Decontamination of radioactive-iodine- and cesium-contaminated spinach using sodium dithionite

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Washing of the spinach with water by the Nishizawa method resulted in the removal of about 13 ± 2% of the 131I and 45 ± 20% of the 134Cs and 137Cs. Removal rates of 131I of approximately 76 ± 3% after further washing with 1.2 wt% Na2S2O4 for 2 days and 47 ± 2% washing with 10 wt% Na2S2O4 for 2 days were obtained. These values agreed with the dissolution rates of 131I in the liquid used for washing within 2 to 3% error. The removal rates of 134Cs and 137Cs after washing with 1.2 wt% Na2S2O4 for 2 days were 69 ± 47% and 100 ± 52%, respectively, which were in poor agreement with the corresponding dissolution rates. The removal rate of 131I after brief washing with 10 wt% Na2S2O4 was negligible and the dissolution rate was 5 ± 2%. The removal rates of 134Cs and 137Cs for 10 wt% Na2S2O4 increased from 26 ± 11% to 42 ± 11% and from 30±20% to 42 ± 20% after washing with Na2S2O4 for 2 days. The high removal rates in 1.2 wt% Na2S2O4 may be due to the effective reduction of the I2 on the surface of the leaves by Na2S2O4 (1.2 wt%) during the 2 day immersion, which occurred without the bleaching of the leaves and stalks, which occurred in the case of 10 wt% Na2S2O4.

Key words: decontamination, radioactive iodine, radioactive cesium, sodium dithionite

1. Introduction

Radioactive iodine was dispersed into the environment and contaminated many agricultural products on the ground in the early part of March 2011 owing to the Fukushima Daiichi nuclear plant accident1). The radioactive iodine attached to leaves and entered plants through injured sites2). An effective method of decontaminating agricultural products was desired and was required for safety at home. Washing with water was tested, but it was not sufficient for the decontamination of 131I3). Therefore, a more effective decontamination method was explored.

In this study, the decontamination of radioactive iodine was investigated using sodium dithionite and the decontamination effect of sodium dithionite on radioactive-iodine- and cesium-contaminated spinach is discussed.

2. Materials and Methods

Sodium dithionite was purchased from Wako Pure Chemicals Co., Ltd. Contaminated spinach (obtained from the Mito branch of Hitachi-Aroka Medical Co., Ltd. on April 8, 2011) was used and its washing procedures and determination of γ-ray spectra were carried out as follows. The spinach was washed using the basic washing procedure of Professor Nishizawa. After the spinach was immersed in a bowl of water for 10 min, it was taken out and again washed with running water for 5 min. At that time, the leaves were spread and washed by scrubbing with fingers. Then, the water was removed from the washed spinach. The roots were cut off and all the leaves were separated. Then, the separated leaves were washed by scrubbing with fingers in running water to remove the remaining contamination without damaging the surface of the leaves. Finally, all the leaves were washed by mixing them gently by hand or using long chopsticks in a clean bowl with running water for 3 to 5 min, and they were taken out to remove the water completely. Before and after washing, their γ-ray spectra were determined using a Ge semiconductor detector; a plastic vessel for 10 wt% Na2S2O4 aqueous solution washing and another plastic vessel (the volume was 200 mL) for 1.2 wt% Na2S2O4 aqueous solution washing were used for the determination of their γ-ray spectra.
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300 mL each of 1.2 wt% and 10 wt% Na$_2$S$_2$O$_4$ aqueous solutions were used to wash and immerse the leaves and roots of the spinach for 2 days. After washing in 300 mL of 10 wt% Na$_2$S$_2$O$_4$ aqueous solution, the roots and leaves were put in the plastic bag and measured. The 300 mL of 10 wt% Na$_2$S$_2$O$_4$ aqueous solution used for washing was also measured. After washing using 1.2 wt% and 10 wt% of Na$_2$S$_2$O$_4$, the leaves and roots were immersed in 1.2 wt% and 10 wt% Na$_2$S$_2$O$_4$, respectively. After immersion in the 10 wt% and 1.2 wt% Na$_2$S$_2$O$_4$ aqueous solutions for 2 days, the roots and leaves were washed with running water. Then, after they were put in plastic bags for washing with 10 wt% and 1.2 wt% Na$_2$S$_2$O$_4$ aqueous solutions, their γ-ray spectra were determined. The 300 mL of 10 wt% and 1.2 wt% Na$_2$S$_2$O$_4$ aqueous solutions used for washing were also measured.

The dissolution rates of $^{131}$I, $^{134}$Cs, and $^{137}$Cs from contaminated spinach were calculated from their net peak counts in each liquid used for washing relative to their peak count after washing by the Nishizawa method. Their removal rates were calculated by subtracting the net count rate in leaves and stalks from 100%. The standard deviation (σ) for both rates was used 3σ calculated using the count rate of background.

3. Results and Discussion

Figure 1 shows a γ-ray spectrum of contaminated spinach measured on April 13, 2011. As shown in Fig. 1, a large peak and some subpeaks were detected and were assigned as follows: $^{129}$Te at 0.459 MeV (7.7%); $^{131}$I at 0.081 MeV (2.6%), 0.284 MeV (6.1%), 0.364 MeV (81.7%), 0.637 MeV (7.2%), and 0.721 MeV (1.8%); $^{134}$Cs at 0.569 MeV (15.4%), 0.605 MeV (97.6%), and 0.795 MeV (85.5%); $^{137}$Cs at 0.661 MeV (85.1% $^{137m}$Ba); $^{129m}$Te at 0.696 MeV (3.1%). Percentages indicate emission rates of γ-rays (%).

Water washing of the spinach by Nishizawa method resulted in about 13 ± 2% removal of $^{131}$I and 45 ± 20% removal of $^{134}$Cs and $^{137}$Cs. The removal rate of $^{131}$I by water washing was not enough. Since most of the radioactive species were $^{131}$I at that time, the removal of $^{131}$I was prioritized. The removal of the remaining $^{131}$I was tested using a reducing reagent, Na$_2$S$_2$O$_4$, to reduce $^{131}$I to $^{131}$I$^{-}$.

Table 1 shows the removal and dissolution rates of $^{131}$I, $^{134}$Cs, and $^{137}$Cs obtained from the contaminated spinach and the liquids used for washing after brief washing with Na$_2$S$_2$O$_4$ and after washing with Na$_2$S$_2$O$_4$ for 2 days. The removal rate of $^{131}$I after brief washing with 10 wt% Na$_2$S$_2$O$_4$ was $-9 ± 2$% and the dissolution rate was 5 ± 2%, indicating that most of the $^{131}$I was not removed. The removal rate of $^{131}$I after washing with 1.2 wt% Na$_2$S$_2$O$_4$ for 2 days was 76 ± 3% and the dissolution rate was 82 ± 3%; both rates were in good agreement and had 3% error. The removal rates of $^{134}$Cs and $^{137}$Cs after washing with 1.2 wt% Na$_2$S$_2$O$_4$ for 2 days were 69 ± 47% and 100 ± 52%, and their dissolution rates were 50 ± 47% and ND, respectively. The removal and dissolution rates agreed but had large errors, which were due to the small observed net count rates. The removal rate

| Content of Na$_2$S$_2$O$_4$ | After brief washing with Na$_2$S$_2$O$_4$ | Washing with Na$_2$S$_2$O$_4$ for 2 days |
|---------------------------|-----------------------------------------|----------------------------------------|
|                          | Removal rate (leaves and stalks)/%      | Dissolution rate (liquid used for washing)/% |
| $^{131}$I                 | $^{134}$Cs                               | $^{137}$Cs                             |
| 1.2 wt%                  | $-9 ± 2$                                | 26 ± 11                                |
| 10 wt%                   | $26 ± 11$                               | $30 ± 20$                              |
|                          | $5 ± 2$                                 | $14 ± 11$                              |
|                          | $16 ± 20$                               | $44 ± 20$                              |
|                          | $41 ± 2$                                | $50 ± 11$                              |
|                          | $61 ± 20$                               | |

Table 1
of $^{131}$I after washing with 10 wt% Na$_2$S$_2$O$_4$ for 2 days was 47 ± 2% and the dissolution rate was 41 ± 2%; both rates were in good agreement and had a 2% error. The removal rates of $^{134}$Cs and $^{137}$Cs after washing with 10 wt% Na$_2$S$_2$O$_4$ for 2 days were 42 ± 11% and 42 ± 20%, and their dissolution rates were 50 ± 11% and 61 ± 20%, respectively. The removal and dissolution rates agreed but had 20% errors. Therefore, the removal rate was used for discussion in the following.

The removal rates after the 2 day treatments by reduction with 10 wt% Na$_2$S$_2$O$_4$ were clearly larger than those after the brief treatment: $-9$ ± 2% for $^{131}$I, 26 ± 11% for $^{134}$Cs, and 30 ± 20% for $^{137}$Cs after brief washing and 47 ± 2% for $^{131}$I, 42 ± 11% for $^{134}$Cs, and 42 ± 20% for $^{137}$Cs after washing with Na$_2$S$_2$O$_4$ for 2 days. The high removal rates are due to the effects of 2 days of immersion, indicating that I$_2$ was reduced to I$^{-}$ and that I$^{-}$ dissolved in the aqueous solution. However, after 2 days, the solution became light green, indicating that the spinach was bleached by the high concentration of Na$_2$S$_2$O$_4$ (10 wt%) and that the greenish components of the spinach dissolved. Comparing the removal rates after washing with 1.2 wt% and 10 wt% Na$_2$S$_2$O$_4$ for 2 days, the removal rates of the former were clearly larger than those of the latter: 76 ± 3% for $^{131}$I, 69 ± 47% for $^{134}$Cs, and 100 ± 52% for $^{137}$Cs compared with 47 ± 2% for $^{131}$I, 42 ± 11% for $^{134}$Cs and 42 ± 20% for $^{137}$Cs. The increase in the dissolution rate for $^{131}$I may have resulted in the effective reduction of the adsorbed I$_2$ on the spinach in the oxidant form to I$^{-}$ in the 1.2 wt% Na$_2$S$_2$O$_4$ solution without the bleaching of the spinach. Therefore, the effective removal of adsorbed $^{131}$I on the leaves should only occur by surface reduction in the dilute Na$_2$S$_2$O$_4$ solution (1.2 wt%).

4. Conclusions

After washing the spinach with water by the Nishizawa method, removal rates of $^{131}$I of approximately 76 ± 3% after further washing with 1.2 wt% Na$_2$S$_2$O$_4$ for 2 days and 47 ± 2% washing with 10 wt% Na$_2$S$_2$O$_4$ for 2 days were obtained. These values agreed with the dissolution rates of $^{131}$I in the liquid used for washing within 2 to 3% error. The removal rates of $^{134}$Cs and $^{137}$Cs after washing with 1.2 wt% Na$_2$S$_2$O$_4$ for 2 days were 69 ± 47% and 100 ± 52%, respectively, which were in poor agreement with the corresponding dissolution rates. The removal rate of $^{131}$I after brief washing with 10 wt% Na$_2$S$_2$O$_4$ was negligible and the dissolution rate was 5 ± 2%. The removal rates of $^{134}$Cs and $^{137}$Cs for 10 wt% Na$_2$S$_2$O$_4$ increased from 26 ± 11% to 42 ± 11% and from 30 ± 20% to 42 ± 20% after washing with Na$_2$S$_2$O$_4$ for 2 days. The high removal rates in 1.2 wt% Na$_2$S$_2$O$_4$ may be due to the effective reduction of the adsorbed I$_2$ on the spinach in the oxidant form to I$^{-}$ in the 1.2 wt% Na$_2$S$_2$O$_4$ solution without the bleaching of the spinach. Therefore, the effective removal of adsorbed $^{131}$I on the leaves should only occur by surface reduction in the dilute Na$_2$S$_2$O$_4$ solution (1.2 wt%).

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