# Development of Continuous Inflow Tritium in Water Measurement Technology by using Electrolysis and Plastic Scintillator

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# Introduction


Fig. 1. A person concerning of Wolseong NPP which is PHWR type and operated since 1983 in Gyeongju, Republic of Korea.

- 4 Pressurized Heavy Water Reactors (PHWR) in Gyeongju have been operated where the oldest one started its operation in April, 1983.
- People living around the nuclear power plants has worried about the safety of drinking water.

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# Introduction

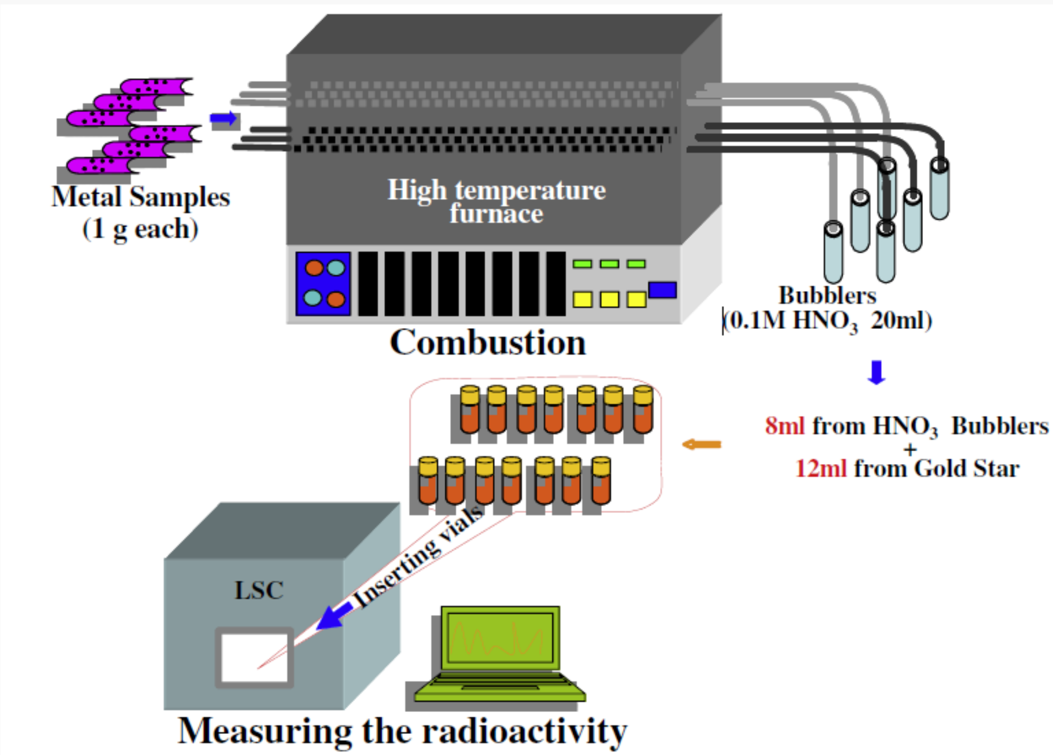


Fig. 2. A system for $^3$H trapping and measurement [1].

- $^3$H analysis using liquid scintillation counter (LSC) requires multiple steps of pre-treatment.
- The pre-treatment for trapping $^3$H requires much time (more than 5 hours for pre-treatment of 6 samples) and radioactivity analysis by LSC produces organic waste.

# Introduction

- $^3H$ radioactivity monitoring using ionization chamber has demerit that it has fluctuation for radioactivity level and it is hard to discriminate which types of particles are entered.

- Tritium in water monitor has high MDA (about 370 kBq/L in 2 hours) and not provides continuous sampling which is not applicable for continuous monitoring purpose.

- To overcome these, continuous tritium monitoring system in water based on plastic scintillation and electrolysis method was suggested.

# Material & Methods

➤ **The Concept of Tritium Continuous Monitoring System**

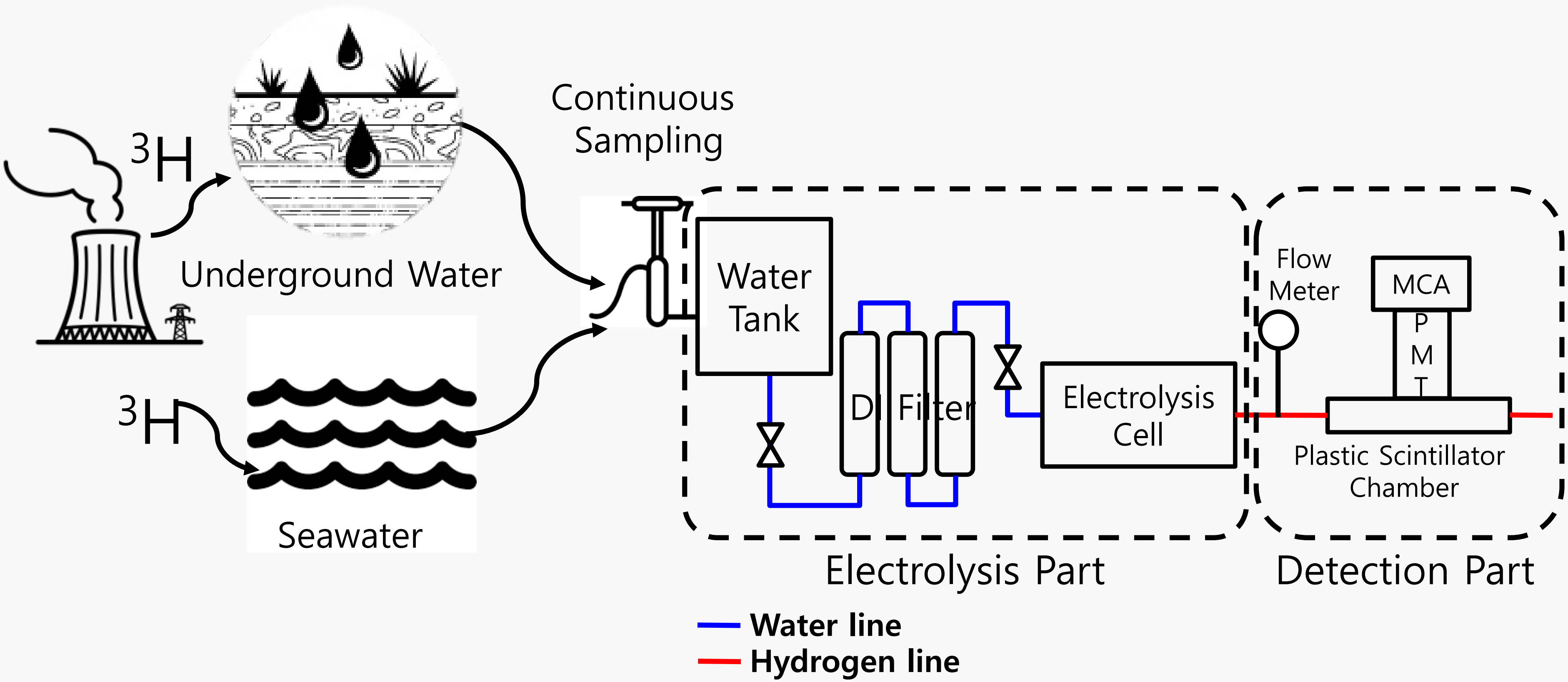


Fig. 3. Conceptual diagram of system for tritium continuous monitoring system using electrolysis and plastic scintillator detector.

# Material & Methods

## ➢ Electrolysis and Tritium Enrichment

- Electrolysis has been used to enrich the tritium concentration but iridium/platinum (Ir/Pt) polymer electrolyte membrane (PEM) electrolyzer has low enrichment factor and could generate the gaseous tritium [2].

- Fractionation factor (β) was adopted to estimate the enrichment factor of electrolyzer and ability to generate the gaseous tritium.

$$E = \frac{T}{T_0} = \left(\frac{V_0}{V}\right)^{1-\frac{1}{\beta}}, \beta = \frac{\ln\left(\frac{V_0}{V}\right)}{\ln\left(\frac{V_0 T_0}{VT}\right)}$$

  - E: Enrichment factor
  - T, $T_0$: Tritium concentration in Bq/L after and before electrolysis
  - V, $V_0$: Volume of the tritiated water sample in L after and before electrolysis

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# Material & Methods

## ➢ The Detector Design

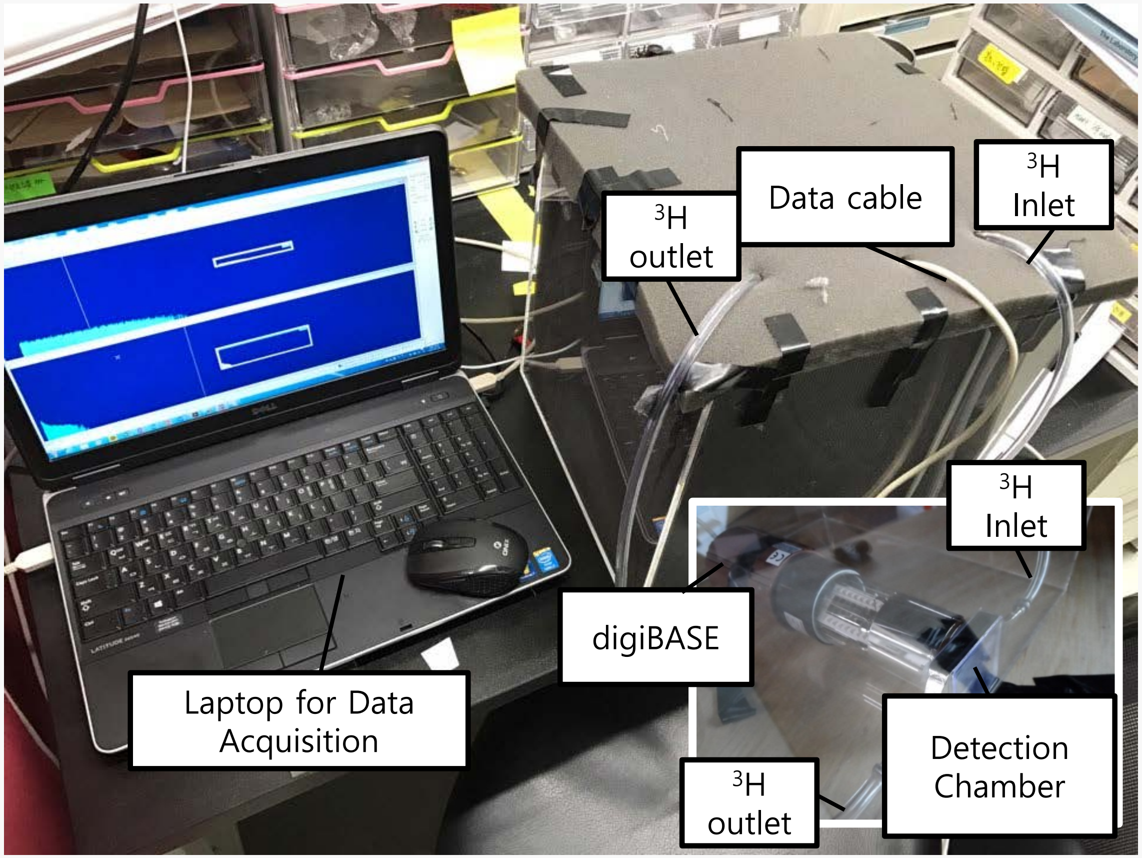


Fig. 4. Picture of the detecor part

- Plastic scintillator has merits for beta-ray measurement because of low probability of back-scattering and stability [3].
- Two 50 × 50 × 1 $mm^3$ planar plastic scintillator were attached to the metal chamber.

# Material & Methods

➤ **The Continuous Sampler and Electrolyzer**

Fig. 5. Picture of the electrolyzer and continuous sampler system

- The continuous sampler system included water pump with carbon filter in its inlet, DI filter, DI water tank.
- The electrolyzer included PEM electrolysis cell, water trap and flow meter.
- The system was operated with 12 V 80 AH lead storage battery.

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# Material & Methods

## ➢ Continuous Measurement Experiment

- The tritium sample (liquid) with activity concentration of 250 kBq/L was used.

- The tritium concertation in the gaseous sample can be calculated by:

$$T_{gas} = T_{liquid} \times \frac{1}{\kappa} \times \frac{1}{\beta}$$

  - ✓ $T_{gas}$, $T_{liquid}$: Tritium concentration for gaseous and liquid sample [Bq/L]
  - ✓ κ: Hydrogen volume correction factor
  - ✓ β: Fractionation factor

- Estimated tritium concentration of gaseous sample was 58.35 Bq/L.

- Measurement was carried out every 5 minutes for 15 times.

# Material & Methods

## ➢ Minimum Detectable Activity Estimation

- The minimum detectable activity (MDA) for gaseous sample in Bq/L was calculated by:

$$MDA_{gas} = \frac{2.71 + 4.65\sqrt{B_{cps} \times T}}{T \times V \times \varepsilon}$$

  - ✓ $B_{cps}$: Background count rate [cps]
  - ✓ T: Measurement time
  - ✓ V: Volume of the detector chamber [L]
  - ✓ ε: Detection efficiency

- The MDA for liquid sample in kBq/L was calculated by:

$$MDA_{liquid} = \frac{2.71 + 4.65\sqrt{B_{cps} \times T}}{T \times V \times \varepsilon} \times \kappa \times \beta \times \frac{1}{1000}$$

  - ✓ κ: Hydrogen volume correction factor (1234.7)
  - ✓ β: Fractionation factor (3.47)

# Results & Discussion

## ➢ Fractionation Factor (β) for the PEM Cell

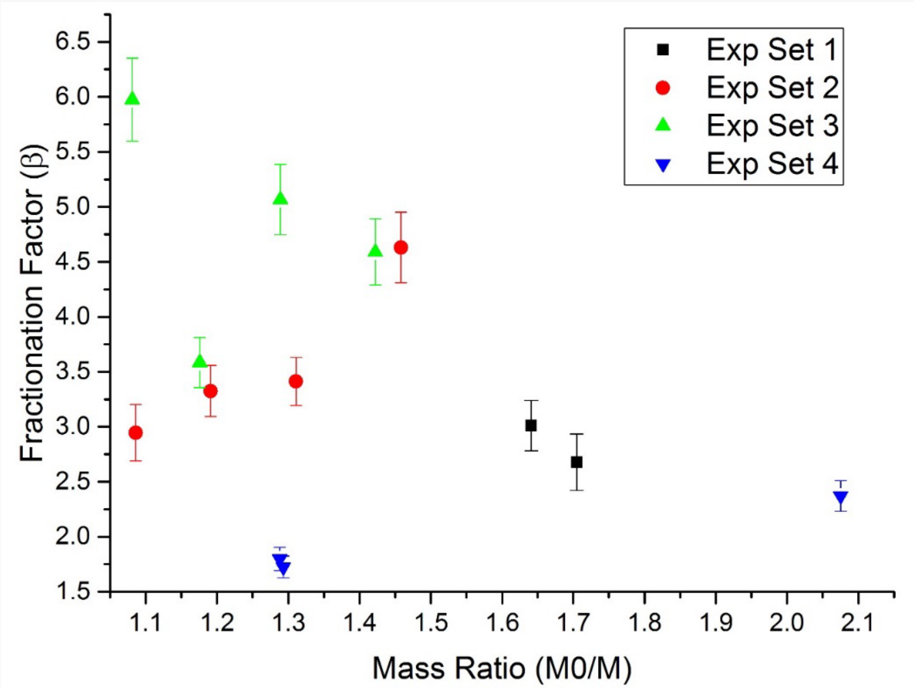


Fig. 6. Serial estimation result of fractionation factor (β).

- The average fractionation factor was 3.47±0.251 while Aurelie reported the value as 4.67 [2].

- The fractionation factor was estimated by mass ratio instead of volume ratio using Quantulus 1220.

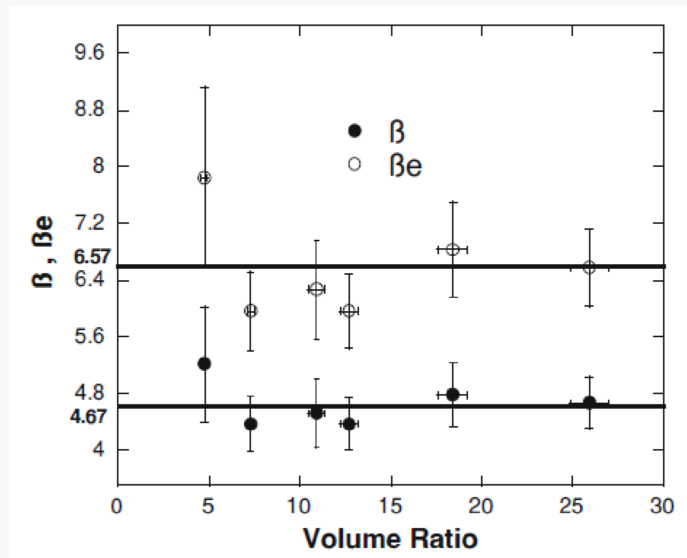


Fig. 7. Fractionation factor for PEM cell reported by Aurelie's work [4]

# Results & Discussion

➢ **Characterization of the Plastic Scintillator Detector**

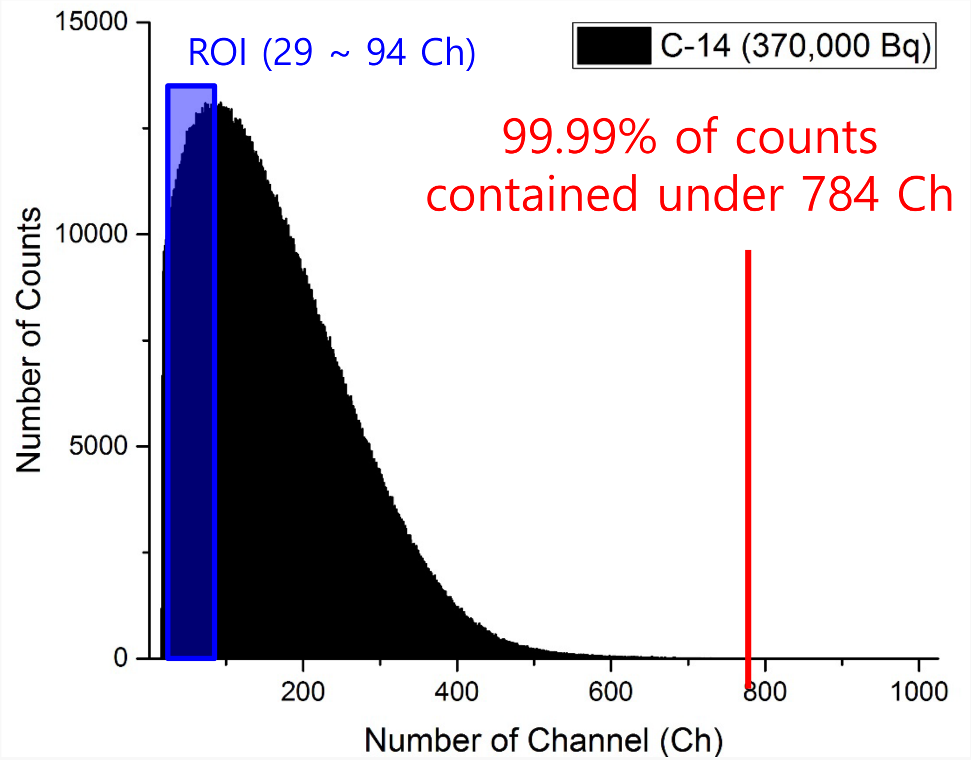


Fig. 8. Energy calibration using $^{14}C$ source and range of interest (ROI) setup based on the tritium energy spectrum.

- The energy calibration was carried out by $^{14}C$ point source.
- 99.99% of counts contained under 784th channel. (156/784 = 0.199 keV/Ch)
- ROI was set between 29 (~5.7 keV) and 94 (~18.6 keV) to obtained a half of beta-ray emitted from tritium.

# Results & Discussion

## ➢ Characterization of the Plastic Scintillator Detector (cont.)

- The efficiency of tritium was calculated using tritium check (surficial) source.

- Corrected beta emission rate of the check source was 189 $s^{-1}$ for 2π direction with 6% relative error.

- Estimated efficiency was 33.28±4.016 % while the uncertainty quoted is the expanded uncertainty with a coverage factor K=2 (95% confidence level).

# Results & Discussion

## ➢ Continuous Measurement Result

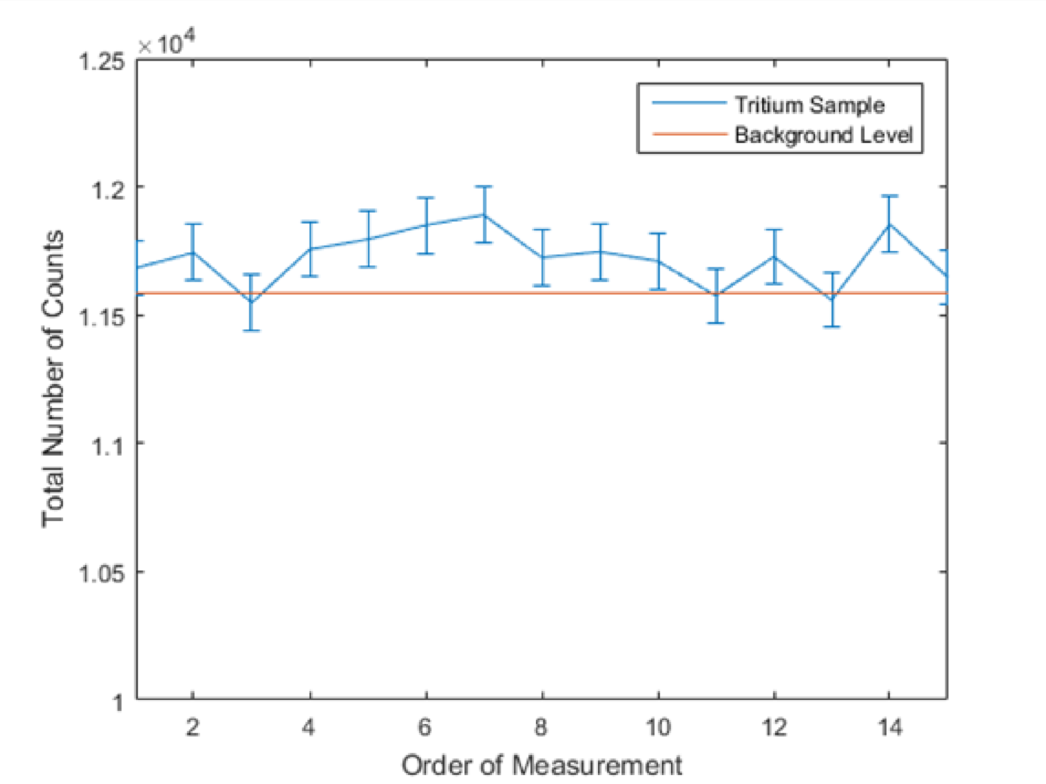


Fig. 9. Continuous measurement result for tritium gas sample.

- The average count rate for gaseous tritium (58.35 Bq/L) and background was 2344±20.37 and 2317±11.75, respectively.
- Count rate of gaseous tritium shows slightly(1.1%) higher value than that of the background level.
- The count rate of lower limit of detection was 100.64 cpm where the net count rate of the tritium gas was 27 cpm.

# Results & Discussion

## ➢ MDA Estimation

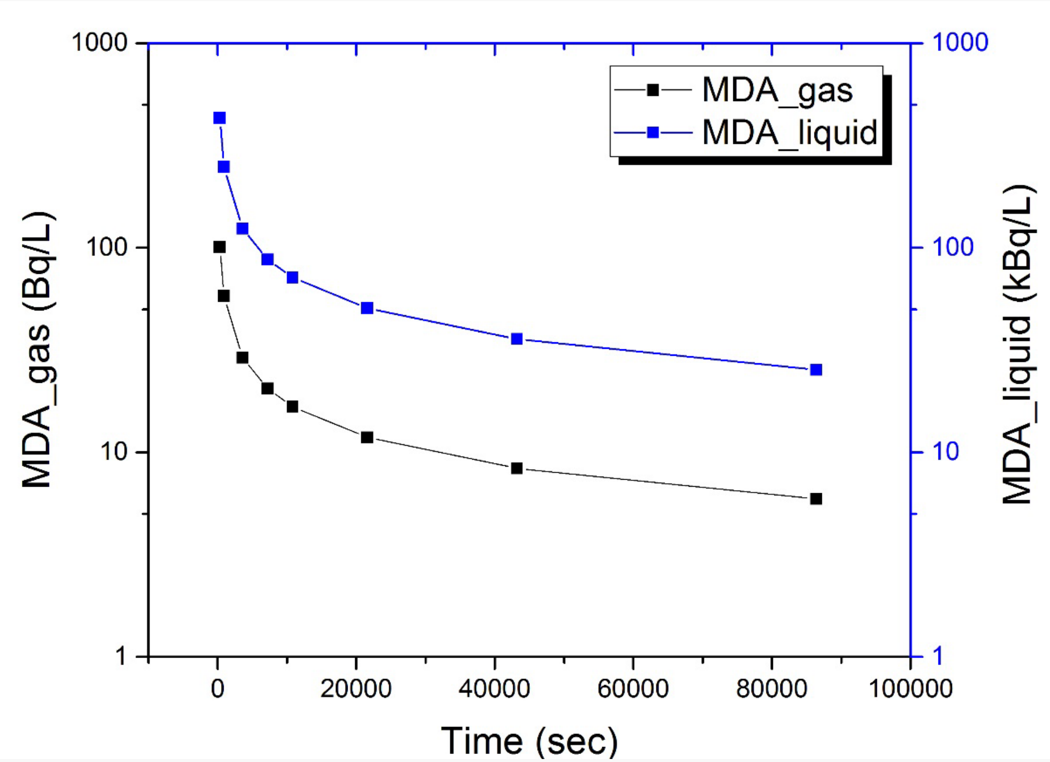


Fig. 10. Minimum detectable activity estimation curve for gaseous and liquid sample.

- The MDA for 5 minute measurement was 100.8 Bq/L for gaseous sample where the activity of gaseous sample was calculated as 58.35 Bq/L.
- For 2 hour measurement, MDA for liquid sample was 87.78 kBq/L while the MDA for commercial tritium monitor (LIQ-X-H3, Technical Associates) for water was 370 kBq/L for 2 hours of measurement time.
- To satisfy the EPA criterion for drinking water (740 Bq/L), background count rate should be reduced by 0.412 in CPM.

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# Conclusion

- A continuous tritium in water monitoring system based on plastic scintillator detector and electrolysis was developed.

- The system had ability to sample the water continuously, gaseous tritium, produced by electrolysis, can be detected by plastic scintillator based detector, and its characteristics were evaluated.

- The developed system showed lower MDA (87.78 kBq/L for 2 hours) than the commercial product (370 kBq/L for 2 hours), but it is not reached to the EPA criterion for drinking water (740 kBq/L).

- For future work, MDA will be decreased by reduce the background count rate using coincidence circuit with two PMTs.

# References

[1] Hee Reyoung Kim, Mun Ja Kang, Geun Sik Choi, "An experiment on the radioactivity characteristics of the tritium contaminated metal sample" Ann. Nucl. Energy, 38 (2011) 1074-1077.
[2] Aurelie M. Soreefan, Timothy A. DeVol, "Proportional counting of tritium gas generated by polymer electrolyte membrane (PEM) electrolysis" J. Radioanal. Nucl. Chem. 282 (2009) 517-521.
[3] Etsuko Furuta, Takao Kawano, A plastic scintillatioin counter prototype, Appl. Radiat. Isot. 104 (2015) 175-180.
[4] Aurelie M. Soreefan, Timothy A. DeVol, "Determinatino of tritium enrichment parameters of a commercially available PEM electrolyzer: a comparison with conventional enrichment electrolysis" J. Radioanal. Nucl. Chem. 282 (2009) 511-515.


Thank you
for your attention