radiocesium contamination of indoor surfaces was inversely proportional to the distance from the FDNPP⁰. This surface contamination was
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mainly due to dry deposition. The estimated ratio of the contribution of surface contamination to the total exposure dose in houses was 3.0%\(^{10}\) or 8.9%\(^{11}\), using different dimension models. Therefore, even the personal effective doses of returnees are estimated to be lower than the annual dose limit of public, precise radiation monitoring and indoor decontamination seem to take an important role in reducing anxiety as well as in controlling the risk to health, which enables returnees to lead a safe life.

Devices and methods for decontamination in the radiation control area are relatively well established for the purpose of radiation protection\(^{12-16}\). The basic method is to wipe the contaminated surface with or without detergents and polishing agents. However, it is unclear whether this is also effective and applicable to the living environment. Surface cleaning and decontamination is also an important issue in medical care facilities in order to avoid the transmission of several key health-related pathogens\(^{17}\). It is noteworthy that indoor health-hazard fungal levels in the evacuation zone were considerably higher than the standard environmental level\(^{18}\). Effective cleaning devices and methods for decontamination and disinfection would be one of the strong supporting tools for returnees' lives.

We have developed cleaning materials and methods for infection control in hospitals and laboratories. We and our colleagues found that a floor-wiping method using a wet microfiber sponge-type device had a good result in terms of the reduction of floating microorganisms, possibly by preventing the resuspension of a dry deposit of microorganisms from the floor\(^{19,20}\). Through such findings, we have chosen several cleaning materials that may be applicable to indoor decontamination by the removal of radiocesium. In this study, we compared these cleaning materials and methods using two newly-developed experimental models that reproduced indoor environments, namely, a wipe test model and a miniature mop model. We then discuss the effective removal of radioactive contamination in ordinary residences.

2. Materials and methods

2-1. Wipe test model

When radioactive substances invade a house, contamination occurs on the surface of household furniture, goods, floors, walls, and other housing materials. In our experimental model, we divided these surfaces into two categories, soft and hard. We used a vinyl floor sheet (2 mm thickness, Art Optima, Achilles Corporation, Tokyo, Japan) and a glass plate (3 mm thickness, Float Glass, Nippon Sheet Glass, Tokyo, Japan) as their representatives, respectively. For the decontamination of these surfaces, four different types of cleaning materials were compared (Fig. 1). The first material (A) was filter paper, which is generally used for wipe test of radioactive contamination (Toyo Roshi Kaisha, Tokyo, Japan). The second material (B) was a sponge made of super-absorbent cellulose that is used for business cleaning (6 mm thickness, Decorators Sponge, Spontex, Paris, France). The third material (C) was a polyester sheet generally used for dust removal (0.8 mm thickness, Clean Cloth Pro, Seiwa, Okayama, Japan). The last material (D) was a multi-purpose dust removal sheet composed of polyester and polyamide (2-mm thickness, TASKI Microquick, CxS Corporation, Yokohama, Japan). All cleaning materials were cut into 25 mm-diameter pieces and used in both dry and wet conditions. For wet conditions, the cleaning material was immersed into water immediately before use, with or without a household detergent (Mypet, Kao Corporation, Tokyo, Japan) at a concentration of 0.1% sodium alkyl ether sulfate, and then squeezed tightly. The weight of a saturated piece was taken as 100% moisture content. Other moisture content percentages were adjusted by weight.

As a model of radioactive contamination, an aliquot of \(^{137}\)CsCl solution (2 kBq/mL, 200 μL, Eckert & Ziegler, Berlin, Germany) was dropped onto the center of a 5 cm × 5 cm area of a vinyl floor sheet or a glass plate. After drying overnight at room temperature (15°C to 25°C), the sheet or plate was placed on a scale and the contaminated area was wiped with each cleaning material by turning it toward the inside of the contaminated spot (Fig. 1) at a constant force of 2 kg, according to a previous investigation\(^{15}\). In some experiments, 30 mg of an abrasive (Cleanser, Lion Corporation, Tokyo, Japan) was put on the contaminated spot before wiping. The sheet or plate was then wrapped with a thin film and exposed to an imaging plate
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(Fujifilm Corporation, Tokyo, Japan) for 3 h. The remaining radioactivity on the surface was visualized and analyzed by a Fluor Imager (FLA-5100, Fujifilm Corporation) using FLA-5100 ImageReader software and Multi Gauge software, respectively. The region of interest (ROI) was set at an area of 5 cm × 5 cm, covering the entire square of decontamination. The photo-stimulated luminescence (PSL) signal from the ROI was measured. After subtracting the signal from the uncontaminated area (background), the rate of remaining radioactivity was determined by dividing the PSL of each spot by the PSL of the decontaminated spot.

2-2. Miniature mop model

If radioactive dust gets blown up during cleaning, it may cause internal exposure through inhalation. The use of wet cleaning devices, such as a wet mop, seems to reduce this possibility and give an effective decontamination. Therefore, we made three different miniature mop models for further experiments to simulate indoor cleaning with a mop for larger areas. The first one (a) was a cellulose sponge (Fig. 2, a), which was the same material as that used in the second cleaning material experiment Fig. 1 (B). The second material (b) is a polyester microfiber mop (Fig. 2, b; CONDOR Microfiber Mop, The Yamazaki Corporation, Osaka, Japan) with an anemone shape that enables a wide adsorption area for dust and dirt. The third model material (c) is a looped cotton yarn mop (Fig. 2, c; Endless mop, NORRIS company, Osaka, Japan). To assemble the miniature mop models, squares of approximately 5 cm × 5 cm of materials “a” and “b” were attached to an acrylic board with a small handle by a hook-and-loop fastener. A 5 cm × 5 cm square of material “c” of was attached by a clip.

An aliquot of 137CsCl solution (2 kBq/mL, 200 μL) was dropped onto one side of the 5 cm × 5 cm area of vinyl floor sheet to create a linear contaminated area 4 cm in length. It was kept at room temperature overnight. In addition, radioactive contamination by solids, such as soil or sand delivered from outdoors, was reproduced by using talc powder (median diameter of 12 μm, Japanese Pharmacopoeia, natural hydrated Magnesium Silicate, Ebisu Yakuhin Kako Corporate, Osaka, Japan) which was pretreated with an aliquot of 137CsCl (2 kBq/mL, 650 μL) and kept overnight for natural drying. It was then placed on one side of the vinyl sheet to create a linear contamination of 25 mg and 4 cm in length. The miniature mop materials were immersed into fresh water and squeezed tightly just before assembly. The vinyl floor sheet was placed on a scale and the linear contaminated surface was wiped in one direction by a wet miniature mop model at a constant force of 500 g. Then the miniature mop was disassembled and the mop material was washed with 100 mL of fresh water, and then squeezed five times. The radioactive material removed from the contaminated area by the mop and in the water was measured with a gamma counter (Model-2470, WIZARD², PerkinElmer Inc., MA, USA). The remaining rate was calculated by subtracting the ratio of the sum of the mop material and washing water to the initial radioactive substance dropped onto the vinyl floor sheet from one.

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R_r = \left(1 - \frac{R_m + R_w}{R_i}\right) \times 100
\]

*Rr*: Remaining rate
*Rm*: Radioactivity in mop material
*Rw*: Radioactivity in washing water
*Ri*: Initial radioactivity

All the experiments were performed more than three times, and the average of all the experiments is shown in the results.

3. Results

3-1. Wipe test model

The visualized radioactive contamination remaining on a vinyl floor sheet, which was used as a model for household furniture and floor soft surfaces, is shown in Fig. 3. The
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Contaminated area is clearly seen as a black spot. When the surface was wiped with a dry cleaning material (0% moisture content), the density of the spot appeared unchanged. In contrast, wiping with a wet cleaning material removed 20% to 100% of contamination, with no significant dependency on moisture content or on the type of cleaning materials. The results demonstrated that the rate of remaining radioactivity after wiping in wet conditions, using only water, was 27–36%, without any difference between cleaning materials. The remaining radioactivity remained as high as 87–96% after wiping in dry conditions (Fig. 4, Left). A statistically significant difference in the rate of remaining radioactivity was observed between wet and dry conditions for each cleaning material. This was observed in wet conditions using only water, with an abrasive, and with a detergent.

When the contaminated spot was wiped with an abrasive, the rate of remaining radioactivity generally decreased compared to that with water alone, with a statistically significant difference both in dry and wet conditions, presumably due to polishing of the surface. However, 21–33% of radioactive contamination remained. In contrast, wiping the contaminated surface with a household detergent at a concentration used for daily cleaning resulted in 31–42% higher rate of remaining radioactivity, compared to wiping with only water.

Figs. 5 and 6 demonstrate the results from similar experiments made using a glass plate to model the hard surfaces.
of housing materials. The size of the contaminated spot on the glass plate looked different than that on the vinyl sheet, i.e., the remaining radioactivity appeared to be concentrated toward the center of the spot. The density of each radioactive spot appeared to be generally reduced by wiping with cleaning materials (Fig. 5). The rate of remaining radioactivity on the glass plate after wiping in dry conditions ranged from 71% to 79% (Fig. 6), which was lower than 87–96% on a vinyl sheet (Fig. 4). When the contamination spot was wiped in wet conditions, the rate of remaining radioactivity decreased similarly to that on the vinyl sheet. Higher moisture content (60–100%) resulted in better decontamination than lower moisture content (20%), with statistically significant differences. However, the rate of remaining radioactivity did not reach the level of 27–36% which was observed on a vinyl sheet (Fig. 4). The use of a detergent improved the removal of radioactivity and the remaining rate was within the range of 21–33%.

In our experimental conditions, the radioactivity remaining on the contaminated spot after wiping was higher than expected. Therefore, we evaluated the removal of radioactivity on the vinyl floor sheet immediately after contamination. As shown in Fig. 7, the radioactively contaminated spot was almost invisible. We also shortened the drying period to 3 h using an air-drier.
Although the rate of remaining radioactivity after wiping in dry conditions was similar to that after drying overnight without an air-drier, wiping in wet conditions decreased the radioactivity to 8–18%, which was similar to or better than using an abrasive after drying overnight (Fig. 8). From these results, it was shown that a radioactive solution left on a sheet after overnight drying, i.e., our experimental conditions, made the contamination spot firmly bound and not easily removable.

3-2. Miniature mop model

As shown in Fig. 9A, the rate of remaining radioactivity on the surface of a vinyl sheet after wiping with a wet miniature mop ranged from 6% to 25%, which was lower than that obtained by the wipe test model (27–36%). This was observed even when the material was the same (wipe test model “B” and miniature model “a”). This could be attributable to differences in the method of measuring, method of wiping, or the size of the contaminated area and cleaning materials. Among the three different mop materials, “b” showed the lowest rate of remaining radioactivity on a sheet whereas “c” showed the highest remaining radioactivity, with a statistically significant difference than the other materials. The surface structure of the material, an anemone shape or a cotton yarn, possibly caused this difference.

Radioactive talc, a model for solid particle contamination, was more difficult to decontaminate than a dried radioactive contamination.
solution spot (Fig. 9B). The rate of remaining radioactivity ranged from 29.2% to 45.1%. The material “b” also showed the best results in this model, presumably due to its specific anemone shape, which held solid talc inside the structure.

4. Discussion

In this study, we compared the removal of radioactive contamination by various cleaning materials using two models of wipe test surface—a relatively soft surface (vinyl floor sheet) and a hard surface (glass plate). Moreover, we assembled a miniature mop model of different cleaning materials as wiping tools and compared the removal of radioactive contamination from a dried spot or a solid particle. Other types of contamination, such as lipophilic stain and oil spot, were not examined in this study.

The household cleaning materials selected for the wipe test model were highly absorbent and commercially available: a filter paper, a cellulose sponge, a polyester sheet, or a polyester and polyamide sheet. There was no significant difference in the remaining radioactivity between the four different materials. Therefore, the type of cleaning material was not a determinant of the effectiveness of decontamination. Instead, the moisture level of the cleaning materials (dry or wet) seemed to play an important role in removal of radioactivity. Wiping in dry conditions removed approximately 10% and 25% of the total surface radioactivity from a vinyl sheet and a glass plate, respectively. Similar results were reported by Shoji et al. in their previous investigation. For the vinyl sheet, when the cleaning materials were moisturized with water more than 60% of radioactivity was removed from the surface, regardless of the level of moisture content. The use of an abrasive appeared to improve the efficiency of radioactive material removal, whereas the use of a household detergent prevented removal. Although the underlying mechanism is not clear, one possible explanation is that radioactive molecules on the vinyl sheet surface infiltrated into the sheet in the presence of a detergent.

For the glass plate, more than approximately 50% of radioactivity was removed in wet conditions, which was further enhanced to 70% or higher using a detergent. Based on these observations, it was suggested that the glass plate surface should be wiped in wet conditions with cleaning materials of any kind, with or without a detergent.

In experiments using wet miniature mop models, the cellulose sponge and polyester microfiber materials demonstrated a higher rate of radioactivity removal from a dried spot, in the range of 90–95%. On the other hand, the removal rate of radioactive talc powder, a solid particle, was in the range of 55–70%. The structure of the cleaning material, such as an anemone shape, largely affects the physical removal of particles. Therefore, using a wet mop was also effective in removal of radioactivity; however, both the type and structure of a cleaning material should be carefully considered. It is obvious that remaining particles should be removed if they are visible. Furthermore, it is noteworthy that 100% removal of radioactive contamination was almost impossible once the radioactivity was fixed onto a surface by drying, regardless of the length of time after contamination. Conversely, the moisture content of cleaning materials may not be critical if the aqueous radioactive spot is wiped immediately after contamination.

The effective dose of Fukushima residents consists mainly of outdoor exposure, whereas the contribution of indoor exposure seems to be smaller. However, routine cleaning of households, which also works as an effective decontamination, is essential for reducing anxiety among returnees. Surprisingly, it has been reported that the radioactivity in house dust reached a value higher than 1000 Bq/g, which was significantly higher than that in soil samples from the same area in the evacuation order zone. Radioactivity may invade indoor spaces by direct ventilation, because the air-exchange rates of houses in the towns of Okuma and Futaba was reported to be approximately 0.69/h. In addition, radioactive dust attached to people and belongings could make them radioactive. In both cases, surface contamination by dry deposition occurs. The resuspension of house dust also raises the risk of internal exposure by inhalation. Our findings suggested that standard wiping in wet conditions, possibly combined with vacuum cleaning in advance, could keep the indoor environment clean. Any cleaning materials for the removal of dust, including a cloth, sponge, sheet or paper, can be used for the wiping. When wet contamination is expected, immediate cleaning before the wet spot dries will give a good result. Although the data were not shown, the radioactivity transferred to cleaning materials after wiping contaminated surfaces was not fully removed by washing and squeezing in fresh water. Approximately 10% of radioactivity was left on cleaning materials. Therefore, frequent replacement of cleaning materials, or use of disposable cleaning devices, is recommended. Furthermore, places where dust easily gathers should be cleaned the earliest because they could build up a hotspot of radioactivity.
Recent reports on the current status of environmental radiation in Fukushima have shown that an effective dose higher than 1 mSv/yr remains even in zones where the evacuation order has been lifted23,24. We easily found locations near the houses of returnees where the environmental dose rate was 0.23 μSv/h or higher, so the risk of indoor radioactive contamination may continue in the long term. Therefore, regular cleaning, keeping the purpose of decontamination in mind, will reduce the risk of indoor radiation exposure and accelerate the return to safe conditions for residents.

5. Conclusion
The effective decontamination of indoor household surfaces was achievable through regular cleaning with commercially available cleaning devices by applying conditions for the types of contamination and surface. These conditions included device structure, moisture content, and the presence or absence of detergent and abrasive. However, radioactivity was not completely removed once it had been fixed on the surface, which may make returnees feel uneasy. Further development of cleaning devices and methods to improve the efficiency of decontamination should be continued by using experimental models such as those established in this study.

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