The Radioactivity Characteristics of the NPP Charcoal Sample Contaminated by Carbon-14 – 13531

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ABSTRACT

The radioactivity of ^{14}C-contaminated charcoal sample was analyzed by using a high temperature oxidation and liquid scintillation counting method. The 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. It showed 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. The higher the combustion temperature was, the higher the recovery during the first 30 minutes was. The first 30 minute recoveries were 100 % at a temperature equal to or greater than 450 ℃. 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. It was understood that the temperature was a critical factor for the complete removal of the ^{14}C from the waste sample.

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, α)^{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 [3-6]. 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. 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.

THE EXPERIMENT

Efficiency Calibration of the Detector

The counting efficiency was measured by using an external standard method (SQPE) at different quench level as is represented in Fig. 1. The counting efficiencies were measured to be 54-64 % for ^{14}C.

$$^{14}C$$
$$y = -6.77 * 10^{-4} x^2 + 1.07 x - 3.59 * 10^2$$
$$R^2 = 9.97 * 10^{-1}$$

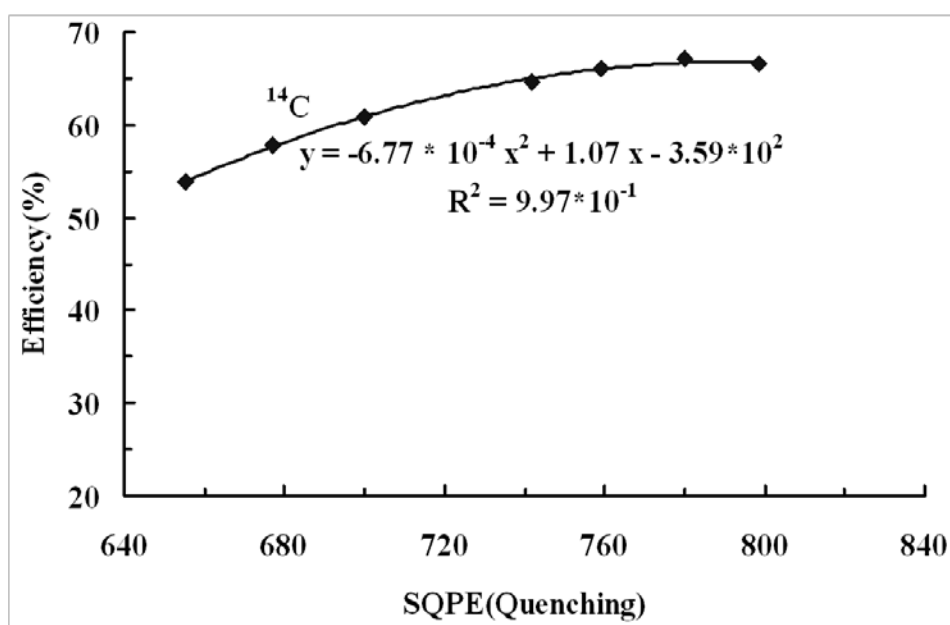Fig. 1. Counting efficiency of ^{14}C for different quench levels (SQP(E))

The Measurement of the Count rates of Samples

The high-temperature furnace system (Raddec Pyrolyser Trio TM) for the oxidation of charcoal samples and the Liquid Scintillation Counter (LSC, 1220 Wallac Quantulus TM) for the sample counting were prepared [3]. Fig. 2 shows the schematic of the experimental system for combusting and trapping samples, and measuring their ^{14}C radioactivity. The vials with the trapped ^{14}C from bubblers after combustion and chemical reaction are placed into the LSC as represented in Fig. 3. A high-energy beta nuclide window mode and low-energy beta nuclide window mode were used for the measurement of ^{14}C in the LSC.
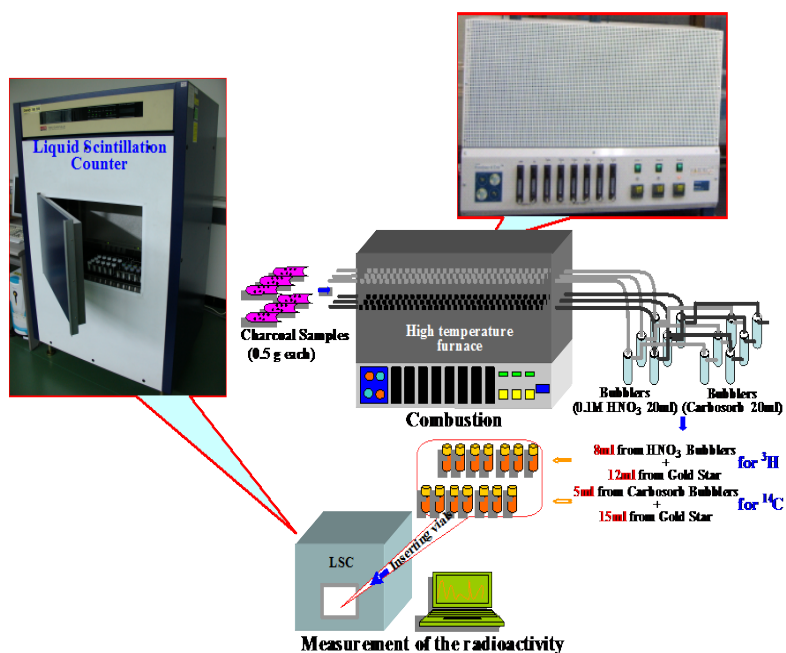
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Fig.2. The experimental system for combustion, trap and ^{14}C radioactivity measurement of the samples.

Fig. 3. The sample vials placed into LSC for measuring

The samples and blank were counted for 90 minutes and 3 cycles. The averages and standard deviation of 3 cyclic counts were calculated. The charcoal sample was combusted in the high temperature furnace 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. 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 [7-9], 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.

Experimental Uncertainty

Some experimental or systematic factors were considered to cause the measurement uncertainty [7-9]. First of all, weighing the samples and bubbler solution could cause experimental errors due to poor measurements. The recovery of the furnace and the efficiency of the LSC could be considered as other factors for the errors as well. For the present analysis, because the measurement errors could be assumed to be independent of each other and the presentation of the radioactivity had a product form as presented in equation (1), its combined relative uncertainty could be expressed in the form of equation (2) through some mathematical process [3, 7-9].

$$A_s = \frac{C-B}{60} \times \frac{100}{E} \times \frac{1}{m} \times \frac{m_f - m_t}{m_s} \times \frac{100}{R} \qquad \text{(Eq. 1)}$$

$$\left(\frac{U_{AS}}{A_S}\right)^2 = \left(\frac{U_{C-B}}{C-B}\right)^2 + \left(\frac{U_E}{E}\right)^2 + \left(\frac{U_m}{m}\right)^2 + \left(\frac{U_{m_f - m_t}}{m_f - m_t}\right)^2 + \left(\frac{U_{m_s}}{m_s}\right)^2 + \left(\frac{U_R}{R}\right)^2 \qquad \text{(Eq. 2)}$$

As = The activity concentration in the sample (Bq/g)

C = The sample count rate (CPM)

B = The background count rate (CPM)

E = The counter efficiency (%)

m = The mass of the bubbler solution taken for analysis (g)

m_f = The final bubbler mass (g)

m_t =   The bubbler tare mass (g)

m_s =   Mass of sample taken (g)

R   =   Furnace recovery (%)

U_{As} =   The uncertainty of the activity concentration in the sample (Bq/g)

U_{C-B} =   The uncertainty of the net count rate (CPM)

U_E =   The uncertainty of the counter efficiency (%)

U_m =   The uncertainty of the mass of bubbler solution taken for analysis (g)

U_{mf-mt} =   The uncertainty of the difference of the final bubbler mass and the bubbler tare mass (g)

U_{ms} =   The uncertainty of mass of sample taken (g)

U_R = The uncertainty of furnace recovery (%)

THE 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. 4. 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. 5, 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. 6, 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 ℃. 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. The furnace recoveries were 100 % ± 4.9 % for ^{14}C.
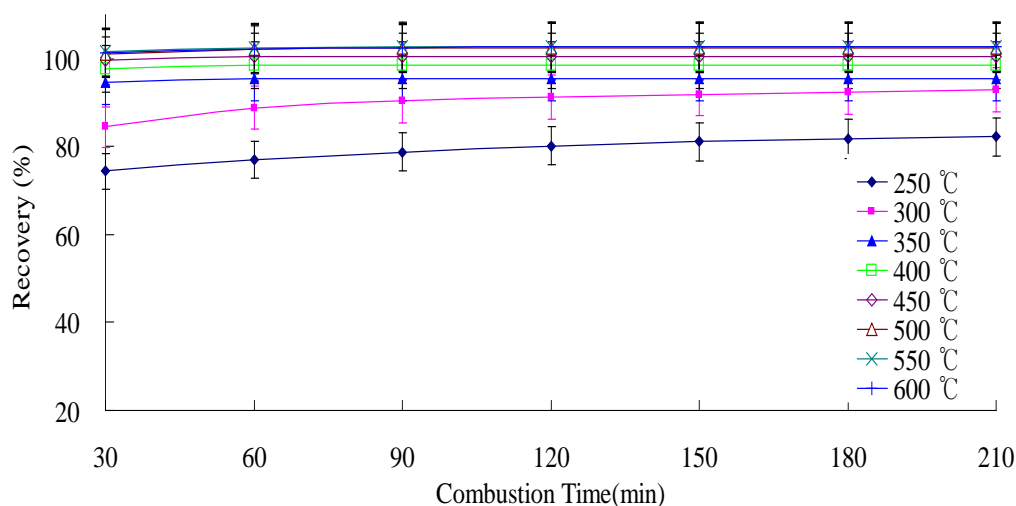
Fig. 4. The ^{14}C recovery on the increasing combustion duration at the various temperatures.
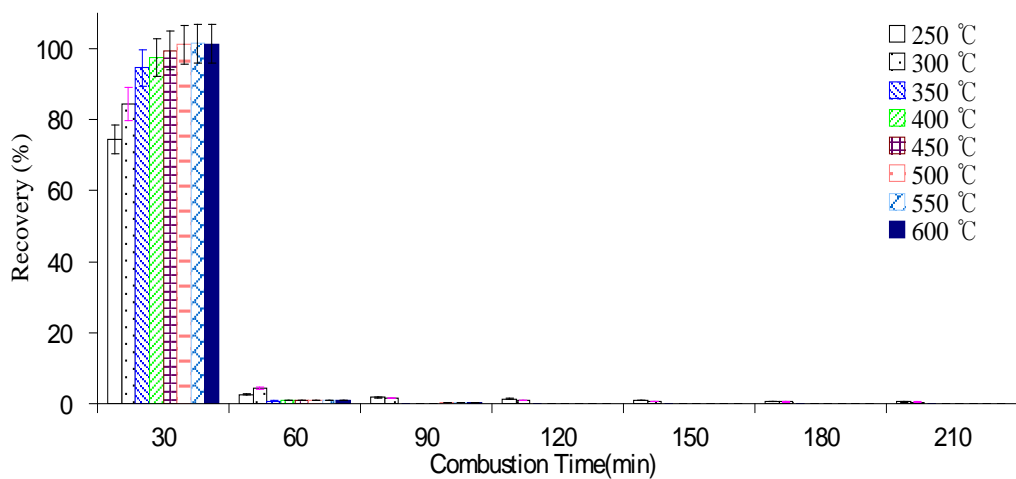

Fig. 5. The ^{14}C recovery on each combustion duration section at the various temperatures.

The background counting rates, which are generally dependent on the counter, the energy range of ^{14}C, the quench level, the vial for the LSC, and the radioactivity level of the blank sample, were 3-4 CPM for ^{14}C, where a polyethylene vial was used. Net count rates were calculated by subtracting the background count rate from the sample count rate. In relation with the experimental uncertainty, it was expected that the background counting rate had little effect on the uncertainty of the radioactivity of the sample.
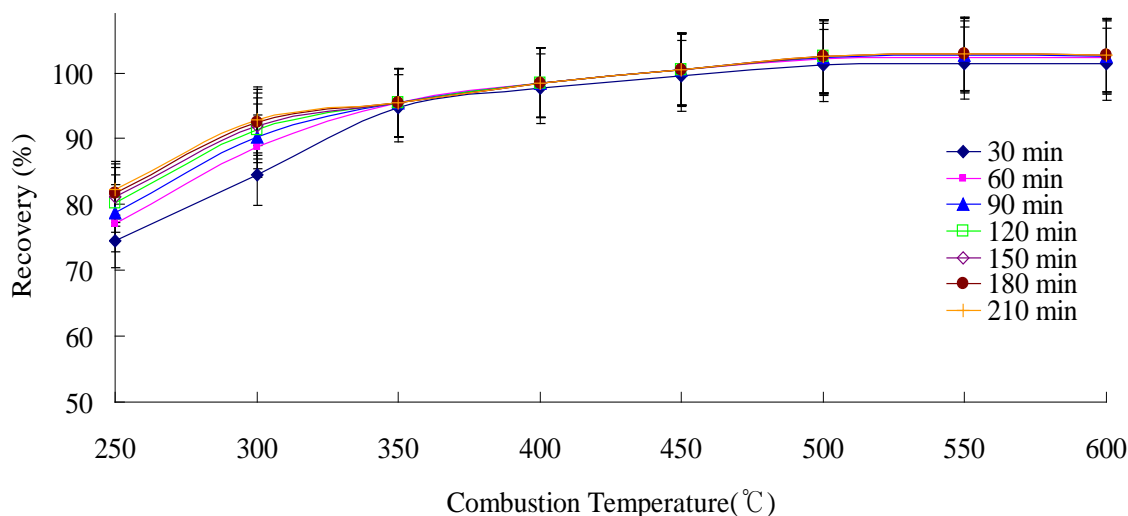
Fig. 6. The ^{14}C recovery on the increasing combustion temperature at the various combustion durations.

As other factors causing an uncertainty in this work, the relative uncertainty of a sample weighing, the mass of a bubbler solution taken for an analysis, and the difference between the final bubbler mass and the bubbler tare mass were 0.01 %, 0.05 %, and 0.03 % respectively, in this experiment. The relative uncertainty of the decay correction and certified ^{14}C (as quoted on certificate) were 0.2 %. The relative uncertainty of the furnace recovery was 4.9 %, and that of the liquid scintillation counter calibration curve was 1.0 %. In addition, the counting uncertainty of the measurement by using an LSC was less than 0.5 %, as was determined its relative standard deviation. Therefore, the combined relative uncertainty was calculated to be less than 5.1 %. While investigating the components of the uncertainty, the uncertainty of the furnace recovery was found to be a dominant factor for causing an experimental uncertainty. In fact, this recovery considerably depended on the condition of the Pt catalyst, temperature, and burning duration. Hence, it was thought that this uncertainty could be remarkably reduced by carrying out an exchange of the catalyst and the maintenance of the suitable temperature and duration.

On the other hand, the minimum detectable activities (MDA) by using these background counting rates were 0.04-0.05 Bq/g for ^{14}C, based on Curie's equation. Curie's equation [9] is defined as equation. (3);

$$\text{MDA} = \frac{2.75 + 4.65\sqrt{N_b}}{t \times E \times R \times m} \qquad \text{(Eq. 3)}$$

where N_b is the count of the background, t is the background counting time (second), E is the counting efficiency, R is a recovery of the furnace, and m is the weight of a sample (g). So, the MDA depends on the background counts, counting time, and the weight of the sample. In this work, the counts of the sample were much higher than those of the blank.

CONCLUSION

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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