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Author(s)	Nakasone, Shunya; Yokoyama, Sumi; Takahashi, Tomoyuki; Ota, Masakazu; Kakiuchi, Hideki; Sugihara, Shinji; Hirao, Shigekazu; Momoshima, Noriyuki; Tamari, Toshiya; Shima, Nagayoshi; Atarashi-Andoh, Mariko; Fukutani, Satoshi; Ishimine, Akinobu; Furukawa, Masahide; Tanaka, Masahiro; Akata, Naofumi
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Preliminary Investigation of Pretreatment Methods for Liquid Scintillation Measurements of Environmental Water Samples Using Ion Exchange Resins^{*)}

Shunya NAKASONE, Sumi YOKOYAMA¹⁾, Tomoyuki TAKAHASHI²⁾, Masakazu OTA³⁾, Hideki KAKIUCHI⁴⁾, Shinji SUGIHARA⁵⁾, Shigekazu HIRAO⁶⁾, Noriyuki MOMOSHIMA⁷⁾, Toshiya TAMARI⁷⁾, Nagayoshi SHIMA⁷⁾, Mariko ATARASHI-ANDOH³⁾, Satoshi FUKUTANI²⁾, Akinobu ISHIMINE, Masahide FURUKAWA, Masahiro TANAKA^{8,9)} and Naofumi AKATA¹⁰⁾

University of the Ryukyus, 1 Senbaru, Nishihara-cho, Nakagami-gun, Okinawa 903-0213, Japan

¹⁾*Fujita Health University, 1-98 Dengakugakubo, Kutsukake-cho, Toyoake, Aichi 470-1192, Japan*

²⁾*Kyoto University, 2 Asashiro-Nishi, Kumatori-cho, Sennan-gun, Osaka 590-0494, Japan*

³⁾*Japan Atomic Energy Agency, 2-4 Shirakata, Tokai-mura, Naka-gun, Ibaraki 319-1195, Japan*

⁴⁾*Institute for Environmental Sciences, 1-7 Ienomae, Obuchi, Rokkasho-mura, Kamikita-gun, Aomori 039-3212, Japan*

⁵⁾*Kyushu University, 744 Motooka, Nishi-ku, Fukuoka 819-0395, Japan*

⁶⁾*Institute of Environment Radioactivity, Fukushima University, 1 Kanayagawa, Fukushima, Fukushima 960-1296, Japan*

⁷⁾*Kyushu Environmental Evaluation Association, 1-10-1 Matsukadai, Higashi-ku, Fukuoka, Fukuoka 813-0004, Japan*

⁸⁾*National Institute for Fusion Science, 322-6 Oroshi-cho, Toki, Gifu 509-5292, Japan*

⁹⁾*SOKENDAI (The Graduate University for Advanced Studies), 322-6 Oroshi-cho, Toki, Gifu 509-5292, Japan*

¹⁰⁾*Institute of Radiation Emergency Medicine, Hirosaki University, 66-1 Hon-cho, Hirosaki, Aomori 036-8564, Japan*

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This paper presents an evaluation of a rapid pretreatment method for tritium analysis, that uses ion exchange resins (the batch method). The standard water samples were prepared with river water collected in Okinawa, Japan. Powdex resin was used in this study, which is an ion exchange resins in the form of powder beads. First, 100 mL of the standard sample was stirred with the cation exchange (PCH) and anion exchange (PAO) resins, as well as with activated carbon. To determine the optimal stirring time, we tested stirring times of 5, 10, 20 and 30 min. Additionally, to determine a suitable amount of ion exchange resin for environmental water samples, variations in the amount of resin and activated carbon were tested. Under the conditions of Experiment-3 (0.3 g, 0.3 g and 0.6 g of PCH, PAO and activated carbon, respectively) and with a stirring time of 5 min, electrical conductivity values of less than 1 mS m^{-1} were achieved. These results demonstrate the effectiveness of the removal of impurities in the sample water by ion exchange resins and confirm that such an effect can be achieved during a short period of time (i.e., in 5 min).

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1. Introduction

Tritium (^3H) is a radioisotope of hydrogen that decays into ^3He with a half-life of 12.32 years, while emitting a maximum electron energy of 18 keV. Tritium is produced naturally in the upper atmosphere by the nuclear interactions between secondary cosmic rays and nitrogen or oxygen atoms [1]. Nuclear weapon tests have released large amounts of anthropogenic tritium into the atmosphere in the Northern Hemisphere [2]. Moreover, anthropogenic tritium is released from nuclear power reactors and nuclear reprocessing facilities. In the future, nuclear fusion reac-

tors will have a large inventory for use as fuel [3].

Tritium was released along with various radionuclides into the environment by the Fukushima Daiichi Nuclear Power Plant accident of March 2011. In the Fukushima Daiichi Nuclear Power Plant, contaminated water (i.e., water containing tritium) is being stored in large quantities [4]. This tritium water will be diluted to less than the regulatory lower limit level and released into the ocean in the future [2]. Presently, it is necessary to perform regular monitoring to evaluate the environmental influence of the tritium released into the ocean tritium. Thus, a rapid data acquisition procedure would be highly desirable. In addition, in the nuclear fusion facilities studied as a large-scale energy source, a large quantity of tritium is handled.

author's e-mail: k198602@eve.u-ryukyu.ac.jp

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During the operations, steady releases of tritium are collected by outside facilities, and concentrations are assumed to be within the regulatory level. Given all of these releases, it would be beneficial to establish a model to better understand the behavior of tritium released into the environment and to improve the analysis method for environmental samples. At present, research is being conducted to improve the tritium transfer model based on a simple compartment model, establish practical parameters, and subsequently estimate tritium transfers in the environment [5]. This paper describes the progress that has been made toward developing new analytical methods in these research fields.

Highly precise analysis for tritium in water samples with the liquid scintillation counting (LSC) method necessitates the removal of impurities such as organic matter and other types of dissolved matter from the water samples to reduce chemical quenching. Generally, a distillation method is adopted as a pretreatment for samples subjected to tritium. The distillation method is, however, a time-consuming process, i.e., it takes 12 hours to remove impurities from 1 liter of water.

The aim of this study was to evaluate a rapid pretreatment method for tritium analysis with ion exchange resins (the batch method).

2. Materials and Methods

2.1 Samples

In this study, standard water samples were prepared with inland water. The inland water was collected from Houe River (26°8'12.38"N, 127°41'11.45"E) in Okinawa, Japan. This river is relatively rich in organic matter. To further enrich the water, water containing organic matter recovered after the burning of pine needles, was added.

2.2 Chemical reagents

Powdex resins (cation exchange resin: PCH, anion exchange resin: PAO, Graver Technologies) were used in this study. The exchange capacities of PCH and PAO were 4.8 and 4.0 meq dry-g⁻¹, respectively. Activated carbon (GW10/32, AS ONE) was also used to remove the impurities within the water samples to reduce chemical quenching.

2.3 Experimental procedure

First, each ion exchange resin was pretreated with deionized distilled water and stirring. After this step, 100 mL of a standard sample was stirred with the PCH and PAO resins, as well as with activated carbon. The sample was stirred by a stirring machine (RS-4DN, AS ONE) at 1000 rpm. To determine the optimal stirring time, we tested stirring times of 5, 10, 20 and 30 min. Additionally, to determine a suitable amount of ion exchange resin for environmental water samples, we tested various amounts of the cation and anion exchange resins

and activated carbon with the batch method in three experiments: Experiment-1 (0.1 g, 0.1 g, 0.2 g, respectively), Experiment-2 (0.2 g, 0.2 g, 0.4 g, respectively), Experiment-3 (0.3 g, 0.3 g, 0.6 g, respectively). After stirring the samples, the supernatant was filtered through 0.45 μm membrane filters (25CS, ADVANTEC). Then, the pH and electrical conductivity (EC) were measured in the filtered supernatant by using an LAQUAtwin-pH-11B meter (HORIBA, Japan) and a B-771COND meter (HORIBA, Japan), respectively. Additionally, we measured the absorption spectra in the UV range by using a single-beam ultraviolet visible light spectrophotometer (ASUV-3100PC, Japan) to confirm the characteristics of the supernatant.

3. Results and Discussion

The electrical conductivity (EC) values of the supernatant decreased with the increasing stirring time, especially in Experiment-3, where the values decreased substantially (Table 1 and Fig. 1). The tritium analysis manual specifies that the EC after pretreatment should be less than 1 mS m⁻¹, the same as pure water [6]. Under the conditions of Experiment-3 and a stirring time of 5 min, EC values of less than 1 mS m⁻¹ were achieved. The removal level was almost the same as that in the distillation method. However, the procedure time of the distillation method was about 12 h. Thus, the results show that, it is feasible to shorten the pretreatment time by using ion exchange resins.

The pH showed a decreasing tendency until the stirring time of 5 min, and afterwards, the pH hardly changed (Table 1 and Fig. 2). It can be presumed that the reactions between the ion exchange resins and the dissolved ions did not begin readily until the stirring time of 5 min. In Experiment-3, the EC was at the level of pure water, although the pH was in the range of 4.0 to 4.3. This was caused by the dissolution of atmospheric carbon dioxide (CO₂), which made the sample become acidic immediately after stirring. From the results on the EC and pH, we could confirm that pretreatment method successfully removed the impurities in the sample water and that such

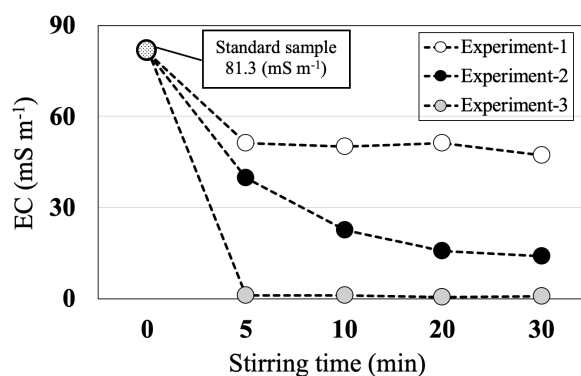


Fig. 1 Declining trends in the electrical conductivity with increasing stirring time.

Table 1 Summary of the parameters for experiments conducted with the batch method.

	Standard sample	Distillation	Experiment-1				Experiment-2				Experiment-3			
			5 min	10 min	20 min	30 min	5 min	10 min	20 min	30 min	5 min	10 min	20 min	30 min
pH	8.3	3.9	7.1	7.2	7.2	6.5	5.9	6.5	6.2	6.2	4.3	4.2	4.1	4.0
EC (mS m ⁻¹)	81.3	0.77	51.3	50.0	51.3	47.1	39.8	22.6	15.8	14.0	0.94	0.97	0.63	0.82

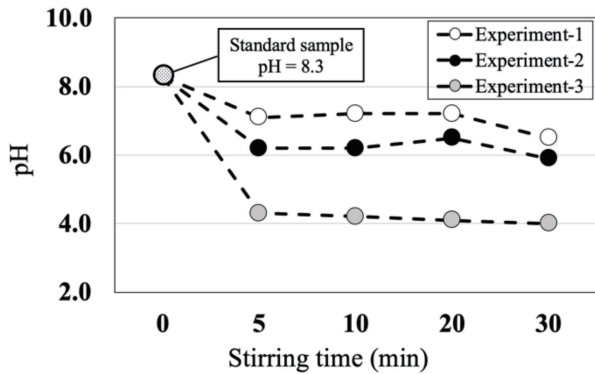


Fig. 2 Changes in pH with increasing stirring time for each experiment.

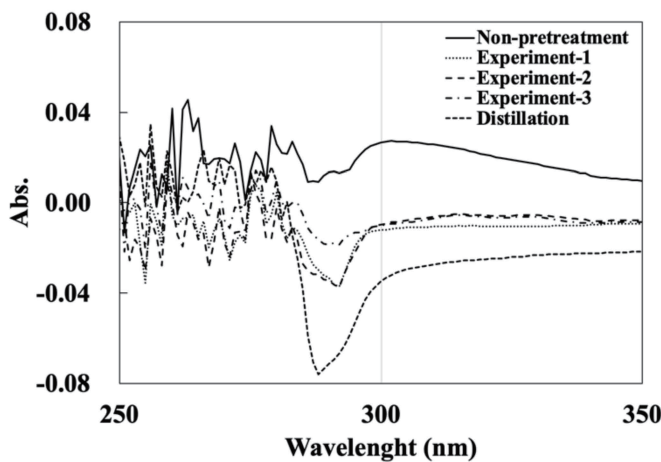


Fig. 3 Absorption spectra in the UV range of recovered water (stirring time; 5 min).

an effect was achievable within a short period of time (i.e., in 5 min).

Lastly, we measured the absorption spectra in the UV range to confirm whether organic matter was included in the collected water. It is well known that UV-Vis absorption hardly occurs in pure water at 250 nm or more. There-

fore, absorption at more than 250 nm is related to organic materials [7]. In this study, the absorption was measured from 280 to 300 nm in all water samples (Fig. 3) and there were no marked absorption peaks in the UV range of interest (250 - 300 nm).

The ion exchange resin samples had lower absorbances than the non-pretreatment samples. From these results, it was confirmed that the dissolved organic matter was adsorbed on and removed by the activated carbon.

4. Summary

We evaluated a rapid ion exchange resin method (the batch method) for pretreating environmental water samples prior to tritium analysis. Appropriate stirring times and amounts of ion exchange resin and activated carbon for environmental water samples were determined. After applying the batch method, the removal of impurities from the sample water was confirmed and such an effect was found to be achievable over a much shorter period of time than that required by the conventional distillation method. As a future task, it is necessary to review the optimal stirring time and amount of resin, e.g., 5 min and 0.3 g, 0.3 g, 0.6 g of PCH, PAO, activated carbon, respectively. Additionally, to apply this method for the rapid pretreatment of tritium measurement, it is necessary to conduct experiments considering the effect of tritium sorption on the ion exchange resins and the activated carbon.

Acknowledgments

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