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The ^{14}C Radioactivity of a NPP Charcoal Sample under a Combustion Condition

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1. Introduction

Carbon fourteen (^{14}C) is produced by a neutron activation of oxygen and nitrogen from a coolant, moderator and structural material in a nuclear reactor. Largely, it is generated by $^{14}N(n, p)^{14}C$ or $^{17}O(n, \alpha)^{14}C$ in a light water reactor or heavy water reactor. It is contained in a charcoal which is used as a filter for nuclear power plants (NPPs). The charcoal more than thousands kilograms is used for NPP annually and some of those will be contaminated by ^{14}C and other radioactive nuclides. ^{14}C from a charcoal, which is a pure beta emitter like ^{3}H, can cause an internal exposure to a human body. Therefore, the charcoals from NPPs are subject to a radioactivity evaluation and then determined for an intermediate/low level radioactive waste repository disposal or to have a clearance in accordance with the national radioactive waste disposal regulation [1]. In the NPPs, ^{14}C is generated as a form of C-compound like $^{14}CO_2$, ^{14}CO and ^{14}C hydrocarbon where ^{14}C hydrocarbon (75 % - 95 %) is dominant at PWR and $^{14}CO_2$ (66 % - 98 %) at PHWR [2]. As one of the methods for ^{14}C determination, the combustion is known to be successful as it can physically extract by oxidation, which requires the combustion duration of several hours generally, and makes it possible to be trapped in an oxide form. Actually, this method is commonly used to combust samples and to trap the separated ^{3}H and ^{14}C from the various kinds of samples such as charcoal, resin, concrete, oil, soil and others [3-7]. Therefore, the combustion condition can be dependent on the sample type and radionuclides to be analyzed. Especially, the number of the samples from NPPs is increasing and faster analysis is required. In this study, the radioactivity of ^{14}C in the charcoal sample used in the NPP is analyzed on the varying combustion time and temperature for finding the efficient conditions.

2. The Experiment

The charcoal sample was combusted by using a high temperature furnace (Raddec Pyrolyser Trio TM) system. It was trapped into the carbosorb in the form of CO_2 and cocktailed with a scintillation solvent. Its radioactivity was measured by using a Liquid Scintillation Counter (1220 Wallac Quantulus TM). Fig.1 shows the schematic of the experimental system for combusting and trapping samples, and measuring their ^{14}C radioactivity. The combustion time was ranged from 30 minutes to 210 minutes every 30 minute. And the combustion temperature varied from 250 ℃ to 600 ℃ in an interval of 50 ℃. The sample with the already known radioactivity, which the radioactivity of the sample was 175 Bq/g ± 8.75 Bq/g with the uncertainty of 95 % confidence level [8-10], was taken. The radioactivity of the sample for the various combustion conditions was calculated as a value of the percentage, which was a ratio of the measured radioactivity to the known activity. On the other hand, the mass of the sample was 0.5 g for the full combustion in the quartz glass pipe with the radius of 1.5 cm of the furnace. Also, the oxygen instead of the air flew from the start of the combustion and the platinum was used as a catalyst.

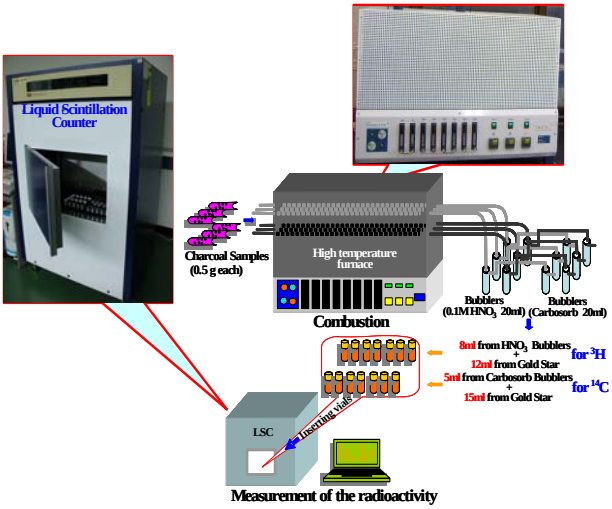

Fig.1. The experimental system for combustion, trap and ^{14}C radioactivity measurement of the samples.

3. Results and Discussion

The ^{14}C radioactivity of the sample was monotonically increased according to the increase of the combustion time at each temperature where the experimental uncertainty was calculated in the 95 % confidence level. The recoveries reached 100 % at a temperature equal to or greater than 450℃ where they were 82.2 %, 92.9 %, 95.5 % and 98.5 at 250 ℃, 300 ℃, 350 ℃ and 400 ℃ in Fig. 2. This experiment represented that the ^{14}C radioactivity was not completely extracted from the sample by simply increasing the combustion time unless the combustion temperature was high enough.

In Fig. 3, the higher the combustion temperature was, the higher the recovery during the first 30 minutes was. Actually, the first 30 minute recoveries were 100 % at a temperature equal to or greater than 450 ℃. Also, in Fig. 4, the ratios of the recovery during the first 30 minutes to the total recovery during whole duration were more than 90 % at each experiment temperature. Especially, the ratio was reached about 100 % in the temperature over 350 ℃.

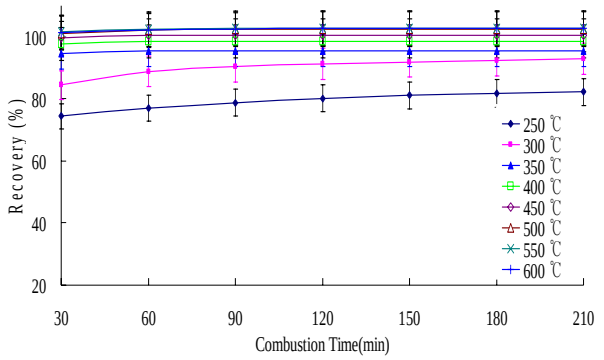

Fig. 2. The ^{14}C recovery on the increasing combustion duration at the various temperatures.

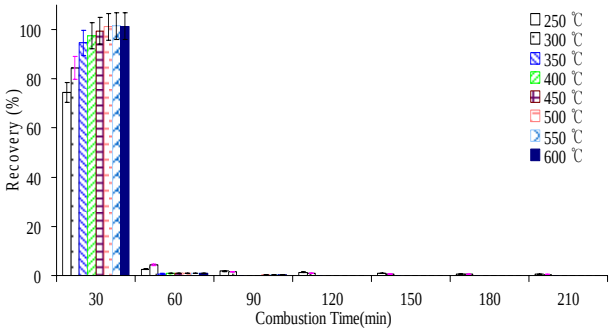

Fig. 3. The ^{14}C recovery on each combustion duration section at the various temperatures.

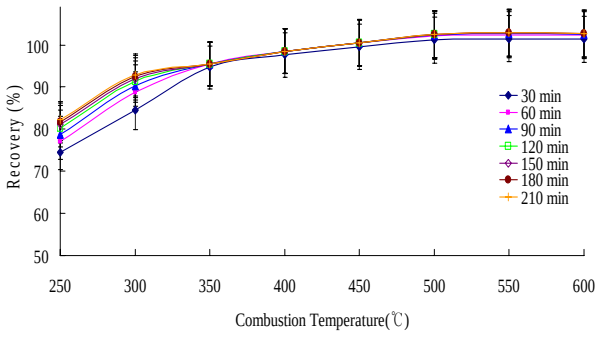

Fig. 4. The ^{14}C recovery on the increasing combustion temperature at the various combustion durations.

After all, it was thought that most of the ^{14}C radioactivity of the sample was extracted during the first 30 minutes. From a practical aspect, when considering a conventional combustion by using a ramped temperature cycle which requires more than five hours, this experiment showed that the time required for the ^{14}C radioactive sample combustion was much reduced under this uniform temperature condition.

4. Conclusions

The recovery of the ^{14}C radioactivity of a charcoal sample used in a nuclear power plant was analyzed by using a uniform temperature combustion method. It was understood that the charcoal had a property whose ^{14}C radioactivity could be completely extracted during the first 30 minutes at a temperature of at least 450 ℃. It was found that the combustion temperature was more important criterion than the combustion duration for the complete extraction of the ^{14}C radioactivity. This study implied that a rapid pretreatment was possible for more NPP charcoal samples.

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