Production method of environmental tracer ^{132}Cs by accelerator-based neutron

Tadahiro Kin[1,*], *Naoto* Araki[1], *Md Kawchar Ahmed* Patwary[1], *Katsumi* Aoki[1], *Kosuke* Yoshinami[1], *Masaya* Yamaguchi[1], *Masatoshi* Itoh[2], and *Yukinobu* Watanabe[1]

[1]Department of Advanced Energy Engineering Science, Kyushu University, Japan
[2]Cyclotron and Radioisotope Center, Tohoku University, Japan

Abstract. Cesium-132 has proposed as an alternative tracer of ^{137}Cs for environment study on radioactive cesium dynamics released by a nuclear power plant accident. In the present study, we conducted a production experiment of the ^{132}Cs by means of accelerator-based neutron method to investigate production amount and radioactive purity. A 12-g Cs_2CO_3 sample was irradiated by the accelerator-based neutron via the C(*d*,*n*) reactions by 1.2 μA of 30-MeV deuterons. As a result, 102 kBq/g of ^{132}Cs was obtained with higher than 98.5% radioactive purity. Following that, a feasibility study of cesium dynamics measurement in andosol soil was performed. We found distribution of absorption of cesium in andosol soil can be clearly measurable by the produced ^{132}Cs tracer.

1 Introduction

Cesium-137 has been known as the most problematic nuclide in a nuclear accident. For instance, in Fukushima Daiichi Nuclear Power Plant Accident, large amounts of radioactive nuclides including the ^{137}Cs were released into the environment [1–3]. There were three dominant nuclides; ^{137}Cs, ^{134}Cs, and ^{131}I just after the accident. Among all, ^{137}Cs is the dominant radiation source of external exposure in the environment in these days because of its long physical half-life ($T_{phys1/2}$ = 30 y) and ecological half-life ($T_{eco1/2}$ = 294±153 y [4]). Therefore, huge effort has been devoted to the cesium dynamics in the environment to reveal residual distribution [5–7] and pollution of agricultural crops [8].

Especially for the cesium distribution study, short-time dynamics in soil is very important to know the mechanism of cesium absorption. Takeda et al. reported that almost all of cesium absorbed to soil within a few days after the contamination [9]. The authors measured amount of water extractive cesium from andosol soil with ICP-MS method, and as a result, only 1.3% of cesium can be extracted after 4 hours from Cs contamination. In other words, almost all of the cesium is rapidly absorbed into soil within the 4 hours. After that, rest of the cesium has been slowly absorbed and the absorption ratio reaches to 99.98% around 200 days after the contamination. The absorption ratio never changes after that at least for 3 years. In summary, contribution of longtime distribution is less than 2% and dominant absorption phenomena are finished in only a few days.

Nevertheless, even for in-laboratory experiment, long half-life radioactive cesium; ^{137}Cs has been using as an environment tracer, because no short half-life radioactive cesium is available at this moment. Moreover, using unsealed ^{137}Cs source requires careful management for long time to avoid environmental pollution. Although such difficulties are existing, it is clear that further study on cesium dynamics in the environment is important for Fukushima enviroment recovery.

To encourage this study, Nagai et al. proposed ^{132}Cs tracer made by using accelerator-based neutrons [10]. The ^{132}Cs tracer has the following properties (See also Figure 1);

1) Cs-132 emits 668-keV gamma ray by 98% emission probability.
2) The half-life of ^{132}Cs is 6.5 days, which is drastically shorter than that of ^{137}Cs.
3) Producible via the (*n*,2*n*) reactions of ^{133}Cs, which has 100% of natural abundance, by means of accelerator-based neutron method [11].

The ^{132}Cs has shorter half-life compared with ^{137}Cs and it is sufficiently long to measure the important short time dynamics of radioactive cesium in soil. Moreover, the gamma-ray energy emitted from ^{132}Cs is close to that from ^{137}Cs; measurement results using ^{132}Cs require no gamma-ray energy dependent correction such as detection efficiency or the G(E) function. However, further study has not been conducted both for the production and tracer demonstration experiments in the past.

Therefore, the objectives of this paper are, actual production of the ^{132}Cs tracer by means of the accelerator-based neutron method, and feasibility study of the produced tracer to survey cesium absorption in soil.

*e-mail: kin@aees.kyushu-u.ac.jp

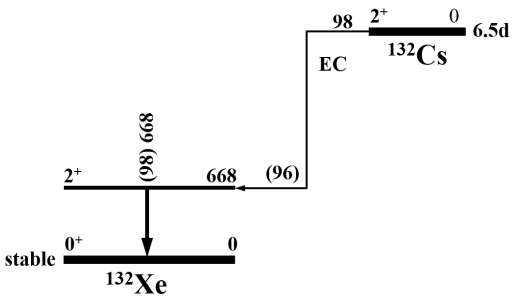

Figure 1. Decay sheme of ^{132}Cs. Cesium-132 decays by electron capture and populates to 1st excited state of ^{132}Xe with 96%. Following that, a 668-keV gamma ray is emitted in de-excitation with almost 100% emission probability.

2 Cesium-132 Production by Means of Accelerator-based Neutron Method

2.1 Cesium-132 Production Experiment

Cesium-132 production experiment was conducted at Cyclotron and Radioisotopes Center (CYRIC) in Tohoku University, Japan. As shown in Figure 2, deuterons were accelerated by AVF930-type cyclotron to 30 MeV and bombarded on a thick carbon target (ϕ40 mm $\times$ 4 mm^t) to generate neutrons via the C(d,n) reactions. The neutrons irradiated a sample of 12-g Cs_2CO_3 to produce ^{132}Cs via the ^{133}Cs(n,2n) reactions. During the irradiation, 1.2 μA deuteron beam was provided for 2 hours.

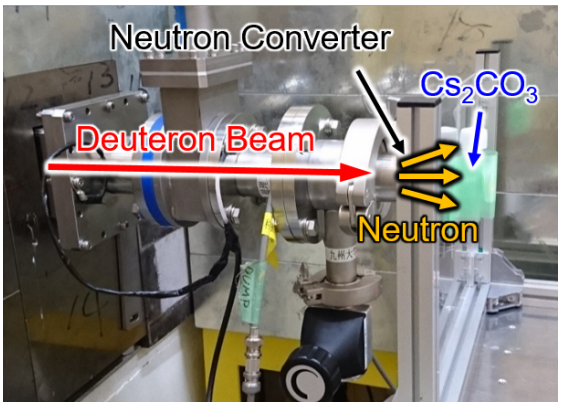

Figure 2. Irradiation system of ^{132}Cs production. Deuterons are provided from right side and bombard on a neutron converter made of carbon installed inside the end of a vacuum tube. Following that neutrons are produced via the C(d,n) reactions and irradiate a Cs_2CO_3 sample to produce ^{132}Cs via the ^{133}Cs(n,2n) reactions.

After the irradiation, gamma rays emitted from the sample were measured by HP Ge detector (ORTEC, GMX50P4-83-RB-B-PL) to determine production amount of ^{132}Cs and radioactive byproducts.

2.2 Results and Discussion

As shown in Figure 3, photo peaks of gamma rays from ^{132}Cs can be observed clearly compared with those of byproducts, ^{134m}Cs and ^{130g}I. As a result, 102 kBq/g of ^{132}Cs was produced during the irradiation, and we found only less than 1.5% of activity is included in the irradiated sample as the byproduct component. The half-lives of the byproducts are 12 h and 3 h for ^{130g}I and ^{134m}Cs, respectively. In addition, because ^{134m}Cs decays to ^{134g}Cs which has 2 y of half life, the activity of the daughter is significantly small. Therefore, we concluded the produced ^{132}Cs has enough quality as an environment tracer. Further quality, such as carrier-added effect and chemical form of the sample, will be discussed in the forthcoming paper.

3 Demonstration of Cesium Distribution Measurement with ^{132}Cs Tracer

3.1 Demonstration Experiment of ^{132}Cs Tracer

A demonstration experiment of the ^{132}Cs tracer produced by means of the accelerator-based neutron method was conducted. We sampled andosol soil at Fukushima in Japan. The andosol soil is widely used for agricultural land in Japan, and it is dominant species of soil also in Fukushima. The sampled soil is not contaminated by fallout released in Fukushima Diichi Nuclear Power Plant Accident. The soil was packed into a acryl cylinder having 3.1 cm in diameter and 15 cm in height. Four packed samples were prepared, because they packed by hand and sample non-uniformity was considered.

At first, we dissolved the irradiated Cs_2CO_3 sample into 1 L of distilled water. As a result, the aqueous solution had about 1.2-kBq/mL ^{132}Cs. Next, 50-mL of the aqueous solution including about 60-kBq ^{132}Cs was flowed into the four andosol soil samples to distribute cesium.

Finally, we measured the gamma rays emitted from ^{132}Cs by an NaI(Tl) detector in 1-cm step from surface to survey the cesium distribution in the andosol soil sample. Figure 4 shows a setup of the cesium distribution measurement.

3.2 Results and Discussion

As shown in Figure 5, resultant cesium distributions of the four andosol soil samples (A, B, C, and D) give similar tendency. It means the samples were uniformly prepared. Almost all of cesium was absorbed within 5 cm from surface of the samples, and this behavior is consistent with the result reported in the previous works [5–7]. Figure 6 shows the distribution comparison with the past result by Ohno et al. [7]. In the past result, the authors measured distribution of ^{137}Cs fallout of Fukushima Daiichi Nuclear Power Plant Accident in brown earth soil just after one month of the accident. Both distributions have similar tendency, but in the past result, cesium distributed deeper than the present one. The difference is caused by the types of soil.

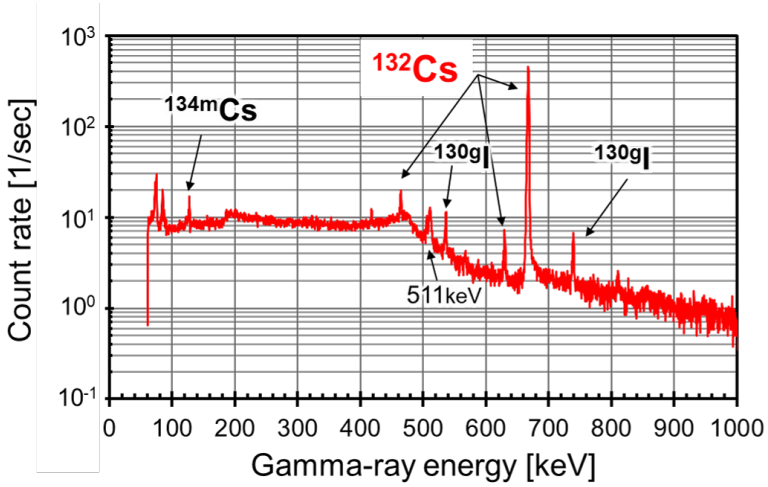

Figure 3. Gamma-ray spectrum obtained by measurement of the irradiated sample, Cs_2CO_3. Gamma-ray peaks from ^{132}Cs are clearly observed, and gamma-ray intensities of byproducts, ^{134m}Cs and ^{130g}I are less than 1.5% compared with that of ^{132}Cs.

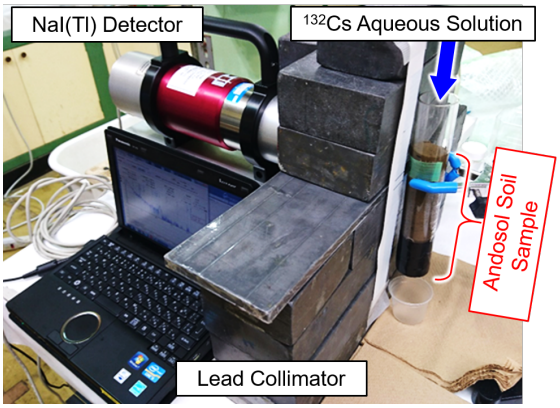

Figure 4. Detection setup of cesium distribution measurement with NaI(Tl) detector. Aqueous solution including ^{132}Cs was flowed from surface of the cylindrical andosol soil sample, and absorbed. To survey the distribution of cesium, gamma rays from ^{132}Cs was measured with NaI(Tl) detector which equiped lead collimator.

Table 1. Component of brown earth and andosol. The andosol includes higher density of clay which has strong cesium absorption ability than that of the brawn earth. Thus, the andosol absorbs cesium shallower region from the surface that contamination occurred.

Soil	Clay	Silt	Fine Sand	Coarse Sand
Brown Earth	12%	12%	26%	49%
Andosol	40%	24%	17%	19%

The brown earth is widely known as a soil species which has higher transmittance of cesium compared with other soil species such as andosol. That is because the brown earth include low density of clay as shown in Table 1. Because the clay is the dominant cesium absorber, the brown earth cannot store cesium around the shallow region from its surface that contamination occurred. The present result shows consistency with this known property.

Therefore, we concluded that the ^{132}Cs tracer produced by the accelerator-based method can be a promising candidate of an alternative environmental tracer of ^{137}Cs one.

4 Conclusion

For the environment recovery from contamination by Fukushima Daiichi Nuclear Power Plant Accident, environmental cesium dynamics study plays an important role. Nagai et al. has proposed ^{132}Cs as a new tracer [10] to encourage the studies, but no further studies has been conducted.

To extend this study, we performed a production experiment of ^{132}Cs by the accelerator-based method at Cyclotron and Radioisotope Center, Tohoku University, Japan. As a result, we found ^{132}Cs tracer having sufficient amount and radioactive purity (102 kBq/g and 98.5%) can be produced by the C(d,n) neutrons induced by 1.2 μA of 30-MeV deuteron beam.

Following the production, we conducted a demonstration experiment as the tracer by using the produced ^{132}Cs tracer. The irradiated sample was dissolved into distilled water and it flowed into a cylindrical shape of andosol soil samples. Cesium distribution in the andosol sample was measured in 1-cm step with NaI(Tl) detector equipping a lead collimator. Measured distribution was compared with the result by Ohno et al. [7] and we found the present result gives same performance and consistent.

In conclusion, the ^{132}Cs tracer produced by the accelerator-based method is a promising candidate as an alternative tracer of the ^{137}Cs. In the future, the tracer will be applied to the further studies on cesium dynamics in the environment, such as development of cesium removal method, absorption time, and different behavior for many types of soil.

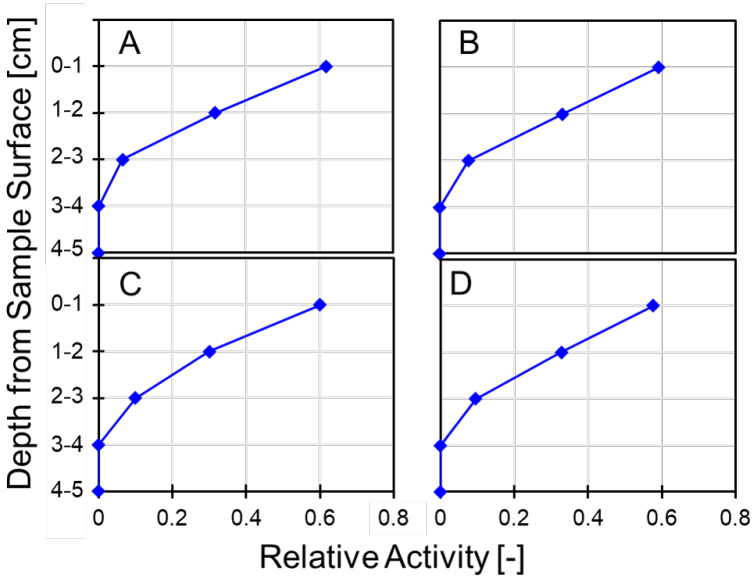

Figure 5. Results of cesium distribution measurement. Four cylindrical andosol soil sample were contaminated from their surface by 50-mL aqueous solution including about 60 kBq of ^{132}Cs produced by the accelerator-based neutron method. The photo peak of 668 keV was measured by NaI(Tl) detector which equip lead collimator to measure the distributions. We can measure that almost all of cesium was absorbed within 5 cm from the surface of the sample by using the proposed tracer, ^{132}Cs.

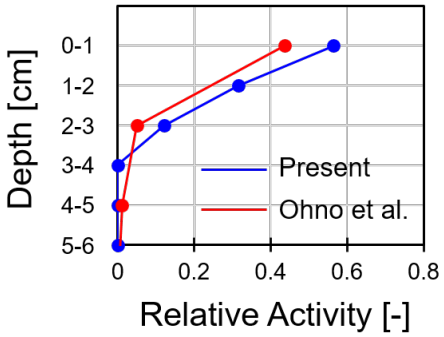

Figure 6. Comparison of cesium absorption distribution with the result by Ohno et al. Ohno et al. measured distribution of fallout ^{137}Cs of Fukushima Daiichi Nuclear Plant Accident for brown earth after one month of the contamination. Brown earth is known to have higher transmittance of cesium compared with andosol, and it follows the result.

Acknowledgements

The andosol soil was provided by Fukushima Agricultural Tecchnology Centre, Japan. I would like to thank Y. Suzuki and M. Sato for their kind effort to provide it. The authors are grateful to accelerator operators of CYRIC, Tohoku University, Japan to provide high quality beam during the irradiation.

References

[1] K. Buesseler, M. Dai, M. Aoyama, C. Benitez-Nelson, S. Charmasson, K. Higley, V. Maderich, P. Masqué, P.J. Morris, D. Oughton et al., Annual Review of Marine Science **9**, 173 (2017), pMID: 27359052

[2] M. Chino, H. Nakayama, H. Nagai, H. Terada, G. Katata, H. Yamazawa, Journal of Nuclear Science and Technology **48**, 1129 (2011)

[3] T.J. Yasunari, A. Stohl, R.S. Hayano, J.F. Burkhart, S. Eckhardt, T. Yasunari, **108**, 19530 (2011)

[4] G. Prohl, S. Ehlken, I. Fiedler, G. Kirchner, E. Klemt, G. Zibold, Journal of environmental radioactivity **91**, 41 (2006)

[5] N. Imamura, M. Komatsu, S. Ohashi, S. Hashimoto, T. Kajimoto, S. Kaneko, T. Takano, Scientific Reports **7**, 8179 (2017)

[6] N. Matsuda, S. Mikami, S. Shimoura, J. Takahashi, M. Nakano, K. Shimada, K. Uno, S. Hagiwara, K. Saito, Journal of Environmental Radioactivity **139**, 427 (2015)

[7] T. Ohno, Y. Muramatsu, Y. Miura, K. Oda, N. Inagawa, H. Ogawa, A. Yamazaki, C. Toyama, M. Sato, GEOCHEMICAL JOURNAL **46**, 287 (2012)

[8] S. Uchida, K. Tagami, Journal of Radioanalytical and Nuclear Chemistry **273**, 205 (2007)

[9] A. Takeda, H. Tsukada, A. Nakao, Y. Takaku, S. Hisamatsu, Journal of Environmental Radioactivity **122**, 29 (2013)

[10] Y. Nagai, H. Makii, S. Namiki, O. Iwamoto, N. Iwamoto, H. Sawahata, Journal of the Physical Society of Japan **81**, 085003 (2012)

[11] Y. Nagai, K. Hashimoto, Y. Hatsukawa, H. Saeki, S. Motoishi, N. Sato, M. Kawabata, H. Harada, T. Kin, K. Tsukada et al., Journal of the Physical Society of Japan **82**, 064201 (2013)