

## Determination of C-14 and H-3 in Radioactive Wastes by Oxidation Method

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### 1. Introduction

C-14 is formed in the upper atmosphere by the interaction between cosmic ray and nitrogen ( $^{14}\text{N}(n,p)^{14}\text{C}$ ) and in Nuclear Power Plant (NPP) via a neutron capture reaction ( $^{17}\text{O}(n,\alpha)^{14}\text{C}$ ,  $^{14}\text{N}(n,p)^{14}\text{C}$ ). H-3 is similarly produced in the atmosphere ( $^{14}\text{N}(n,^{12}\text{C})^3\text{H}$ ) and in the coolant water ( $^6\text{Li}(n,\alpha)^3\text{H}$ ,  $^7\text{Li}(n,n\alpha)^3\text{H}$ ) [1,2,3]. Most of C-14 is found in  $\text{CH}_4$  for organic and  $\text{CO}_3^{2-}$ ,  $\text{HCO}_3^-$  for inorganic form. H-3 is usually formed HTO.

In 1982, U.S. Nuclear Regulatory Commission (NRC) regulated radioactive wastes through 10 CFR 61 [4]. The volatile elements (H-3, C-14, Tc-99, and I-129) in these wastes are the difficult-to-measure (DTM) radionuclides, and need to be measured to an established method.

The new method of wet oxidation has been developed for the separation of C-14 and H-3 in concentrated boric acid waste solution (CB) both simulated and NPP radioactive wastes. Among C-14 wastes, inorganic form is easily oxidized to  $\text{CO}_2$  by the acid, but organic C-14 needs powerful oxidant for an oxidation. Inorganic and organic C-14 is oxidized to  $^{14}\text{CO}_2$  by using the method of wet oxidation. The counting result of C-14 and H-3 is compared with a sample oxidizer.

### 2. Experiments and Result

In this section, experiments for the separation of C-14 and H-3 in the radioactive waste of concentrated boric acid waste solution (CB) are described. Wet oxidation and sample oxidizer (Model A307, Perkin-Elmer) are shown in Figure 1. The sampling of CB for simulated radwaste was prepared a boric acid including the standard. Counting of NPP radioactive wastes was measured and compared with wet oxidation and sample oxidizer. The determination of C-14 and H-3 were counted to Liquid Scintillation Counting (LSC).

#### 2.1. Wet Oxidation

In the reactant flask, 5 g of  $\text{K}_2\text{S}_2\text{O}_8$  and 0.5 g of  $\text{AgNO}_3$  for the oxidant with about 1.0 g of the sample (0.1 mL of  $\text{Na}_2^{14}\text{CO}_3$ , 0.05 mL of  $^{14}\text{C}$ -toluene, and 0.1 mL of HTO) were inserted. And 20 mL of 3 N  $\text{H}_2\text{SO}_4$  was dropped under a He gas. Then  $^{14}\text{CO}_2$  for C-14 was trapped to the absorber (Carbo-Sorb E, Perkin-Elmer) at reflux. After 2 hr, the collection of HTO is distilled to the collector (Figure 1A). The cocktail is Permafluor E<sup>+</sup>

(Perkin-Elmer) for C-14 counting and Ultima-Gold XR (Perkin-Elmer) for H-3.

#### 2.1. Sample Oxidizer

In sample oxidizer (Figure 1B), 0.1 mL of a sample was directly ignited in an ignition flask.  $\text{CO}_2$  gas formed during combustion was trapped to the absorber (Carbo-Sorb E). This solution was mixed with the cocktail (Permafluor E<sup>+</sup>). HTO steam was mixed with Monophase S (Perkin-Elmer).

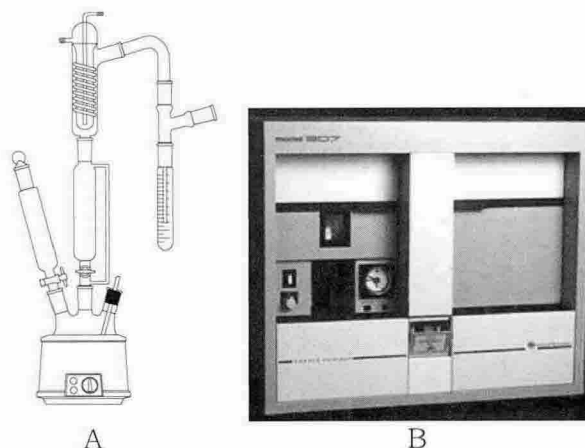


Figure 1. The equipment for oxidation method. (A) wet oxidation equipment and (B) sample oxidizer.

#### 2.3. Determination of C-14 and H-3 Activities

The recovery for the standard and simulated radioactive waste (CB) are measured to 91 for C-14 and 93% for H-3 (Table 1).

Table 1. The recovery of the standard and simulated radioactive waste.

Nuclide	Sample	Real kdpm	Exp. kdpm	%REC	%RSD
C-14	Standard	164	150	91	1.02
	CB <sup>a</sup>	142	130	91	0.62
H-3	Standard	14.3	13.2	93	1.02
	CB <sup>a</sup>	7.3	6.8	93	2.58

<sup>a</sup> CB is simulated concentrated boric acid waste solution with standard C-14 and H-3.

Radioactive waste, concentrated boric acid waste solution (CB), produced in NPP was found that the

activity of C-14 is 8-90 Bq/g and H-3 is 240-288 Bq/g by wet oxidation. In the case of the waste No. 1, A system was measured to 62.1 Bq/g for C-14 and 247 for H-3. The result for sample oxidizer was obtained with 67.5 for C-14 and 217 for H-3, respectively (Figure 2). Data of six NPP wastes (CB) for two methods were similar confirmed within about 10% difference.

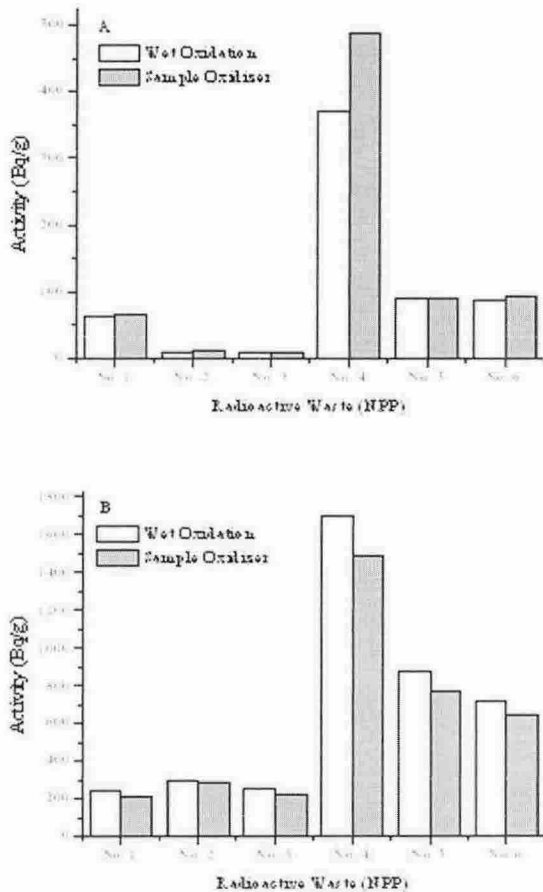


Figure 2. The comparison of the activity for C-14 (A)

and H-3 (B) by using wet oxidation (white bar) and sample oxidizer (gray bar) at NPP radioactive waste (CB).

### 3. Conclusion

In simulated radioactive waste, the result of C-14 and H-3 counting was obtained over 90% by using wet oxidation.

In the case of NPP radwaste, wet oxidation and sample oxidizer were similarly measured to the difference of about 10%.

Wet oxidation method can analyze C-14 and H-3 at the same time and oxidize C-14 for inorganic and organic form.

### REFERENCES

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