DEVELOPMENT OF THE DUAL COUNTING AND INTERNAL DOSE ASSESSMENT METHOD FOR CARBON-14 AT NUCLEAR POWER PLANTS

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In a pressurized heavy water reactor (PHWR), radiation workers who have access to radiation controlled areas submit their urine samples to health physicists periodically; internal radiation exposure is evaluated by the monitoring of these urine samples. Internal radiation exposure at PHWRs accounts for approximately 20 ~ 40% of total radiation exposure; most internal radiation exposure is attributed to tritium. Carbon-14 is not a dominant nuclide in the radiation exposure of workers, but it is one potential nuclide to be necessarily monitored. Carbon-14 is a low energy beta emitter and passes relatively easily into the body of workers by inhalation because its dominant chemical form is radioactive carbon dioxide ($^{14}CO_2$). Most inhaled carbon-14 is rapidly exhaled from the worker's body, but a small amount of carbon-14 remains inside the body and is excreted by urine. In this study, a method for dual analysis of tritium and carbon-14 in urine samples of workers at nuclear power plants is developed and a method for internal dose assessment using its excretion rate result is established. As a result of the developed dual analysis of tritium and carbon-14 in urine samples of radiation workers who entered the high radiation field area at a PHWR, it was found that internal exposure to carbon-14 is unlikely to occur. In addition, through the urine counting results of radiation workers who participated in the open process of steam generators, it was found that the likelihood of internal exposure to either tritium or carbon-14 is extremely low at pressurized water reactors (PWRs).

Keywords : Dual Counting, Internal Dose Assessment, Carbon-14, Tritium, Liquid Scintillation Counter

1. INTRODUCTION

Internal radiation exposure at pressurized heavy water reactors (PHWRs) accounts for approximately 20 ~ 40% of total radiation exposure; most internal radiation exposure is attributed to tritium. Radiation workers who have access to radiation controlled areas submit their urine samples to health physicists every fifteen days and additional urine samples are demanded from workers who participated in important work in high radiation exposure areas [1]. Measurement and internal dose assessment of tritium in urine samples of radiation workers are performed after radiation work. For monitoring of the internal exposure by gamma emitters including ^{58}Co, ^{60}Co, and ^{131}I, whole body counting is conducted periodically or after important radiation work. Carbon-14 is not a dominant nuclide from the radiation exposure of workers, but it is one potential nuclide to be necessarily monitored.

At PHWRs, carbon-14 is generally produced by the irradiation of oxygen present as impurities in the moderator system from $^{17}O(n,\alpha)^{14}C$ reaction. Because carbon-14 release from PHWRs takes place mostly as carbon dioxide ($^{14}CO_2$), in the case of carbon-14 release to the work area, it is easily inhaled into the body of radiation workers [1,2]. Most inhaled carbon-14 is rapidly exhaled from the worker's body, but a small amount of carbon-14 remains inside the body and is excreted by urine. Internal dose assessment for carbon-14 is conducted using the measurement results of carbon-14 in urine samples of radiation workers since carbon-14 is a low energy beta emitter.

In this study, the origin of carbon-14 and its metabolism through inhalation were reviewed for internal dose assessment

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of carbon-14 at PHWRs. In particular, the method of dual analysis of tritium and carbon-14 in urine samples was developed with the use of Liquid Scintillation Counters (LSCs) and methods for tritium measurements at the Wolsong nuclear power plants (NPPs). In addition, methods for determination of intake and internal dose assessment were established based on the measurement results of carbon-14 and its excretion rate data.

2. PRODUCTION AND EMISSION OF CARBON-14 AT PHWRS

In general, it is estimated that annual amounts of carbon-14 production are approximately 370 Ci for a PHWR and 5 ~ 8 Ci for a PWRs [2-5]. At PHWRs, carbon-14 is produced in the fuel, moderator, reactor coolant, and annulus gas system by radioactive reactions between neutrons and the parent isotopes ^{13}C, ^{14}N, ^{17}O [2,5,6]. These parent isotopes are involved in the three major types of reactions to produce carbon-14 at a 600MWe PHWR, as listed in Table 1.

At PHWRs, generation of carbon-14 from ^{13}C is lower due to its low cross section of ^{13}C and its small concentration in the coolant. On the other hand, ^{14}N has enough isotopic abundance and high cross section, and thus it is possible to produce a large amount of carbon-14, but the actual amount of carbon-14 production is small since generation of carbon-14 from ^{14}N primarily occurs in the annulus gas system. Despite the low cross section and small isotopic abundance of ^{17}O compared with ^{14}N, the oxygen reaction, $^{17}O(n,\alpha)^{14}C$, is the dominant carbon-14 production mechanism at PHWRs because a large amount of heavy water including ^{17}O is used as reactor coolant and moderator [2,5]. The carbon-14 production rate at Wolsong units 2, 3 and 4 is shown in Fig. 1.

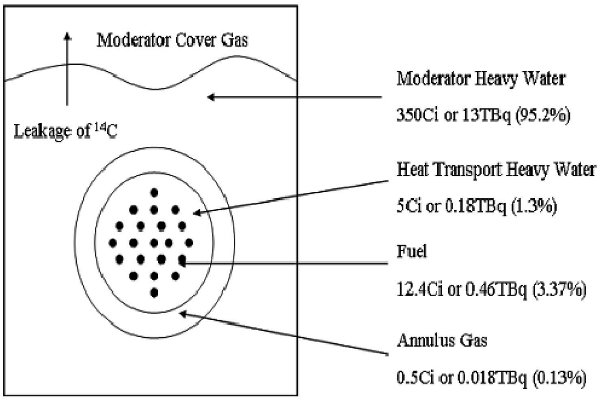

Fig. 1. Annual carbon-14 production rates at Wolsong uint 2, 3 and 4.

3. DUAL ANALYSIS OF TRITIUM AND CARBON-14

Liquid scintillation counting is generally used to measure the radioactivity of carbon-14 in urine samples of radiation workers [1,5,7,8]. The effective urine analysis method that is commonly used is to set the lower and higher energy channels of the Liquid Scintillation Counter (LSC) to rise and to fall, respectively, for detection of liquid scintillation released from only carbon-14 within a specific channel interval. Carbon-14 measurement is conducted with tritium simultaneously and a one minute count is appropriate for detection. For more than a certain radioactive level, it is necessary to employ a longer counting time, approximately ten minutes, for carbon-14 measurement [5,9,10].

In development of the method of dual analysis of tritium and carbon-14, measurement and analysis were performed with LSC and both artificial and real urine samples of radiation workers [5,9,10]. Prior to measurements, sensitivity analysis was conducted for each parameter, which can affect the counting results. The important parameters include the background of counting vials, stabilization interval of samples, change of efficiency depending on the mixture proportion of sample and cocktail, irradiation interval of external sources, and Quenching Index Parameter (QIP) depending on samples. After sensitivity analysis, effective channels of LSC were determined through the optimization process, which increases the effective count's maximum and reduces the background minimum [5,9,10].

Counting results of tritium and carbon-14 in urine samples using an LSC are illustrated by channel counts in

Table 1. Dominant carbon-14 production mechanisms at PHWRs.

Target nuclide	Natural abundance (%)	Reaction	Thermal neutron cross section
^{17}O	0.037	$^{17}O(n, \alpha)^{14}C$	0.235 barn
^{14}N	99.63	$^{14}N(n, p)^{14}C$	1.820 barn
^{13}C	1.11	$^{13}C(n, \gamma)^{14}C$	0.009 barn

Fig. 2. Tritium has a continuous spectrum with a maximum energy of 18.6 keV, but after measurement of tritium spectrum using an LSC, it was found that 86.5% of total spectrum is distributed lower than 4.0 keV. In addition, 95.6% of total counts for tritium are distributed within 12.0 keV. Thus, for tritium measurement, determination of channel from 0 to 4.0 keV is more effective to reduce the backgrounds despite the small decrease in efficiency compared with that of channel from 0 to 12.0 keV. On the other hand, carbon-14 has a maximum energy of 156 keV and, after measurement, it was found that 95.6% of total counts are distributed within 42 keV. If the upper channel is set from 0 to 60.0 keV, 99.7% of total counts are measured.

The effective channels of LSC were determined through the optimization process, which maximizes the counting efficiency and reduces the Minimum Detectable Activity (MDA). First, for determination of the lower channel, the higher channel was fixed at Channel 200 and then the lower channel was increased gradually by one channel from Channel 1 to optimize counting channels. Here, one channel is a unit that has 0.5 keV. On the other hand, for determination of the higher channel, the lower channel was fixed at Channel 1 and then the higher channel was decreased gradually by one channel from Channel 200 to optimize counting channels similar to the previous process. In this process, the effective channels were determined using the spectrums of both tritium and carbon-14, in order to maximize the counting efficiency and minimize the MDA. Figs. 3 and 4 show the determination process of effective channels for tritium and carbon-14, respectively. As a result, the effective channels were set from Channels 1 to 9 for single and dual analysis of tritium, from Channels 4 to 85 for single analysis of carbon-14, and both from Channels 30 to 85 and Channels 40 to 85 for dual analysis of carbon-14.

A reliability test for the developed method of dual analysis was conducted using standard samples made for the use of verification. To make standard samples, tritium was mixed at a level of radioactivity 100 times higher than that of carbon-14, with consideration of the practical level of radioactivity in urine samples at NPPs. As a result of the reliability test, the results demonstrated good performance, showing that the mean values of analysis for tritium and carbon-14 were almost similar to the calculated values, within 10% for tritium and 5% for carbon-14. Table 2 displays the verification results of the analyses for tritium and carbon. The standard deviation of the measured values was regarded as appropriate, and was shown to be within 8 ~ 10% of average measured values [5,9,10].

The reason for suggestion of effective channels for both single and dual analysis of tritium and carbon-14 is to make users possible to select either single or dual analysis depending on types of radioactive urine sample. That is, if only carbon-14 exists in urine samples, Channels 4 to 85 for single analysis is used. In case that both tritium and carbon-14 exist in urine samples, Channels 1 to 9 and Channels 30 to 85 or 40 to 85 for dual analysis of tritium

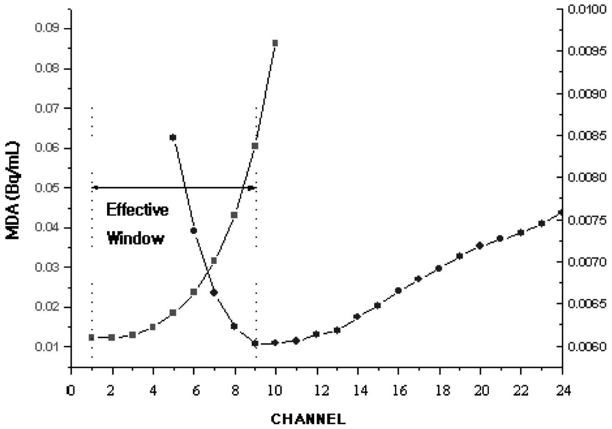

Fig. 3. Channel determination for the optimization of tritium analysis.
[a] “ ■ ” indicates a lower channel determination process.
[b] “ ● ” indicates a upper channel determination process.

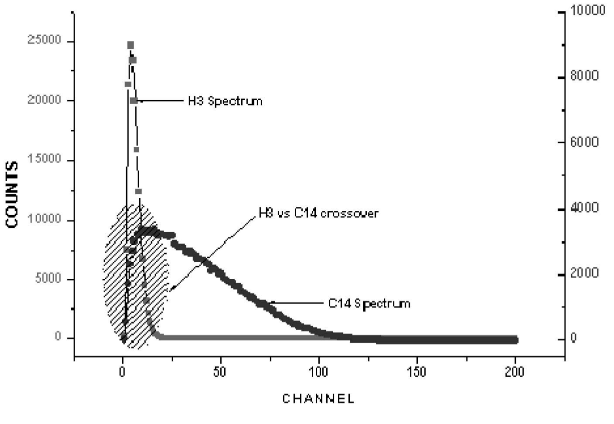

Fig. 2. Spectrum of tritium and carbon-14.

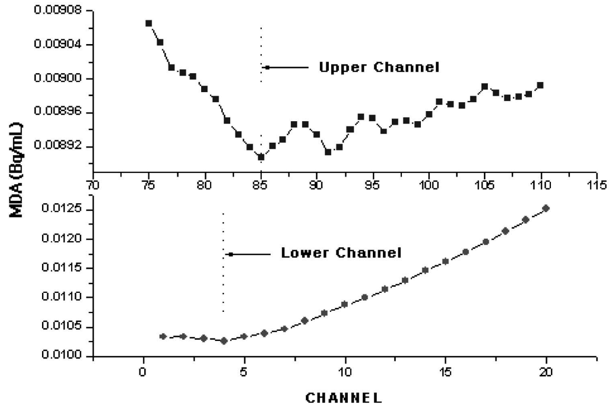

Fig. 4. Channel determination for the optimization of carbon-14 analysis.

and carbon-14, respectively are used. For dual analysis, Channels 30 to 85 has a low standard deviation and provides the close value to spiked value compared with those of Channels 40 to 85; thus, it is regarded that Channels 30 to 85 is appropriate for dual analysis of carbon-14. However, in terms of MDA, it was found that Channels 40 to 85 has a better MDA level for dual analysis of carbon-14 [5].

As a result of the application of dual analysis to urine samples of radiation workers at Wolsong NPPs, it was found that measurement results of tritium activity almost corresponded to previous measurement results of tritium activity. Thus, the validity of the dual analysis method was indirectly demonstrated. In the meantime, there was the phenomenon of crossover, the interchange of sections between tritium and carbon-14 counts, for the results of carbon-14 analysis when Channels 4 ~ 85 were set for carbon-14 analysis; this caused overestimation of carbron-14 counts. On the other hand, the phenomenon of crossover of tritium counts was properly eliminated when Channels 30 ~ 85 and Channels 40 ~ 85 were set for carbon-14 analysis. In addition, it was found through dual analysis that counting results of carbon-14 for urine samples of radiation workers at PHWRs were always lower than MDA, and that the likelihood of internal exposure to carbon-14 is extremely low [5,10]. Several measurements of tritium and carbon-14 activity for actual urine samples of radiation workers at PHWRs were conducted applying the developed method for dual analysis of tritium and carbon-14. Tables 3 ~ 6 show the some results of the dual analysis [5].

The dual counting method for tritium and carbon-14 in workers' urine was carried out for radiation workers who participated in tasks for which high radiation exposure was expected, such as the open process of steam generators during the planned maintenance period at PWRs. As a

Table 2. Verification of dual analysis results for tritium and carbon-14.

Sample No.	Spiked value (Bq/mL)		Amount of sample (mL)		tSIE	Channel	Eff (%)	Measured value			
								Net counts[a]		Activity (Bq/mL)	
	^{3}H	^{14}C	^{3}H	^{14}C				Mean	S.D	Mean	S.D
1-1	1084.8	9.5	0.528	0.642	312.7	1-9	31.7	11055	90.5	1099.3	9.0
						4-85	86.7	10465	102.4	313.2	3.1
						30-85	45.3	169	13.3	9.7	0.8
						40-85	31.0	119	12.7	10.0	1.1
1-2			0.529	0.527	320.7	1-9	32.1	11217	108.0	1101.0	10.6
						4-85	86.4	10701	92.4	391.5	3.4
						30-85	45.9	144	12.8	9.9	0.9
						40-85	31.8	101	10.2	10.0	1.0
1-3			0.512	0.558	320.1	1-9	32.1	10814	118.0	1097.5	12.0
						4-85	86.4	10359	110.5	357.8	3.8
						30-85	45.9	151	12.3	9.9	0.8
						40-85	31.8	107	9.9	10.1	0.9
2-1	958.5	8.8	0.537	0.602	312.8	1-9	31.7	9726	81.4	951.9	8.0
						4-85	86.7	9228	92.4	294.7	3.0
						30-85	45.3	147	11.9	9.0	0.7
						40-85	31.0	105	8.8	9.4	0.8
2-2			0.552	0.594	309.6	1-9	31.6	9936	119.0	949.6	11.4
						4-85	86.8	9355	99.5	302.4	3.2
						30-85	45.0	147	8.6	9.2	0.5
						40-85	30.7	104	8.7	9.5	0.8
2-3			0.533	0.583	312.1	1-9	31.7	9642	96.5	951.3	9.5
						4-85	86.7	9150	92.3	301.5	3.0
						30-85	45.2	146	13.2	9.2	0.8
						40-85	30.9	103	10.2	9.5	0.9

[a] Net counts = Gross counts – Background counts

result, it was found that tritium activity in urine samples of radiation workers at PWRs indicated almost MDA levels, much lower than those at Wolsong NPPs (PHWRs). Thus, this result demonstrated indirectly that the likelihood of internal exposure to tritium at PWRs is extremely low. Furthermore, there was no detection of carbon-14. This result confirms that there was no carbon-14 in urine samples of radiation workers at PWRs and that internal exposure to

Table 3. Tritium analysis for radiation workers at Wolsong NPPs (Windows: Channels 1 ~ 9).

No.	Given value (Bq/mL)[a]	Amount of sample (mL)	tSIE	Eff (%)	Gross counts (cpm)	Measured value (Bq/mL)	
						Gross	Net[b]
1	120	1.154	258.7	28.68	2329	116.7	116.4
2	143	1.017	199.8	22.92	1897	133.7	133.3
3	11	1.012	272.8	29.76	226	12.4	12.1
4	59	1.008	252.8	28.19	943	54.6	54.4
5	40	1.107	248.3	27.80	707	37.7	37.5
6	2	1.055	277.2	30.09	30	1.6	1.3
7	127	1.035	228.8	26.00	1984	120.5	120.2
8	391	1.033	246.6	27.65	6626	379.4	379.1
9	68	1.196	241.2	27.17	1407	70.7	70.5
10	129	0.991	291.5	31.04	2285	121.8	121.5
11	104	1.018	246.1	27.61	1593	91.4	91.2
12	233	1.051	192.8	22.12	3002	210.3	209.8
13	65	1.315	248.0	27.78	1380	61.6	61.4
14	144	1.038	249.7	27.92	2460	138.0	137.8
15	94	1.022	307.7	31.99	1892	95.0	94.8
16	49	1.012	265.1	29.18	842	46.5	46.2
17	152	1.040	250.8	28.02	2708	151.8	151.5
18	14	1.067	247.8	27.76	232	12.8	12.5
19	295	1.171	223.8	25.50	5199	281.7	281.4
20	318	1.040	273.5	29.82	5898	310.8	310.6
21	51	1.008	258.6	28.67	913	51.7	51.5
22	615	1.138	217.9	24.90	10405	596.4	596.1
23	370	1.235	209.2	23.97	6454	355.7	355.4
24	104	1.236	273.1	29.79	2379	106.2	106.0
25	100	1.136	268.0	29.41	2063	101.5	101.2
26	190	1.167	266.6	29.30	3879	185.8	185.6
27	67	1.109	261.4	28.89	1252	64.0	63.7
28	124	1.042	278.9	30.20	2269	118.4	118.1
29	64	1.051	267.8	29.39	1084	57.4	57.1
30	132	1.403	166.2	18.78	1993	122.6	122.3
31	16	1.138	264.7	29.16	306	15.2	14.9
32	85	1.274	263.8	29.08	1845	80.8	80.6
33	751	1.216	253.3	28.23	15696	749.9	749.7
34	64	1.131	229.5	26.06	1129	62.7	62.4
35	73	1.063	261.4	28.89	1354	72.2	71.9
36	108	1.044	273.9	29.84	1891	100.0	99.7

[a] Counting results from Wolsong NPPs

[b] Net (counts or Bq/mL) = Gross – Background

Table 4. Carbon-14 analysis for radiation workers at Wolsong NPPs (Windows: Channels 4 ~ 85).

No.	Eff (%)	Gross counts (cpm)	Measured value (Bq/mL)	
			Gross	Net[a]
1	88.85	1943	31.6	31.3
2	86.30	1421	27.0	26.6
3	88.80	204	3.8	3.4
4	88.79	793	14.8	14.4
5	88.72	606	10.3	10.0
6	88.73	33	0.6	0.3
7	88.11	1618	29.6	29.2
8	88.69	5457	99.3	98.9
9	88.56	1160	18.3	18.0
10	88.34	2068	39.4	39.0
11	88.68	1329	24.5	24.2
12	85.71	2223	41.1	40.8
13	88.72	1175	16.8	16.5
14	88.75	2049	37.1	36.7
15	87.59	1811	33.7	33.4
16	88.86	709	13.2	12.8
17	88.77	2238	40.4	40.1
18	88.71	212	3.7	3.4
19	87.87	4164	67.4	67.1
20	88.79	5149	92.9	92.6
21	88.85	773	14.4	14.0
22	87.56	8119	135.8	135.5
23	87.00	4930	76.5	76.3
24	88.80	2049	31.1	30.8
25	88.85	1809	29.9	29.6
26	88.85	3290	52.9	52.6
27	88.86	1069	18.0	17.8
28	88.70	2010	36.3	35.9
29	88.85	963	17.2	16.9
30	82.87	1413	20.3	20.0
31	88.86	253	4.2	3.9
32	88.86	1618	23.8	23.6
33	88.80	13135	202.8	202.5
34	88.14	921	15.4	15.1
35	88.86	1190	21.0	20.7
36	88.79	1670	30.0	29.7

[a] Net (counts or Bq/mL) = Gross – Background

The conditions of tSIE and efficiency for liquid scintillation counting are the same with those of liquid scintillation counting in Table 3 (Channel 1 to 9).

Table 5. Carbon-14 analysis for radiation workers at Wolsong NPPs (Windows: Channels 30 ~ 85).

No.	Eff (%)	Gross counts (cpm)	Measured value (Bq/mL)	
			Gross	Net[a]
1	38.26	8	0.3	0
2	27.26	3	0.2	-0.4
3	40.42	10	0.4	0
4	37.31	10	0.4	0.1
5	36.55	16	0.7	0.3
6	41.07	2	0.1	-0.3
7	33.07	10	0.5	0.1
8	36.26	9	0.4	0
9	35.33	7	0.3	-0.1
10	43.01	17	0.7	0.3
11	36.17	9	0.4	0
12	25.75	8	0.5	-0.1
13	36.51	10	0.4	0
14	36.78	16	0.7	0.3
15	44.99	10	0.4	0
16	39.26	9	0.4	0
17	36.98	8	0.4	0
18	36.47	11	0.5	0.1
19	32.12	9	0.4	0
20	40.52	15	0.6	0.2
21	38.25	9	0.4	0
22	30.99	9	0.4	0
23	29.22	10	0.5	0.1
24	40.47	14	0.5	0.2
25	39.70	6	0.2	-0.1
26	39.50	11	0.4	0.1
27	38.69	4	0.2	-0.2
28	41.30	9	0.4	0
29	39.67	8	0.3	0
30	19.57	9	0.6	0
31	39.21	9	0.3	0
32	39.06	9	0.3	0
33	37.39	10	0.4	0
34	33.20	12	0.5	0.1
35	38.68	21	0.9	0.5
36	40.58	16	0.6	0.3

[a] Net (counts or Bq/mL) = Gross – Background

The conditions of tSIE and efficiency for liquid scintillation counting are the same with those of liquid scintillation counting in Table 3 (Channel 1 to 9).

Table 6. Carbon-14 analysis for radiation workers at Wolsong NPPs (Windows: Channels 40 ~ 85).

No.	Eff (%)	Gross counts (cpm)	Measured value (Bq/mL)	
			Gross	Net[a]
1	23.15	7	0.4	0
2	13.57	3	0.4	-0.4
3	25.54	9	0.6	0.2
4	22.17	6	0.5	-0.1
5	21.41	15	1.1	0.6
6	26.31	2	0.1	-0.3
7	18.20	8	0.7	0.1
8	21.13	5	0.4	-0.1
9	20.24	4	0.3	-0.2
10	28.78	13	0.8	0.4
11	21.05	9	0.7	0.2
12	12.48	4	0.5	-0.3
13	21.37	8	0.5	0.1
14	21.64	14	1.0	0.5
15	31.63	5	0.3	-0.1
16	24.23	6	0.4	0
17	21.84	4	0.3	-0.2
18	21.33	10	0.7	0.2
19	17.39	7	0.6	0
20	25.66	14	0.9	0.5
21	23.14	8	0.6	0.1
22	16.45	6	0.5	-0.1
23	15.05	6	0.5	-0.1
24	25.60	8	0.4	0.1
25	24.72	3	0.2	-0.2
26	24.49	8	0.5	0.1
27	23.61	4	0.3	-0.2
28	26.59	5	0.3	-0.1
29	24.69	2	0.1	-0.3
30	8.41	7	1.0	0.1
31	24.17	8	0.5	0.1
32	24.01	7	0.4	0
33	22.25	8	0.5	0.1
34	18.31	10	0.8	0.3
35	23.60	16	1.1	0.6
36	25.73	8	0.5	0.1

[a] Net (counts or Bq/mL) = Gross – Background

The conditions of tSIE and efficiency for liquid scintillation counting are the same with those of liquid scintillation counting in Table 3 (Channel 1 to 9).

carbon-14 is unlikely to occur [5,10]. Accountings of the results for tritium and carbon-14 for actual urine samples of radiation workers at Yonggwang NPPs (PWRs), applying the developed method for dual analysis, are shown in Tables 7 ~ 10 [5].

4. CARBON-14 METABOLISM AND ITS EXCRETION RATE

The International Commission on Radiological Protection (ICRP) provided some information about carbon-14 in ICRP publication 10, stating that inhaled CO_2 remains inside the body for a 0.4-day retention period, and that 30% of inhaled CO_2 is deposited in the bones. In ICRP publication 30, results were presented that showed that 99% of inhaled CO_2 exhibit two short-term behaviors that have a 5-minute or a 60-minute retention period and that the final 1 % of residual carbon-14 remains inside body for 60,000 minutes (approximately 40 days). This carbon metabolism is based on test results on animals. In the meantime, ICRP publication 54 (1983) and publication 78 (1997) do not provide detailed information about carbon-14 metabolism or give any guidance on internal exposure monitoring. According to ICRP publications, the central compartment of radioactive carbon dioxide ($^{14}CO_2$) inhaled by breathing or ingested by diet is the behavior of CO_2/HCO_3^-. Carbon-14 metabolism for CO_2 inhalation is illustrated in Fig. 5 [6].

In Canada, measurement experiments have been performed to examine the excretion rate of carbon-14 for volunteers who have inhaled low levels of the substance, and for whom the density of inhaled $^{14}CO_2$ was known; these experiments were done to estimate the internal exposure of NPP workers to inhaled CO_2. It was found that radiocarbon excreted was present as bicarbonate in urine and that it had retention periods of 0.4, 1.2, and 40 days, respectively. Based on this metabolism data, the Canadian Nuclear Safety

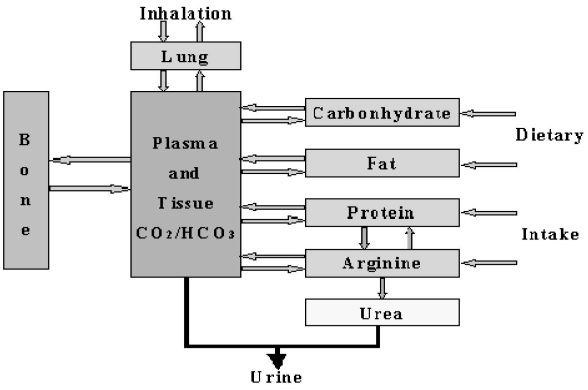

Fig. 5. Typical carbon-14 metabolism of human.

Table 7. Tritium analysis for radiation workers at Yonggwang NPPs (Windows: Channel 1 ~ 9).

No.	Radiation workers	Gross counts (cpm)		Gross specific activity (Bq/mL)		Net specific activity[c] (Bq/mL)		MDA (Bq/mL)	
		Before[a]	After[b]	Before[a]	After[b]	Before[a]	After[b]	Before[a]	After[b]
1	A	11.3	11.2	0.7	0.7	0.5	0.5	0.8	0.7
2	B	8.6	10.6	0.5	0.6	0.3	0.4	0.7	0.7
3	C	8.9	12.8	0.6	0.7	0.3	0.5	0.7	0.7
4	D	7.6	8.7	0.4	0.5	0.2	0.3	0.7	0.7
5	E	7.3	8.9	0.4	0.5	0.2	0.3	0.7	0.7
6	F	22.5	24.6	1.4	1.6	1.2	1.4	0.7	0.8
7	G	165.3	171.5	9.2	9.4	9.0	9.2	0.7	0.7
8	H	9.2	9.2	0.5	0.5	0.3	0.3	0.7	0.7
9	I	9.0	9.6	0.5	0.6	0.3	0.3	0.7	0.7
10	G	-	8.6	-	0.4	-	0.2	-	0.6

[a] Counting results in urine samples before radiation work
[b] Counting results in urine samples after radiation work
[c] Net (counts or Bq/mL) = Gross – Background

Table 8. Carbon-14 analysis for radiation workers at Yonggwang NPPs (Windows: Channels 4 ~ 85).

No.	Radiation workers	Gross counts (cpm)		Gross specific activity (Bq/mL)		Net specific activity[c] (Bq/mL)		MDA (Bq/mL)	
		Before[a]	After[b]	Before[a]	After[b]	Before[a]	After[b]	Before[a]	After[b]
1	A	19.4	20.3	0.4	0.4	0.01	0.03	0.4	0.4
2	B	19.9	20.1	0.4	0.4	0.02	0.03	0.4	0.4
3	C	20.0	25.2	0.4	0.5	0.02	0.12	0.4	0.4
4	D	19.6	20.5	0.4	0.4	0.02	0.03	0.4	0.4
5	E	18.5	21.4	0.4	0.4	-0.01	0.05	0.4	0.4
6	F	30.9	32.7	0.6	0.6	0.23	0.26	0.4	0.4
7	G	157.2	164.2	3.0	3.1	2.65	2.78	0.4	0.4
8	H	17.6	22.4	0.3	0.4	-0.02	0.07	0.4	0.4
9	I	21.0	21.4	0.4	0.4	0.04	0.05	0.4	0.4
10	G	-	22.1	-	0.4		0.07		0.5

[a] Counting results in urine samples before radiation work
[b] Counting results in urine samples after radiation work
[c] Net (counts or Bq/mL) = Gross – Background

Commission (CNSC), the nuclear regulatory body, organized the Working Group on Internal Dosimetry and provided bioassay guideline for carbon-14 inhalation [5,10-12].

In Canadian carbon-14 metabolism, the retention of radiocarbon activity, R(t), and the fractions of intake excreted per day in breath, E'$_b$(t), and in urine, E'$_u$(t), over a specific period after intake were described in Eq. 1 ~ 3 [5,10-13]. These relations are presented in Fig. 6.

$$R(t) = 0.06e^{-1.7t} + 0.02e^{-0.58t} + 0.02e^{-0.017t} \tag{1}$$

$$E_b^{'}(t) = 0.1e^{-1.7t} + 0.01e^{-0.58t} + 0.0003e^{-0.017t} \tag{2}$$

$$E_u^{'}(t) = 0.02e^{-1.7t} + 0.002e^{-0.58t} + 0.00004e^{-0.017t} \tag{3}$$

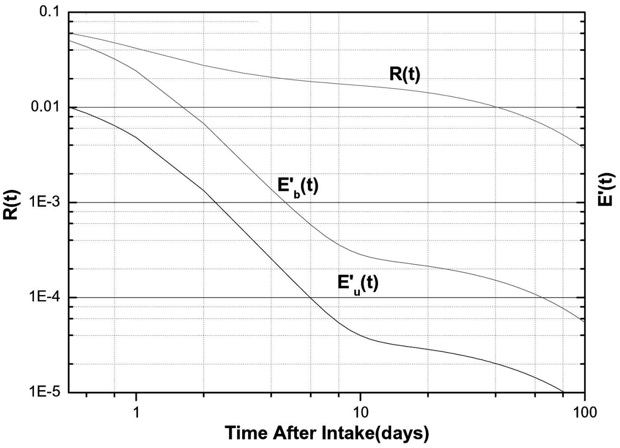

Fig. 6. Excretion rate of carbon-14 over time after intake.

Table 9. Carbon-14 analysis for radiation workers at Yonggwang NPPs (Windows: Channels 30 ~ 85).

No.	Radiation workers	Gross counts (cpm)		Gross specific activity (Bq/mL)		Net specific activity[c] (Bq/mL)		MDA (Bq/mL)	
		Before[a]	After[b]	Before[a]	After[b]	Before[a]	After[b]	Before[a]	After[b]
1	A	9.3	8.5	0.4	0.4	0.1	0.1	0.8	0.8
2	B	9.4	8.6	0.4	0.3	0	0	0.6	0.6
3	C	8.0	10.1	0.4	0.4	0.1	0.1	0.8	0.7
4	D	9.3	9.0	0.4	0.4	0.1	0	0.7	0.7
5	E	8.4	9.4	0.3	0.4	0	0.1	0.7	0.7
6	F	9.1	9.5	0.4	0.5	0.1	0.2	0.8	0.9
7	G	9.1	9.0	0.4	0.3	0	0	0.7	0.6
8	H	7.6	9.8	0.3	0.4	0	0.1	0.6	0.6
9	I	9.4	9.5	0.4	0.4	0.1	0.1	0.7	0.7
10	G	-	9.1	-	0.3		0		0.6

[a] Counting results in urine samples before radiation work
[b] Counting results in urine samples after radiation work
[c] Net counts (Bq/mL) = Gross counts – Background counts

Table 10. Carbon-14 analysis for radiation workers at Yonggwang NPPs (Windows: Channels 40 ~ 85).

No.	Radiation workers	Gross counts (cpm)		Gross specific activity (Bq/mL)		Net specific activity[c] (Bq/mL)		MDA (Bq/mL)	
		Before[a]	After[b]	Before[a]	After[b]	Before[a]	After[b]	Before[a]	After[b]
1	A	6.6	6.2	0.5	0.5	0.2	0.1	1.2	1.1
2	B	7.4	6.8	0.4	0.4	0.1	0	0.9	0.8
3	C	5.8	7.7	0.4	0.5	0.1	0.1	1.1	0.9
4	D	6.7	6.4	0.4	0.4	0.1	0	0.9	0.9
5	E	6.5	7.1	0.4	0.4	0.1	0.1	0.9	0.9
6	F	6.4	6.6	0.5	0.6	0.1	0.3	1.1	1.3
7	G	6.7	6.3	0.4	0.4	0.1	0	0.9	0.9
8	H	5.6	7.1	0.3	0.4	0	0.1	0.8	0.8
9	I	6.8	7.0	0.4	0.5	0.1	0.1	0.9	1.0.
10	G	-	7.0	-	0.4		0		0.7

[a] Counting results in urine samples before radiation work
[b] Counting results in urine samples after radiation work
[c] Net counts (Bq/mL) = Gross counts – Background counts

5. INTERNAL DOSE ASSESSMENT

The dose rate (rem/d) to a tissue of mass m (g) resulting from radioactivity Q (μCi) of carbon-14 is given by Eq. 4. Here, E (MeV) is the mean energy of the beta emission. The committed effective dose (rem) resulting from an initial radioactivity Q_o (μCi) is described in Eq. 5 [5,10,13].

$$Dose\ Rate = \frac{51.2 \times E \times Q}{m} \tag{4}$$

$$Committed\ Effective\ Dose\ = \frac{51.2 \times E \times Q_o}{m} \int R(t) e^{\frac{-0.693\,t}{T_{1/2}}}\, dt \tag{5}$$

Inhaled carbon-14 is distributed throughout all soft tissues of the body. Thus, the committed effective dose, resulting from the mean energy 0.049 MeV and the soft tissue mass of Reference Man 63,000 g, is given by Eq. 6. For a urine sample of concentration C (μCi/L) obtained at time t after single or acute intake, the intake can be estimated by Eq. 7. Here, "1.4 L" is the nominal daily excretion of urine. The final committed effective dose is then given by Eq. 8, which can be described in Eq. 9 using SI unit (mSv) [5,10,13].

$$Committed\ Effective\ Dose\ = 5.0 \times 10^{-5} \times Q_o \quad [\text{rem}] \tag{6}$$

$$Intake = \frac{1.4 \times C}{E_u(t)} \tag{7}$$

$$\textit{Committed Effective Dose} = \frac{7.0 \times 10^{-5} \times C}{E_u(t)} \quad [\text{rem}] \tag{8}$$

$$\textit{Committed Effective Dose} = \frac{25.9 \times C}{E_u(t)} \quad [\text{mSv}] \tag{9}$$

6. CONCLUSION

In this study, carbon-14 metabolism was investigated to establish a methodology for internal dose assessment of carbon-14 inhalation. Furthermore, a method for dual analysis of tritium and carbon-14 in urine samples of NPP workers was developed. Finally, the procedures for measurement and dose calculation of tritium and carbon-14 activity were provided based on the experiment and investigation results.

For dose calculation, both Canadian and ICRP carbon-14 metabolism were reviewed in detail and used for the establishment of the technical background of internal dose assessment. In addition, the method for dual analysis of tritium and carbon-14 was developed linked with the current method for single analysis of tritium. The effective channels of LSC were set as Channels 1 ~ 9 for analysis of tritium and both Channels 30 ~ 85 and 40 to 85 for analysis of carbon-14, respectively; then, the validity of the developed dual analysis was demonstrated using several experiments. In the meantime, analysis of carbon-14 in actual urine samples of radiation workers at PHWR was conducted using the developed method for dual analysis. As a result, it was found that there was no internal exposure by carbon-14 at both PWRs and PHWRs. Thus, it is expected that this research result can be implemented for monitoring the internal exposure to carbon-14 during large maintenance periods including the change of pressurized tubes at Wolsong unit 1, planned for April, 2009. It is also expected that the developed technology can make the internal dose assessment of carbon-14 possible and contributed to the health of radiation workers at NPPs.

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