Development of radioactivity measurement using isothermal calorimetric system by comparing with TDCR counter

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Abstract. The radioactivity measurement method using a calorimeter uses the 1st law of thermodynamics. The decay energy of a nuclide is absorbed in isothermal system (calorimeter) and converted into thermal energy. The converted thermal energy causes a temperature change in the chamber. Heat flow occurs due to temperature difference between sample and background. The heat flow rate of calorimeter is converted into a measurable electrical power by the thermoelectric sensor using the Seebeck effect. Radioactivity can be calculated using the decay energy of the radionuclide, the absorption efficiency at which decay energy is absorbed by the calorimeter, the calibration factors of the system, and the measured heat flow rate. The isothermal calorimeter measurement method can overcome the problem of counting loss due to dead time. However, since the measurement range of the calorimeter differs from that of the ionizing radiation detector, studies are being conducted to secure traceability of radioactivity. Comparison between TDCR and calorimeter was performed using ^{32}P, a pure beta-emitter. As a result, the results of comparing the radioactivity (calorimeter) and the TDCR measurement had a difference of 2.3 %. The result was at the time when the effect of dead time is expected to be large. After a period of time to overcome the effects of dead time, the comparison seems to be good. In addition, it would be possible to measure radioactivity more precisely in the calorimeter if the effect of heat defect on the liquid ratio of liquid source is demonstrated.

KEYWORDS: Isothermal Microcalorimetry(IMC), thermal energy, absorption efficiency, calibration factor, Triple-to-Double Coincidence Ratio(TDCR), P-32

1 INTRODUCTION

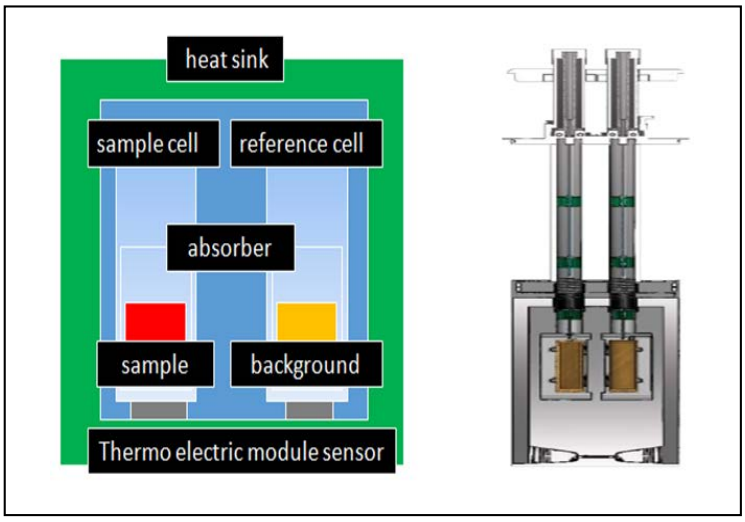
The radioactivity detector using the ionizing radiation has a dead time due to the characteristics of the circuit. The dead time causes a counting loss and increases the error between the prediction value and the measurement value. An isothermal microcalorimetry was chosen as an alternative to high-level radioactivity measurement. The isothermal microcalorimetry measurement can measure the radioactivity with the total heat energy absorbed in the system, so that the influence of the dead time can be removed. But the calorimeter has a problem that the range of coefficient efficiency is different from other radiation detector. We measured the heat flow rate of the source in calorimeter, and divided it by the decay energy of the radionuclide absorbed in calorimeter per Bq, and calculated the radioactivity by applying the calibration factor. We then attempted to obtain the traceability of radioactivity measurement by comparing with the TDCR counter in KRISS. [1, 2]

1.1 Measurement principles of radioactivity in IMC system

The relation of the heat flow from the radioactive source, temperature, and work follow the 1st law of thermodynamics. Therefore, the decay energy (from radioactive source) absorbed in the IMC system without temperature change is completely converted into heat energy. The isothermal microcalorimeter is composed of two cells (sample cell, reference cell) as shown Fig. 1, and the temperature changes between cells due to the converted heat energy of source. The thermo-electric module sensor senses the temperature change and converts it to electrical power using the Seebeck effect. The unit of electrical power is same as the heat flow rate of the calorimeter and enables DAQ. [1, 2] After measuring the heat flow rate, we use the decay energy absorbed in the calorimeter and calorimetric calibration factor to determine the radioactivity of source. Therefore if we know the decay energy ($\hat{E}$) of a given radionuclide, the absorption efficiency (k) of the absorber, and calibration factor

of calorimeter(c), we can determine the radioactivity (A) of the radionuclide by measuring the heat flow rate (P) in the calorimeter. The parameters have a related expression such as $P\ c = k\ A\ \hat{E}$. The efficiency of energy absorption is determined by the amount of decay energy and type of decay of radionuclide. [1, 2, 6, 7]

Figure 1: Internal structure of IMC system (left), and structure of TAM III (right, resource : www.tainstrument.com).



1.2 Equipment specifications

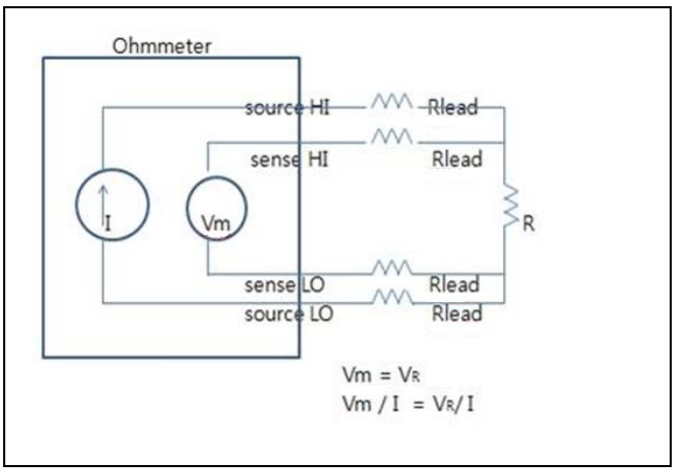
The calorimeter used is a TAM III microcalorimeter (from TA Instruments, Inc). TAM III uses mineral oil to reduce the temperature gradient inside the system to less than 10 μK at room temperature between 5 and 150 ° C. The performance test was conducted considering the deviation and drift errors and the baseline width was within 150 nW. In addition, the calorimeter was tested to take the time required to stabilize after entering the external temperature when the source was placed in the calorimeter. As a result, it was confirmed that the time required to measure the radioactivity was 12 h on average.

2 EXPERIMENT

2.1 Obtaining calibration factor of calorimeter

The calorimeter internal calibration factor was determined by the joule-heating method. Attempts have been made to eliminate thermal offset to precisely measure the resistance. After connecting 6221 AC and DC current source (KITHELY Instruments) and 2182 nanovoltagemeter (KITHELY Instruments) to the resistor (Precision Resistor Co., Inc) as shown Fig. 2, the voltage was measured by supplying the current of ± 5 ~ 20 μA of the current source. The voltagemeter was set to digital, 10 points were measured by the moving average method and the moving average value was measured per one minute for minimizing the deviation error. In addition, a voltage of 0 μA was measured and subtracted to the voltage, and the supplied current was divided. The average resistance of resistor was 199.978 kΩ. [3]

Figure 2: Experiment to know the resistance value to be used for power calculation.



The resistor of 199.978 kΩ connected with current source and voltagemeter was placed inside the calorimeter as shown Fig. 3. The current of current source was supplied and voltage was measured by DAQ. The expected power (generated from known voltage and resistance) and the heat flow rate measured in the calorimeter were compared to obtain the calibration factor. The heat flow rate was generated up to 2 - 400 μW, and linear function fitting was performed by comparing expected power and measured heat flow rate as shown Fig. 3. As a result, a calibration factor was obtained in the form of an equation such as y = 1.015 x + 0.078 and the fitting for comparison is shown in Fig. 4. [3, 4]

Figure 3: The diagram of experiment system for the purpose of getting calibration factor.

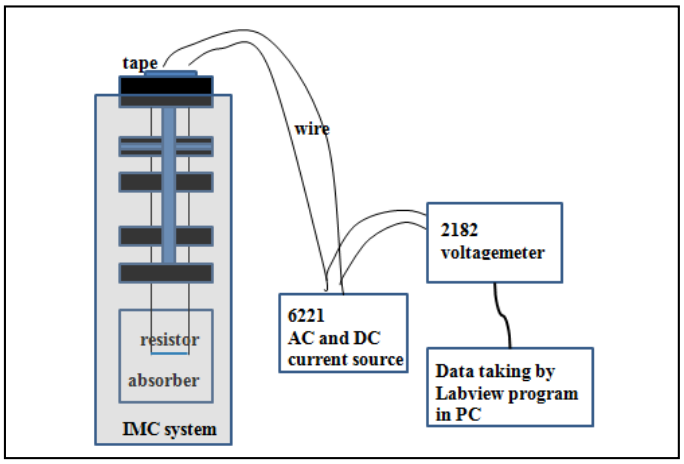
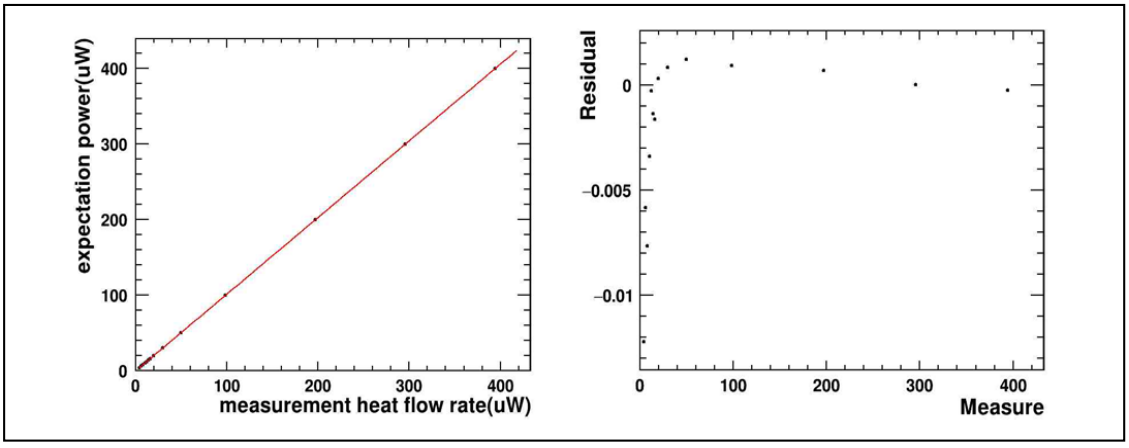


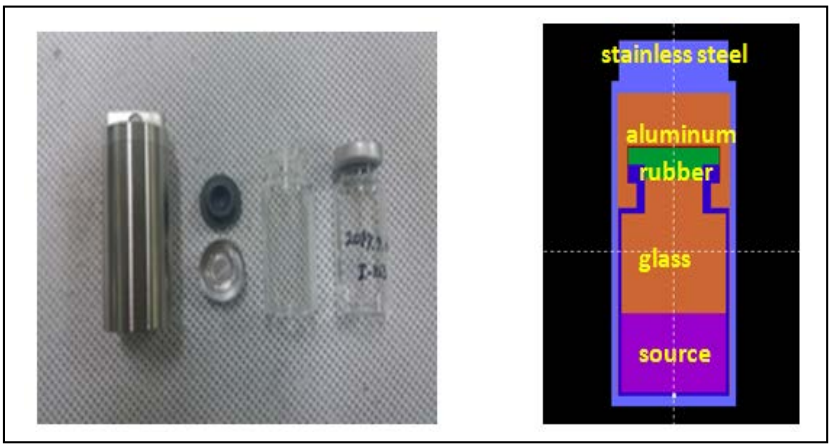
Figure 4: Graph showing fitting between the measured heat flow rate (x-axis) and expected power (y-axis).



2.2 Simulation for Energy Absorption

Considering the stabilization time of the calorimeter, ^{32}P was used with a half life time of 14.284 d for radioactivity measurement. ^{32}P is a pure-beta radionuclide with average beta energy of 695.5 keV per Bq. The absorber is made of stainless steel material (Fe : 70.80 %, Cr : 18.06 %, Ni : 8.06 %, etc) for the purpose of 100% absorption of average decay energy as shown Fig. 5. [5, 6] As a result of using Monte Carlo simulation (PENELOP 2012), the decay energy absorbed in the absorber was almost equal to the average beta energy. The result of summing the absorption energy of each spectrum after generating the spectra with 1000 channels continuously with the maximum decay energy (1720.7 keV) of ^{32}P was also the same as the previous result. Thus, the absorption efficiency of ^{32}P inside the calorimeter was 40.65% of the maximum decay energy. Finally, ^{32}P radioactivity in the calorimeter can be obtained by applying the calibration factor after dividing the measured heat flow rate by 695.5 keV per Bq, which is 40.65% of the maximum decay energy. [5, 6, 7]

Figure 5: Actual appearance of absorber, and vial, materials used in simulation (left) and simulation geometry for simulating energy absorption (right, from PENELOPE2012)



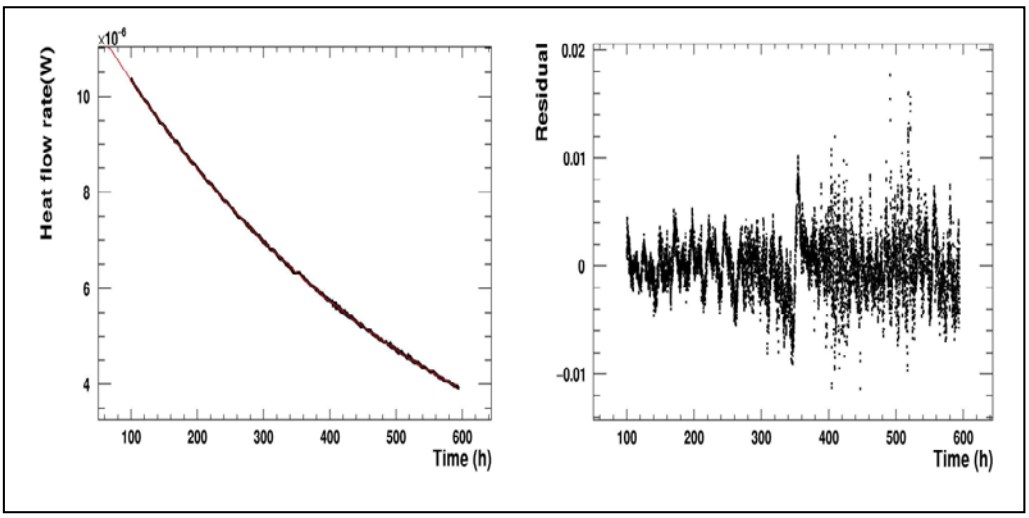
3 RESULT

The type of source used in the experiment was a 5 mL solution source (Eckert & Ziegler) with a nominal radioactivity of 92.5 MBq. The radioactivity measurement of ^{32}P source in calorimeter was about 3 weeks and Subsequent radioactivity measurement was replaced by TDCR. Considering the dead time of the TDCR counter, 5 samples were sampled in mg unit and TDCR measurements were performed. Since the TDCR system is difficult to measure high-level radioactivity due to dead time, measurements were made after a certain period of time (4 months). [1, 6]

3.1 Data Analysis

The measurement time of ^{32}P source is about 3 weeks, and it is confirmed that the half life time of the heat flow rate in the calorimeter corresponds to the half life time of ^{32}P. As a result of the fitting, the half life time was 14.30 days, and the error was 0.089% with the reference data, and it was confirmed that the radioactivity measurement of ^{32}P source proceeded as expected in the calorimeter. The rear part of total data (350 ~ 600 h) was analyzed to remove the influence of the stabilization time of calorimeter. Including the baseline of the calorimeter, the data were fitted with equation $y = a\, e^{(-(\ln 2/b)t)} + c$ (a = initial heat flow rate (μW), b = half life time (min), c = baseline (μW)) as shown Fig. 6., and fitting results for each parameter are shown in the Table 1.

Figure 6: Graph showing fitting for radioactivity measurement results of 32P radioactivity of IMC (left) and residual between fitting and measurement results (right).



Initial heat flow rate was 12.509 μW, so the initial radioactivity was 113.97 MBq after applying the calibration factor (y = 1.015 x + 0.078) after dividing the heat flow rate (12.509 μW) by the absorbed energy (1710.66 keV × 40.65 %). The uncertainty evaluation for radioactivity was calculated as the combined standard uncertainty of calibration factor in calorimeter, measured heat flow rate in calorimeter, decay energy of ^{32}P as shown in Table 2.

Table 1: Fitting parameter of ^{32}P radioactivity measurement in IMC system

Fitting parameter	Fitting value
Initial heat flow rate (μW)	12.509
Half life time of heat flow rate (h)	342.816
Baseline of calorimeter (μW)	0.168

Table 2 : Uncertainties of radioactivity measurement in IMC system

Parameter	Value	Uncertainty (%)
Calibration factor in calorimeter	1.015 x ± 0.078	0.5
Heat flow rate in calorimeter	12.509 ± 0.168	1.34
Decay energy	1710.66 ± 0.21	0.01

Combined standard uncertainty u_c of radioactivity of calorimeter was 1.43 %, and initial radioactivity was 113.97 ± 1.63 MBq.

3.2 Results Comparison

The total mass of the solution source was 5.0014 g and the radioactivity per mass was 0.028 MBq / mg. After a certain period of time (about 4 months), the radioactivity of 5 samples measured by TDCR counter was calibrated to the measurement median time, and the results were shown in Table 3.

Table 3 : Radioactivity measurement results of TDCR counter

Sample	Mass(mg)	Radioactivity per mass (Bq / mg)
1st sample	22.23	18.55
2nd sample	6.56	18.53
3rd sample	4.81	18.62
4th sample	16.89	18.66
5th sample	8.33	18.60

The initial radioactivity of the calorimeter was calculated by applying a half-life equation to match the TDCR measurement time. Radioactivity per mass of calorimeter at TDCR measurement time was 18.40 Bq / mg. The calculated values were compared with those measured by TDCR counter to confirm the radioactivity traceability. The average measured radioactivity per mass of TDCR counter was 18.59 ± 0.06 Bq / mg. Compared to the 18.40 Bq / mg calorimeter results, the TDCR results were 1.1% greater. With respect to this difference, it is judged that the effect of the impurities because the remaining ^{32}P is almost absent.

4 CONCLUSION

As a result of comparison with calorimeter and TDCR using ^{32}P pure-beta emission source, two characteristics were confirmed. First, heat flow rate decrease rate of the calorimeter is almost the same as the half life time of the ^{32}P radionuclide. Second, the expected value of the calorimeter shows a 1% difference from the measured value of TDCR after a certain period of time. Therefore, it is expected that the IMC system will ensure the traceability of radioactivity measurement if the TDCR counter is matched with the range which the influence of dead time is minimized and the influence of impurities is minimized. The results suggest that the heat defect that could occur in the calorimetric measurements of the radioactive solution are negligible in comparison of the magnitude of the evaluated uncertainty because of a small amount of source solution.[8]

5 ACKNOWLEDGEMENTS

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