Radon-in-water secondary standard preparation

ERRICCA-2 Kick-off Meeting
D.J. Karangelos, N.P. Petropoulos, E.P. Hinis and S.E. Simopoulos,
NTUA, Greece
Introduction

- Radon-in-water standards are necessary for the calibration and quality control of instruments.
- Availability is limited by the short half-life of radon.
- A method for the generation of such secondary standards from an ordinary radon source is presented.
Solubility of radon in water (1)

The equilibrium state for a radon – air – water system with respect to the diffusion process is described by the dimensionless Ostwald coefficient:

$$ k = \frac{\text{concentration in the liquid phase}}{\text{concentration in the gas phase}} $$
Solubility of radon in water (2)

- Published radon solubility data:
  
  *IUPAC Solubility Data Series, Vol.2*
  *Krypton, Xenon and Radon*
  Clever H.L. (editor)

- Reproduced partly in:
  
  *CRC Handbook of Chemistry and Physics, 74th ed.*
  Lide, D.R. (editor)
  CRC Press, Boca Raton, 1994
The method in principle

- A reference concentration of radon gas is generated inside a calibration chamber.
- A closed circuit with the chamber and a smaller vessel containing water is formed.
- Air is bubbled through the water for a time (5-20 min) sufficient for the system to attain equilibrium.
- The water vessel is then isolated from the circuit and its water content is measured.
- At equilibrium, the radon content of the water can be calculated from the initial concentration in the chamber and the Ostwald coefficient.
The method in detail (1)
The method in detail (2)
The method in detail (3):
The water vessel
The method in detail (4) :
Drawing a sample

The sample can be drawn through the exit valve to avoid contact with the atmosphere.
Calculation of the radon concentration

For our set-up, air volume is much larger than water volume. This allows simplifying assumptions to be made. The radon concentration in the water phase at equilibrium is given by:

\[ C = k \frac{R}{V} \]

where:
R : initial radioactivity in the chamber
V : total volume
k : Ostwald coefficient
Sources of uncertainty

\[
\frac{\delta C}{C} = \sqrt{\left(\frac{\delta R}{R}\right)^2 + \left(\frac{\delta k}{k}\right)^2 + \left(\frac{\delta V}{V}\right)^2}
\]

- **Systematic:**
  - Primary standard
  - Ostwald coefficient
  - Total volume

- **Random:**
  - Temperature – through Ostwald coefficient
The method in detail (5):
Operating Parameters

- Total volume: 1.9 m³
- Maximum water volume: 4lt
- Air flow rate: 10 lt/min
- Radon sources:
  - Pylon RN-2000A
    (Maximum activity 102.8 kBq)
  - Czech Metrological Institute RF200
    (Maximum activity 274.3 kBq)
- Maximum Rn concentration in water: 37 Bq/lt
- Overall systematic uncertainty: 5%
Validation of the method

Water samples with Radon concentration ranging from 3 – 17 Bq/lt were generated. Subsamples of these were measured using an active radon monitor, together with the accessories for water measurement provided by the manufacturer of the instrument.
Radon-in-water instrumentation:
AlphaGuard & Genitron AquaKit (1)

A water sample (~100ml) is transferred to the bubbler. The radon is then expelled by bubbling and led to the ionization chamber of the AlphaGuard radon monitor. Initial radon content of the sample can be calculated from total radon balance.
Radon-in-water instrumentation: AlphaGuard & Genitron AquaKit (2)

- Degassing of the water sample is quantitative – No other calibration factors are needed apart from the original calibration of the AlphaGuard.
- A canister of active charcoal can be incorporated in the measurement set-up, in order to lower background for low-level measurements.
### AlphaGuard measurement results (1)

<table>
<thead>
<tr>
<th>#</th>
<th>Nominal Concentration (Bq/l)</th>
<th>Measured Concentration (Bq/l)</th>
<th>Standard Deviation (Bq/l)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>3.1</td>
<td>3.4</td>
<td>0.32</td>
</tr>
<tr>
<td>2.</td>
<td>3.2</td>
<td>3.5</td>
<td>0.47</td>
</tr>
<tr>
<td>3.</td>
<td>4.7</td>
<td>4.6</td>
<td>0.29</td>
</tr>
<tr>
<td>4.</td>
<td>6.6</td>
<td>6.4</td>
<td>0.43</td>
</tr>
<tr>
<td>5.</td>
<td>8.3</td>
<td>8.5</td>
<td>0.41</td>
</tr>
<tr>
<td>6.</td>
<td>13.8</td>
<td>16</td>
<td>1.1</td>
</tr>
<tr>
<td>7.</td>
<td>16.9</td>
<td>16.4</td>
<td>0.97</td>
</tr>
</tbody>
</table>
AlphaGuard measurement results (2)
The intercept of the least-squares line is not significant statistically.

The slope does not differ statistically from unity.

RMS-deviation is equal to 10%.

Measured values are within overall uncertainty.
An example:
Calibration of a measuring apparatus

Using subsamples from the same set of samples produced for our previous experiment, calibration of a radon-in-water measuring apparatus based on a gas-transfer membrane was performed.
Radon-in-water instrumentation:
RM2000 & Sarad AquaKit (1)

A microporous membrane tube is submerged in the sample (~ 2lt). While the sample is stirred, radon diffuses through the membrane for a fixed amount of time and is led to the counting chamber of an RM2000 monitor. A calibration factor can be determined to give the initial radon concentration of the water sample.
Radon-in-water instrumentation: RM2000 & Sarad AquaKit (2)

- The fraction of radon that diffuses must be determined experimentally – A calibration factor for the membrane is required.
- Ageing or progressive wear of the membrane might be an issue.
- The system can be used for continuous measurement in flowing water.
RM2000 measurement results (1)

<table>
<thead>
<tr>
<th>#</th>
<th>Nominal Concentration in water (Bq/l)</th>
<th>Measured Concentration in air (Bq/l)</th>
<th>Standard Deviation (Bq/l)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>3.1</td>
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<td>0.12</td>
</tr>
<tr>
<td>2.</td>
<td>3.2</td>
<td>1.3</td>
<td>0.15</td>
</tr>
<tr>
<td>3.</td>
<td>4.7</td>
<td>1.9</td>
<td>0.19</td>
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<tr>
<td>4.</td>
<td>6.6</td>
<td>2.8</td>
<td>0.40</td>
</tr>
<tr>
<td>5.</td>
<td>8.3</td>
<td>3.5</td>
<td>0.10</td>
</tr>
<tr>
<td>6.</td>
<td>13.8</td>
<td>6.7</td>
<td>0.35</td>
</tr>
<tr>
<td>7.</td>
<td>16.9</td>
<td>7.5</td>
<td>0.32</td>
</tr>
</tbody>
</table>
RM2000 measurement results (2)
Radon content of the water sample is indeed reproducible from the amount that diffuses through the membrane in given time ($R^2 = 0.99$), for the range of concentrations tested.

For our set-up, the calibration factor is equal to $2.22 \pm 0.05$. 
Conclusions

- A method to generate secondary radon-in-water standard solutions has been given.
- This method can be easily applied by any Laboratory having access to a radon source and a calibration chamber.
- Its ease of use and low running cost make it ideal for the calibration and quality control of instruments and measurement methods.