

Determination of H-3 and C-14 in Solidified Radioactive Wastes

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1. Introduction

In 1982, U.S. Nuclear Regulatory Commission (NRC) regulated radioactive wastes through 10 CFR 61 [1]. 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.

Immobilization is one of the options for dealing with radioactive waste. The objective of immobilization is to convert radioactive waste into a stable form, which minimizes the probability of radionuclide release to the environment during interim storage, transport and final disposal [2].

The activity of H-3 and C-14 in cement and paraffin drum would be counted to the oxidation method. The measuring method of H-3 and C-14 counting recognized generally two types, sample oxidizer and wet oxidation [3]. Evaporated bottom (EB) and spent resins (SR) would be measured by two types [4]. Then counting for two types was obtained a similar result. But because the measurement of H-3 and C-14 in the drum sample was difficult to sample oxidizer, the separation and measurement of H-3 and C-14 in the drum solidified to cement and paraffin both simulated and NPP radioactive wastes have been tried by wet oxidation method.

2. Experiments and Result

In this section, the experiment for the separation of H-3 and C-14 in the radioactive wastes within the drum solidified to cement and paraffin is described. Counting of radioactive wastes was measured with wet oxidation. The determination of H-3 and C-14 were counted to Liquid Scintillation Counting (LSC) by using the quench correction curve.

2.1. Experiments

In the reactant flask, 5 g of $K_2S_2O_8$ and 0.5 g of $AgNO_3$ for the oxidant with 1.0 g of cement or paraffin added the standard (HTO is 0.1 mL of 7300 dpm/mL and $Na_2^{14}CO_3$ (aq) is 0.1 mL of 7140 dpm/mL) were inserted for the simulated sample. And 20 mL of 3 N H_2SO_4 was dropped under a He gas. Then $^{14}CO_2$ for C-14 was trapped to the absorber (Carbo-Sorb E, Perkin-Elmer) at reflux. After 3 hr, the collection of HTO is distilled to the collector (Figure 1). The cocktail is

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

In the case of NPP sample, the procedure was same such as the simulated sample except using the NPP sample solidified to cement and paraffin instead of the standard and normal cement and paraffin.

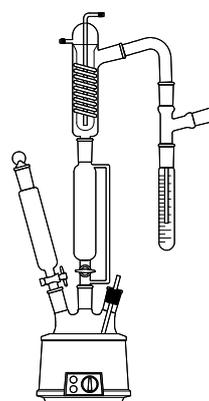


Figure 1. The equipment for wet oxidation method.

2.2. Determination of H-3 and C-14 Activities

The recovery for the simulated radioactive waste (the drum solidified to cement and paraffin) is measured to 93 for H-3 and 91% for C-14 (Table 1).

Table 1. The recovery of the simulated radioactive waste.

Nuclide	Sample	Real dpm	Exp. dpm	% Yield	%RSD
H-3	Cement	730	670	92	1.02
	Paraffin	730	656	90	2.58
C-14	Cement	714	665	93	1.02
	Paraffin	714	663	93	3.62

Radioactive wastes produced in NPP was found that the activity of H-3 was 65-1000 Bq/g and C-14 was 1-300 Bq/g for cement (Table 2). The result of paraffin drum was obtained with 120-1500 for H-3 and 37-470 Bq/g for C-14 (Table 3).

Table 2. The activity of H-3 and C-14 in the drum solidified cement at NPP radioactive waste.

Sample	$\mu Sv/hr g$	Bq/g	
		H-3	C-14

CE-1-SR-U	0.00	339±1	1.4±0.2
CE-1-SR-M	0.01	250±1	0.9±0.1
CE-1-SR-B	0.00	87±4	0.7±0.1
CE-2-EB-U	0.01	290±5	3.3±0.6
CE-2-EB-M	0.01	302±1	2.7±0.4
CE-2-EB-B	0.01	317±4	3.6±0.6
CE-3-EB-U	0.22	68±1	118±12
CE-3-EB-M	0.26	75±2	93±6
CE-3-EB-B	0.14	65±1	52±10
CE-4-EB-U	0.26	174±1	122±7
CE-4-EB-M	0.24	176±9	195±18
CE-4-EB-B	0.24	170±3	307±23
CE-5-EB-U	0.50	679±7	11±0
CE-5-EB-M	0.50	975±20	12±1
CE-5-EB-B	0.45	929±16	5.5±1.9

[2] IAEA, "Management of Waste Containing Tritium and Carbon-14", Technical Reports Series No. 421, 2004.

[3] Packard, "Liquid Scintillation Analysis Science and Technology".

[4] Lee, H. N., *et al.*, "Determination of C-14 and H-3 in Radiation Wastes by Oxidation Method", Proceeding of KNS Autumn Meeting, 2004.

Table 3. The activity of H-3 and C-14 in the drum solidified paraffin at NPP radioactive waste.

Sample	$\mu\text{Sv/hr g}$	Bq/g	
		H-3	C-14
PA-1-EB-U	0.04	1220±35	38±0.5
PA-1-EB-M	0.03	1090±12	38±2
PA-1-EB-B	0.03	779±15	37±1
PA-2-EB-U	0.04	135±2	56±1
PA-2-EB-M	0.03	129±1	51±0.3
PA-2-EB-B	0.03	127±1	53±1
PA-3-EB-U	0.08	153±4	44±1
PA-3-EB-M	0.10	135±15	51±2
PA-3-EB-B	0.08	163±3	52±2
PA-4-EB-U	0.13	516±16	45±1
PA-4-EB-M	0.17	738±28	201±61
PA-4-EB-B	0.16	697±2	522±128
PA-5-EB-U	0.21	1425±8	78±2
PA-5-EB-M	0.20	1398±15	83±1
PA-5-EB-B	0.25	1352±2	75±3
PA-6-EB-U	0.38	1495±13	445±12
PA-6-EB-M	0.39	1520±13	442±2
PA-6-EB-B	0.27	1489±19	468±5

3. Conclusion

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

As the radiation dose increase, the activity of C-14 was high in the case of cement drum. But that of H-3 was independent with the radiation dose. On the other hand, the result of H-3 and C-14 counting for paraffin was reverse.

REFERENCES

[1] EPRI, "Assessing the Impact of NRC Regulation 10 CFR 61 on the Nuclear Industry", EPRI NP-5983, 1988.