

## Analysis of $^{14}\text{CO}_2$ trapped $^{14}\text{C}$ Sorbent, and $^{14}\text{C}$ and $^3\text{H}$ Radioactivity Determination in Resins and Oils from Nuclear Power Plants Using a Combustion Method

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### 1. Introduction

Tritium ( $^3\text{H}$ , T) generated in the heavy water and C-fourteen ( $^{14}\text{C}$ ) originated from the graphite moderator or structural materials of the nuclear power plant can cause acute and/or chronic harmful effects by inhalation and ingestion of these radionuclides owing to their binding affinity toward biomolecules and gas phase [1]. To determine the  $^3\text{H}$  and  $^{14}\text{C}$  radioactivities, the most widely applied technique is ambient pressure decomposition where the sample is thermally oxidised in air or  $\text{O}_2$ -enriched air within a tube furnace assembly [2]. This combustion method provides a very powerful tool to rapidly process many different sample types with a high degree of precision and accuracy [3]. And the method is ideally suited for the single- and dual-label  $^3\text{H}$  and  $^{14}\text{C}$ . Following thermal combustion, the liberated decomposition products are fully oxidized with a catalyst (Pt-alumina) to ensure that  $^3\text{H}$  and  $^{14}\text{C}$  are present only as HTO and  $^{14}\text{CO}_2$  which can be subsequently trapped using dilute acid solutions and  $^{14}\text{C}$  sorbents respectively.  $^{14}\text{C}$  sorbent (Carbo-Sorb<sup>®</sup> E) has been used to trap the  $^{14}\text{CO}_2$ . However, the trapping mechanism has not been analyzed well. In this study, we focused our efforts on the radioactivity determination of  $^3\text{H}$  and  $^{14}\text{C}$  of radwastes such as oils and ion exchange resins from nuclear power plants using a dry oxidation (combustion) method, and study of  $^{14}\text{C}$  trapping mechanism based on FT-IR analysis of  $^{14}\text{C}$  sorbents.

### 2. Materials and Methods

#### 2.1 Equipment and Chemicals

The combustion system (Pyrolyser-6 Trio<sup>™</sup>) was purchased from RADDEC. 0.1 M  $\text{HNO}_3$  was prepared by the dilution of concentric  $\text{HNO}_3$  (Merck).  $^{14}\text{C}$  sorbent (Carbo-Sorb<sup>®</sup> E) was bought from PerkinElmer. Gold Star Multi-Purpose (Meridian) was used as a scintillation cocktail.

#### 2.2 Combustion of Samples

Samples of ion exchange resins and oils were obtained from Nuclear Power Plants in Korea. The equipment for sample combustion method is depicted in Fig. 1. The sample boat including radwastes such as ion exchange resins or oils is placed in the a silica glass work tube

within the sample oxidation zone of the furnace and heated using a preset heating profiles based on experimentally optimized studies (Fig. 2). The combustion gas released from the sample is fully oxidized in a catalysis zone (800 °C). The formed HTO and  $^{14}\text{CO}_2$  are trapped in a 0.1 M  $\text{HNO}_3$  solution and a  $^{14}\text{C}$  sorbent (Carbo-Sorb<sup>®</sup> E) respectively.

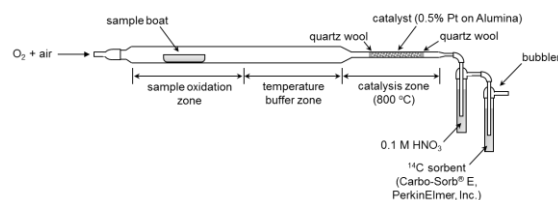


Fig. 1. Schematic diagram of the combustion system.

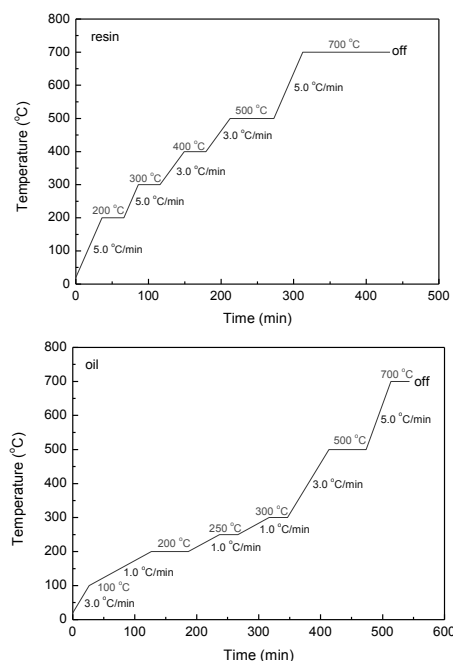


Fig. 2. Temperature profiles for the sample combustion of ion exchange resins and oils.

#### 2.3 Radioactivity Determination of $^{14}\text{C}$ and $^3\text{H}$ by Liquid Scintillation Counter

The 5 mL of the  $^{14}\text{CO}_2$  trapped  $^{14}\text{C}$  sorbent (Carbo-Sorb<sup>®</sup> E) in the bubbler or the 8 mL of the HTO trapped 0.1 M  $\text{HNO}_3$  solution in the bubbler was mixed with 15

or 12 mL Gold Star Multi-Purpose scintillation cocktail in a 22 mL polyethylene vial, respectively. The radioactivity determination of  $^3\text{H}$  and  $^{14}\text{C}$  of the prepared vial samples were carried out using a Quantulus<sup>TM</sup> 1220 Ultra Low Level Liquid Scintillation Spectrometer (PerkinElmer).

### 2.4 FT-IR Analysis of $\text{CO}_2$ Trapped $^{14}\text{C}$ Sorbent

A 10 mL of Carbo-Sorb<sup>®</sup> E contained glass vial was connected with a  $\text{CO}_2$  gas line (flow rate: 1000 mL/min). The FT-IR spectra of the  $\text{CO}_2$  gas trapped Carbo-Sorb<sup>®</sup> E at various elapsed times were obtained using Fourier transform infrared (FT-IR) spectroscopy (Frontier, PerkinElmer). The FT-IR spectra of  $\text{CO}_2$  from real samples trapped Carbo-Sorb<sup>®</sup> E were also analyzed.

## 3. Results and Discussion

### 3.1 Radioactivity Concentrations of $^3\text{H}$ and $^{14}\text{C}$ in radwastes

During the combustion of samples, other volatile nuclides that are produced during irradiation in the nuclear reactor and which therefore have a possibility of existing in the samples, may also be released from the sample and become trapped in the  $^{14}\text{C}$  sorbent (Carbo-Sorb<sup>®</sup> E) and the 0.1 M  $\text{HNO}_3$  solution. No significant contribution of the interfering nuclides was observed in the  $^3\text{H}$  and  $^{14}\text{C}$  spectra of samples (Fig. 3).

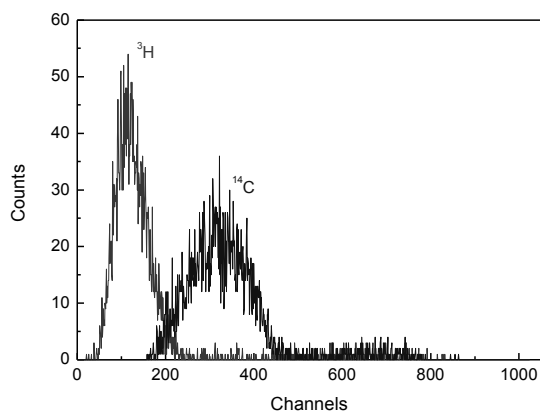


Fig. 3.  $^3\text{H}$  and  $^{14}\text{C}$  spectra of an ion exchange resin from the nuclear power plant.

The analytical results of  $^3\text{H}$  and  $^{14}\text{C}$  an ion exchange resin and an oil from a nuclear power plant are summarized in Table I.

Table I: Analytical Results of  $^3\text{H}$  and  $^{14}\text{C}$  an Ion Exchange Resin and an Oil

	$^3\text{H}$ activity (Bq/g)	$^{14}\text{C}$ activity (Bq/g)
Ion exchange resin	$9.56 \pm 2.95$	$7.21 \pm 2.14$
Oil	$0.04 \pm 0.02$	$0.22 \pm 0.08$

### 3.2 FT-IR study of $\text{CO}_2$ Trapped $^{14}\text{C}$ Sorbent

The FT-IR spectra of the  $\text{CO}_2$  gas trapped Carbo-Sorb<sup>®</sup> E at various elapsed times with 1000 mL/min of  $\text{CO}_2$  flow rate are shown in Fig. 4. Until 10 min, peaks for carbamate groups were observed which are indicating the bonding of the amine group of  $^{14}\text{C}$  sorbent and  $\text{CO}_2$ . However, with the increase of time for  $\text{CO}_2$  blowing into the  $^{14}\text{C}$  sorbent, the peaks for carbamic acid group arose.

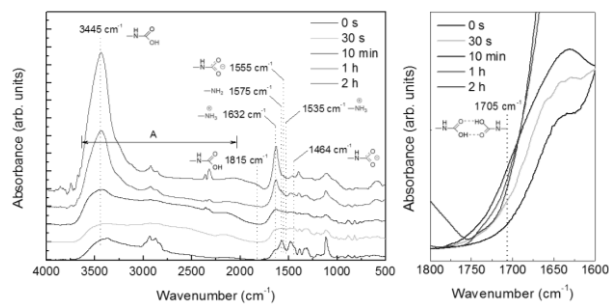


Fig. 4. FT-IR spectra of the  $\text{CO}_2$  gas trapped Carbo-Sorb<sup>®</sup> E.

## 4. Conclusions

$^3\text{H}$  and  $^{14}\text{C}$  radioactivity in ion exchange resins and oils from nuclear power plants were determined by an oxidation (combustion) method. The 0.1 M  $\text{HNO}_3$  solution and the  $^{14}\text{C}$  sorbent trapped the  $^3\text{H}$  and  $^{14}\text{C}$  respectively in the gas from the combustion of samples. All samples were burned without ash in the combustion system. The reaction of  $\text{CO}_2$  and  $^{14}\text{C}$  sorbent was investigated by FT-IR analysis. The study demonstrated the different reaction mechanism according to the  $\text{CO}_2$  concentration. In the FT-IR study, it is clearly confirmed that  $\text{CO}_2$  from the burned 1 g of sample can be trapped in the  $^{14}\text{C}$  sorbent completely. During the reaction of  $\text{CO}_2$  and  $^{14}\text{C}$  sorbent, the temperature and the viscosity of  $^{14}\text{C}$  sorbent increased due to the decrease of enthalpy change and the bonding between each molecules of the sorbent. We expect that our FT-IR study could motivate the development of  $^{14}\text{C}$  sorbent and confirm the  $^{14}\text{C}$  trapping performance of the  $^{14}\text{C}$  sorbent.

## REFERENCES

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