Report on "Radiation Disaster Recovery Studies"

Course ：Radioactivity Environmental Protection

Name ：Herry Wijayanto

○ Regarding "Radiation Disaster Recovery Studies"

On March 11, 2011, the strongest earthquake ever recorded in Japan occurred in Pacific Ocean, east of Tohoku Region. Immediately after the earthquake, a powerful tsunami came and caused flooding in the emergency power supplies of the Fukushima Daiichi Nuclear Power Plant (FDNPP) facing the Pacific Ocean. It led to cooling system interruption which induced the nuclear reactors to melt followed by the release of large amounts of radioactive substances into the environment. Consequently, more than 70% of municipalities in Fukushima Prefecture were designated as high-level radiation areas.[1,2]

In order to reduce the impact of high radiation levels on human health, Japanese government had been implemented decontamination work since the enactment of Special Measures Act in January 2012 regarding the handling of contaminated areas.[3] Decontamination work that includes removal of radioactive topsoil, grass, branches, leaves, etc. was completed on March 19, 2018.[1] Therefore, the radiation dose level in Fukushima Prefecture has been decreasing significantly due to decontamination efforts as well as natural dispersion. Accordingly, as of April 1st, 2017, most evacuation orders had been lifted and the residents had been able to return to their hometowns, except the Difficult-to-Return Zones (DRZ) where the annual cumulative radiation doses are over 50 mSv, which covered southern part of Iitate Village, northeast part of Katsurao Village, Namie Town, Okuma Town, Futaba Town, and some part of Tomioka Town.[4]

The most recent news informed that finally for the first time, partial evacuation order has been lifted in Futaba town (DRZ) on August 30, 2022—11 years since the accident, particularly 5.55 km^2 of area surrounding Futaba Station.[5] Even though the residents have been allowed to come back, however, more than 80% of the town is still considered as DRZ. In addition, many residents are reluctant to return home. A survey conducted by Reconstruction Agency in 2021 showed that only 11.3% of residents wanted to return, while 60.5% of them decided not to return. The remaining residents (24.8%) had not yet decided.[6] It is because former residents have already found jobs and restarted new lives in other cities in Japan. Another reason is that the current situation in Futaba town is still lack of employment and poor facilities such as schools, shops, hospitals or other public services. The similar condition also happened in Katsurao Village although evacuation order has been lifted on June 12, 2022. In fact, previous studies revealed that public anxiety (self-stigma) about health effects of radiation exposure when living in the ex-radioactive contaminated area is also inhibiting factor for residents to come back.[7,8] It seems the residents are pessimistic as well as traumatic about the future of their hometowns.

Indeed, Japan has advanced technology and financial capability to clean up radiation-affected areas quickly, nevertheless, the social problems faced by residents are very complex and more difficult to recover. In other words, it should be realized that there are "tangible" and "intangible" problems as the result of nuclear disaster. Contaminated land due to radioactive pollution is one of the tangible problems, but intangible problem like distrust and psychological issues is prolonged problems. For that reason, it is important to pay more attention to the social problem treatment by, for example, giving them more empathy and intense public consultation.

On the other hand, however, decontamination work produces high amount of radioactive contaminated waste. Basically, there are two kinds of waste: combustible (leaves, grass, branches, trees, etc.) and non-combustible which is cesium-contaminated soil. The combustible materials can be burned to reduce its volume, then the ash will be stored at waste storage facility at the Interim Storage Facility (ISF).[1] But, soil is difficult to be burned for volume reduction. Moreover, the volume of soil being stored in ISF is approximately 14 million m^3 (equivalent to 11 times Tokyo Dome) acquiring 1,600 ha of land area.[1]

To reduce the amount of contaminated soil in final disposal after 30 years of storing at ISF, the government actively proposes soil recycling.[1] The soil containing less than 8,000 Bq/kg can be used for embankment materials for roads, and other building levees, not only in Fukushima but all over Japan. However, this project faced protests from residents who doubted its safety level, especially its effects of radiation on health.[9] Meanwhile, according to survey of nation's prefectural governors (except Fukushima Prefecture), 7 governors opposed or rejected the use of radioactive soil, while others decided not to answer.[10]

In terms of storage space and cost saving, soil washing treatment can be considered as one of the possible choices to reduce soil volume that is stored at ISF, including supporting government projects about soil recycling by decreasing radiation levels in soil. In my research, I conducted very basic experiment about cesium desorption using cationic surfactant from clays and clay minerals of the soil by analyzing Cs desorption efficiency and its mechanism involved. The results obtained from this study hopefully afford new insights for surfactant selection in the washing remediation technology of cesium-contaminated clays and clay minerals including soil.

References

1 Ministry of the Environment, Japan. *Off-Site Environmental Remediation in Affected Areas in Japan.* August 2020.

2 NHK News. 2022. *Contaminated soil piles up in vast Fukushima cleanup project.* https://www3.nhk.or.jp/nhkworld/en/news/backstories/1942/

3 Ozawa, S. 2021. *Progress on Off-Site Cleanup Efforts in Fukushima.* Originally published in Journal of the Atomic Energy Society of Japan, 2015, **57** (6), 399-402 in Japanese.

4 Fukushima Prefectural Government. 2019. *Transition of evacuation designated zones.* https://www.pref.fukushima.lg.jp/site/portal-english/en03-08.html

5 The Japan Times. 2022. *Evacuation order finally lifted for Fukushima nuclear plant town.* https://www.japantimes.co.jp/news/2022/08/30/national/futaba-evacuation-order/

6 The Japan Times. 2022. *Fukushima town ready to lift evacuation order but only 10% of residents wish to return.* https://www.japantimes.co.jp/news/2022/07/27/national/fukushima-town-residents-not-returning/

7 Matsunaga, H., Orita, M., Iyama, K., Sato, N., Aso, S., Tateishi, F., Yasuyuki T., Kudo, T., Yamashita, S., and Takamura, N. *J. Radiat. Res.,* 2019, **60** (1), 51-58.

8 Orita, M., Hayashida, N., Nakayama, Y., Shinkawa, T., Urata, H., Fukushima, Y., Endo, Y., Yamashita, S., and Takamura, N. *PLoS ONE,* 2015, **10** (6), e0129227.

9 NHK News. 2019. *Disposing of Fukushima's contaminated soil.* https://www3.nhk.or.jp/nhkworld/en/news/backstories/399/

10 The Asahi Shimbun. 2021. *Survey: Not a single prefecture backs reuse of radioactive soil.* https://www.asahi.com/ajw/articles/14311546

○ Title of Doctoral Thesis

Cationic surfactant for remediation of ^{137}Cs-contaminated soil and the influence of its head group structure on the Cs desorption from clays and clay minerals

○ Summary of Doctoral Thesis

Substantial amounts of radionuclides released following the 2011 Fukushima Daiichi Nuclear Power Plant accident have contaminated lands in the Fukushima area of Japan. Of the radionuclides found in Fukushima soil, radioactive cesium (^{137}Cs) is still of major concern because of its long half-life ($t_{1/2}$ = 30.1 years), consequently, being major contributor to public health impact of long-term effect. Particularly, deposited ^{137}Cs in the soil takes part in the internal exposure when crops cultivated from contaminated land are consumed. It has been observed that Cs ions are strongly bonded to clay minerals of the Fukushima soil, causing their retention in the upper 5-cm soil layer.[1]

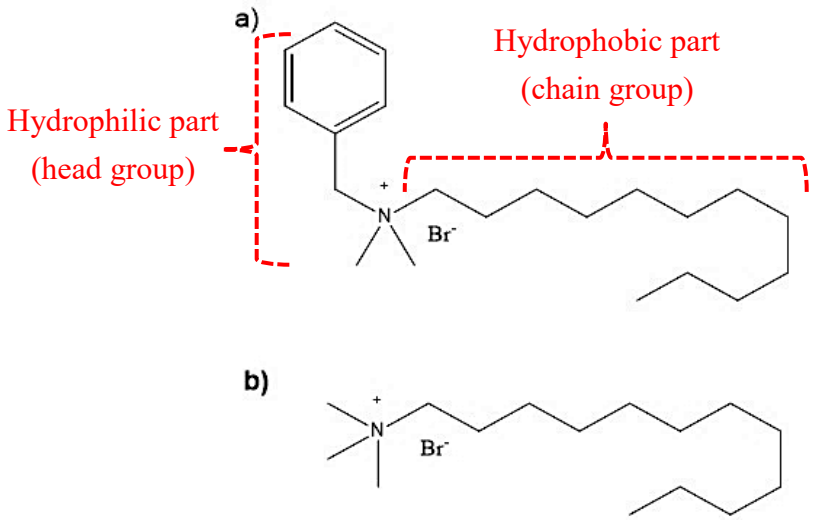

Fig. 1. Structure of (a) benzyldodecyldimethylammonium bromide (BDAB) and (b) dodecyltrimethylammonium bromide (DTAB)

Cs was mainly adsorbed to micaceous clay minerals[1] and probably also presented in kaolinite-, and smectite-like clay minerals.[2] In order to overcome the issues, soil remediation strategy is necessary both to reduce radiation effect on public health and to recover the land. The soil washing method by cationic surfactants ($R–N(CH_3)_3^+$; R=long hydrocarbon chain group; $N(CH_3)_3^+$ = head group) has been widely used as desorption agent (desorbent) to effectively desorb Cs from clay minerals via ion exchange between positively charged amine groups with Cs ions. Moreover, the length of their hydrocarbon chain group plays essential role in enhancing Cs desorption. The longer the surfactant chain length, the more Cs were released from kaolinite due to the stronger affinity of longer chain with kaolinite surfaces.[3] In the case of montmorillonite, a longer hydrocarbon chain increased its interlayer distance, therefore enhancing Cs desorption efficiency.[4] Although the influence of different surfactant chain lengths on the Cs desorption from clay minerals has been studied extensively, the contribution of their head group to such desorption mechanism has not been well understood. This study aims to examine the effectiveness of cationic surfactant of benzyldodecyldimethylammonium bromide (BDAB) to desorb Cs from actual ^{137}Cs-contaminated soil taken from Fukushima Prefecture and to investigate the influence of cationic surfactant head group on the desorption of Cs from specific clays and clay minerals by comparing two kinds of surfactants having the same 12 hydrocarbon chain lengths but different head group types. In this case, BDAB which has bulky head group was compared to dodecyltrimethylammonium bromide (DTAB) with linear structure (Fig. 1). The clays used in this study included Na- and Ca-bentonite, which are dominantly composed of montmorillonite. Clay minerals such as kaolinite and illite were also used.

To examine the effectiveness of cationic surfactant for soil remediation, first we compared BDAB with hydrochloric acid (HCl; strong acid), the commonly used desorbent in the soil washing method. The experimental results indicated that cationic surfactant of benzyldimethyldodecylammonium bromide (BDAB) was more effective in desorbing ^{137}Cs from contaminated soil of Fukushima even by using a low concentration in comparison to HCl. It should be noted that performance of desorbents for soil remediation depends on contaminants and soil characteristics, including the content of clay minerals, the content of other metals in soil, organic matters, etc.[5]

Therefore, in the next experiment, we used specific clays and clay minerals to investigate the influence of cationic surfactant head group on the Cs desorption. Clays and clay minerals samples were initially contaminated by Cs solution with maximum adsorption capacity, followed by desorption experiment using desorbents. Batch desorption experiment indicated that, in general, desorption efficiency for all studied samples increased by increasing surfactants concentration. In swelling bentonites that highly contain montmorillonite, Cs desorption was accompanied by interlayers'

expansion. In the case of Na-bentonite, BDAB showed higher Cs desorption efficiency than DTAB due to larger Na-bentonite interlayers expansion after intercalation of BDAB (bulkier ammonium head group structure). In contrast, Cs desorption efficiency from Ca-bentonite was relatively similar, which corresponded to the identical interlayers' expansion after intercalation of both surfactants. It was because divalent cation of Ca^{2+} must associate with two negatively charged interlayers, limiting interlayer expansion of Ca-bentonite. Cs desorption from kaolinite was more governed by surfactant micelles. In comparison to surfactant ions, surfactant micelles had a stronger affinity to the clay surface, enhancing Cs desorption efficiency. Fig. 2 shows that as the surfactant micelle formed earlier for BDAB, a faster Cs desorption was achieved. Thus, BDAB desorbed Cs more efficiently than DTAB due to formation of surfactant micelle at low concentration in BDAB solution. Fig. 3 displays possible mechanism of Cs desorption from kaolinite based on surfactant micelles formation. For Cs desorption from illite, both surfactants even showed relatively similar performance with HCl used as a desorbent control. This was due to the existence of frayed edge sites in illite, which provokes strong cesium retention. Desorption kinetic model of pseudo-second order fitted well to all studied samples by BDAB and DTAB suggested that desorption process was ion-exchange process.

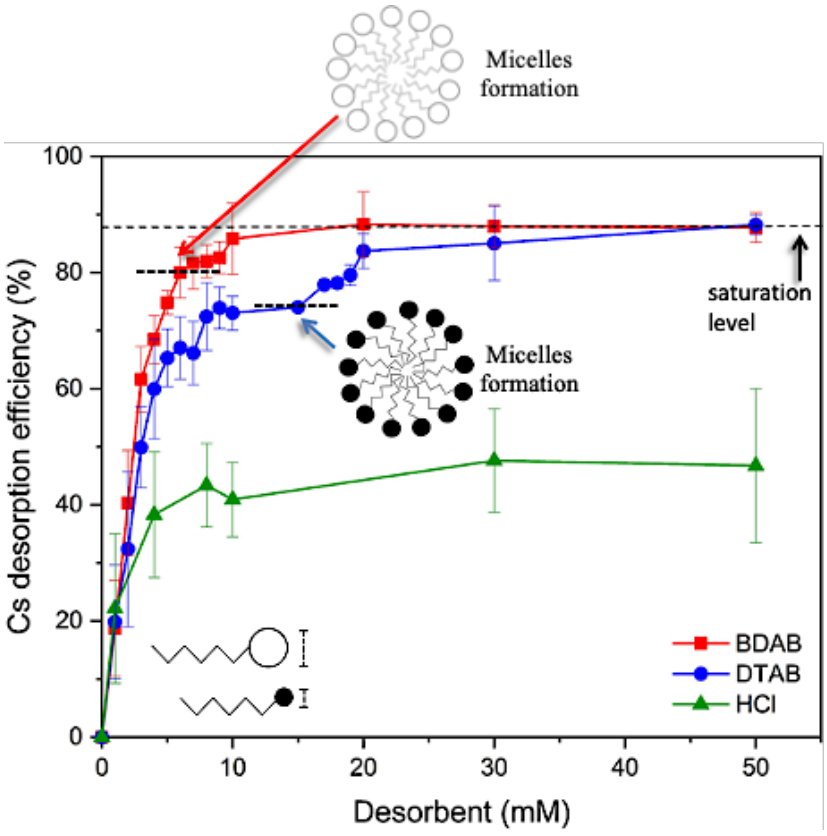

Fig. 2. Cs desorption from kaolinite

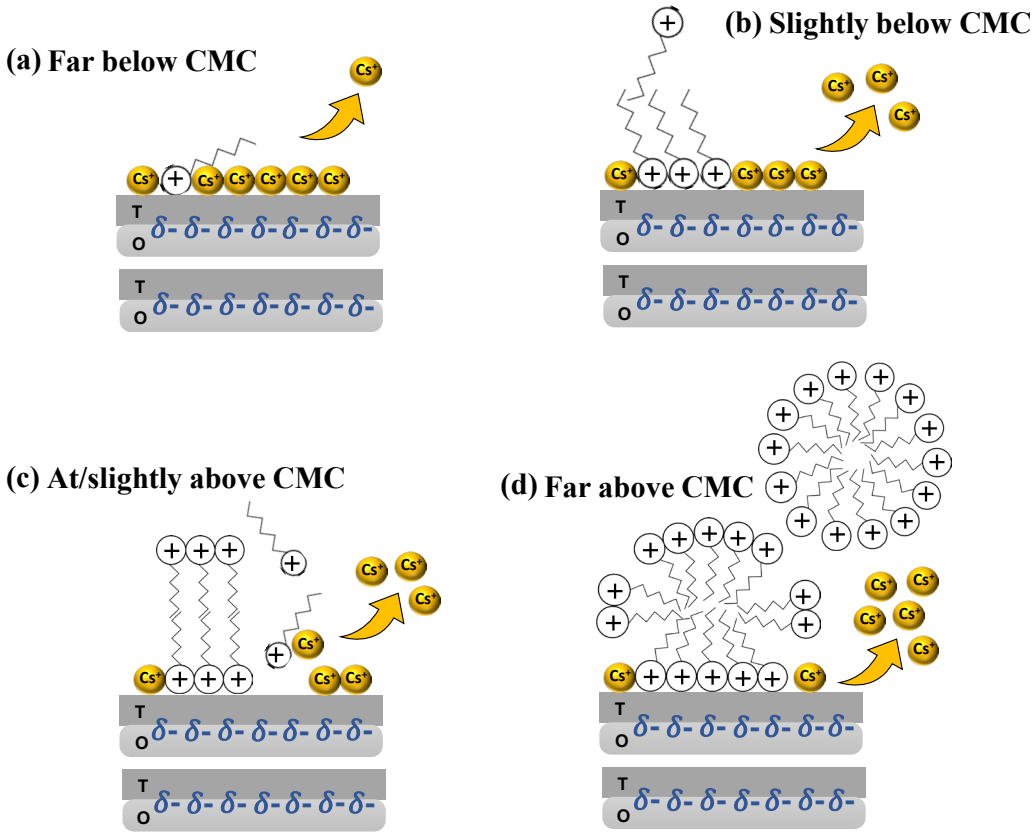

Fig. 3. Schematic diagram of Cs desorption from kaolinite based on micellar formation

References

1 H. Mukai, T. Hatta, H. Kitazawa, H. Yamada, T. Yaita and T. Kogure, *Environ. Sci. Technol.*, 2014, **48**, 13053–13059.

2 N. Kozai, T. Ohnuki, M. Arisaka, M. Watanabe, F. Sakamoto, S. Yamasaki and M. Jiang, *J. Nucl. Sci. Technol.*, 2012, **49**, 473–478.

3 C. Willms, Z. Li, L. Allen and C. V. Evans, *Appl. Clay Sci.*, 2004, **25**, 125–133.

4 B. H. Kim, C. W. Park, H.-M. Yang, B.-K. Seo, S.-J. Park and K.-W. Lee, *J. Nucl. Fuel Cycle Waste Technol.*, 2017, **15**, 27–34.

5 S. M. Park, J. G. Kim, H. Bin Kim, Y. H. Kim and K. Baek, *Environ. Geochem. Health*, 2021, **43**, 3263–3272.

○ **Other theses published in academic research journals**

1. Wijayanto, H., Tsujimoto, M., Basuki, T. & Nakashima, S. Comparison of cationic surfactant and acid solution for remediation of actual ^{137}Cs-contaminated soil from Fukushima Prefecture. *AIP-CP.* **2381**, 020107 (2021).

2. Wijayanto, H. & Nakashima, S. Influence of head group structure of cationic surfactants on the desorption of cesium from clays and clay minerals. *Appl. Clay Sci.* **228**, 106649 (2022).