Research on the Conversion Coefficient for Measuring Radon Concentration in Water Based on Bubbling Method

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Research on the conversion coefficient for measuring radon concentration in water based on bubbling method

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Abstract: Radon is soluble in water, and the radon in drinking water is one of the sources of indoor radon. The China national standard for drinking water (GB5749-2006) indicates that the radon concentration in drinking water cannot be higher than 300pCi/L. The widely used method for measuring radon concentration in water is to use the RAD7 and the H₂O accessory which provided by the DurrIDGE Company for bubbling measurement. There are two kind sample bottles: 40ml and 250ml; the measurement steps: bubbling for 5 minutes, and measuring for 20 minutes. For the 40ml sample volume the conversion coefficient is around 25. For the 250ml sample volume the conversion coefficient is around 4. Since the price of the accessory is more than one thousand dollars, and the drying tube of the accessory is small, it needs to be replaced after each measurement which makes the operation is troublesome. We used a big drying tube to perform the radon concentration in water measurement. A new model for description the radon concentration in the gas circuit is proposed, and the conversion coefficient in any measurement condition is obtained. Any volume of sample bottle and drying tube can be used, selecting the suitable bubbling time and measuring time, the radon concentration in water can be obtained.

Keywords: Radon concentration in water measurement; radiation measurement; RAD7; H₂O accessory; bubbling method; conversion coefficient

29 ***Introduction***

30 Radon (Rn-222) is a chemically inert and odorless radioactive gas from the
31 uranium (U-238) decay chain ([Tan et al. 2020](#)). And its half-life is 3.8 days. It is one
32 of the most important naturally occurring radioactive nuclides and contributes to more
33 than 50% of the ionizing radiation dose of humans ([UNSCEAR 2000](#)). In addition,
34 atmospheric particles containing Rn decay products are of high relevance to humans
35 since they may be deposited in the respiratory tract, causing high radiation hazards
36 ([Eichler B et al. 2000](#)). Radon is soluble in water, which allows radon to enter the
37 house through groundwater aquifers and indoor pipelines. The radon gas in water is
38 mainly derived from the decay and release of radium dissolved in water. It should be
39 noted that around 1–2% of radon in the air comes from drinking water ([USEPA 1991](#)).
40 The sanitary standard for drinking water ([GB5749-2006](#)) stipulates that the
41 concentration of radon in drinking water cannot be higher than 300pCi/L. Despite
42 being less of a health risk than its airborne counterpart, radon carried in water is often
43 a major transportation mechanism leading to high airborne radon concentration
44 ([Welch LE. 1994](#)). As a result of its noble gas configuration radon behaves
45 biologically and chemically inert. Due to that property, it is applied as environmental
46 tracer in hydrology, geochemistry, and oceanography for tracking of transport
47 pathways in aquifers and surface water systems and for the quantification of
48 admixture rates ([Axel Schmidt et al. 2008](#)). Especially for seismology, earthquakes
49 and other similar geological activities like volcanic explosion will affect the
50 concentration of radon in groundwater, which is usually measured for earthquake
51 prediction ([Igarashi G et al.1995. Pradeep Talwani et al. 1980. Liu et al. 1984](#)).

52 Therefore, the measurement of radon concentration in water is a very meaningful
53 work. Based on different purpose and requirement, the radon-in-water measurement
54 methods and systems can be quite different. Commonly used methods are liquid
55 scintillation counting (LSC) method, gamma spectrometry method and emanometric
56 method, which are recommended by ISO 13164 ([ISO 2013](#)). The measuring method
57 includes diffusion, degassing membrane method and the bubbling method. In

bubbling method, the balance time of radon-in-water and radon-in-air concentration is shortened by increasing the contact area between water and air with bubbling probe (Wang et al. 2020). Therefore, this method has been widely used, and new measurement methods and equipment have been developed, such as an improved version of the ionization chamber, a series of equipment based on bubble chambers and RAD7 monitors (D Lane-Smith. 2001. H Dulaiova et al. 2005. Dimova et al. 2009. Stieglitz et al. 2010. Lee et al. 2014), as well as commercial instruments like RAD H₂O (Malik et al. 2014), RAD AQUA and some other devices (Teng. 1980).

The widely used device for bubbling to measure radon concentration in water is to use the RAD7 and the H₂O accessory which provided by the DurrIDGE Company. For the 40ml sample volume the conversion coefficient is around 25. For the 250ml sample volume the conversion coefficient is around 4 (<https://durrIDGE.com>). Since the price of the accessory is more than one thousand dollars, and the drying tube of the accessory is small, it needs to be replaced after each measurement which makes the operation is troublesome. We used a big drying tube to perform the radon concentration in water measurement. A new model for description the radon concentration in the gas circuit is proposed, and the conversion coefficient in any measurement condition is obtained. Any volume of sample bottle and drying tube can be used, selecting the suitable bubbling time and measuring time, the radon concentration in water can be obtained.

Theoretical Descriptions

Measurement process

The water radon measurement device based on the bubbling method is shown in Fig.1. The method for measuring radon concentration in water by the above-mentioned bubbling method water includes such process as follows:

Fig.1 Water radon measurement device based on bubbling method

(1) Pour the water sample to be tested into the water sample measuring bottle and close the cap tightly. The air outlet of the inlet pipe of the water sample bottle is in the water sample to be tested and is close to the bottom of the water sample bottle, and the air inlet of the gas outlet pipe of the water sample bottle is located on the liquid surface of the water sample to be tested.

(2) Start the air pump and control the flow rate of the air pump to make air bubble into the water sample through the intake pipe of the water sample bottle. The generated bubbles will fully contact the water sample during the ascent process, and the radon in the water will enter the bubbles through the diffusion effect. At the same time, the radon in the water sample is carried out, and enters the drying tube through the air outlet pipe of the water sample bottle, and then passes through the air pump, and then enters the internal cell of the RAD7 radon monitor.

(3) After several periods of measurement, the water radon concentration can be obtained from the measurement value of RAD7.

Assuming that the initial radon concentration in water is C_0 , the initial concentration of radon and Po-218 in the air circuit of the device for measuring radon concentration in water by the bubbling method is 0. Thus, the water sample measuring

bottle was replaced with a bottle filled with charcoal before the experiment to adsorb the radon in the air loop. Using the pump of the RAD7, the air is continuously circulated in the loop, and the radon concentration in the loop will reduce until it drops to 0.

Calculation process

Assuming the water radon concentration in the water sample measuring bottle is $C_1(t)$, the radon concentration in the air circuit is $C_2(t)$, and the Po-218 concentration in the internal cell of RAD7 is $C_3(t)$.

Assuming that the radon concentration in the bubble has reached equilibrium with the radon in the water during the process from the inlet pipe of the water sample measuring bottle to the water surface, the following relationship exists between $C_1(t)$ and $C_2(t)$:

$$\frac{dC_1(t)}{dt} = \frac{L[C_2(t) - XC_1(t)]}{V_1} \quad (1)$$

where V_1 is the water sample volume; L is the flow rate of the pump. And X is the ratio of radon in air to radon in water at equilibrium. According to the reference (Weigel. 1978):

$$K_{w/air} = \frac{C_w}{C_{air}} = f(T) = 0.105 + 0.405e^{-0.0502T[^\circ\text{C}]} \quad (2)$$

where T is the temperature of the water sample ($^\circ\text{C}$). So $X = \frac{C_{air}(t)}{C_w(t)} = \frac{1}{K_{w/air}}$, the value of X is related to temperature.

$$\frac{dC_2(t)}{dt} = \frac{L[XC_1(t) - C_2(t)]}{V_2} \quad (3)$$

where V_2 is the volume of air circuit.

According the measurement principle of the RAD7, $C_2(t)$ and $C_3(t)$ have the following relationship (Tan et al. 2011. Tan et al. 2013):

$$\frac{dC_3(t)}{dt} = \lambda_{Po} C_2(t) - \lambda_{Po} C_3(t) \quad (4)$$

where λ_{Po} is the decay constant of Po-218.

For $C_1(0) = C_0, C_2(0) = 0, C_3(0) = 0$, the solutions of Eq.(1),(3) and (4) are:

$$C_1(t) = \frac{C_0 V_1}{V_1 + V_2 X} + \frac{C_0 V_2 X e^{-t \frac{LV_1 + LV_2 X}{V_1 V_2}}}{V_1 + V_2 X} \quad (5)$$

$$C_2(t) = \frac{C_0 V_1 X}{V_1 + V_2 X} - \frac{C_0 V_1 X e^{-t \frac{LV_1 + LV_2 X}{V_1 V_2}}}{V_1 + V_2 X} \quad (6)$$

$$C_3(t) = \frac{C_0 V_1 X}{V_1 + V_2 X} - \frac{C_0 L V_1 X e^{-\lambda_{Po} t}}{L V_1 + L V_2 X - V_1 V_2 \lambda_{Po}} + \frac{C_0 V_1^2 V_2 X \lambda_{Po} e^{-t \frac{LV_1 + LV_2 X}{V_1 V_2}}}{(V_1 + V_2 X)(L V_1 + L V_2 X - V_1 V_2 \lambda_{Po})} \quad (7)$$

Assuming the Sampling time is t_1 , and the internal cell of the RAD7 radon monitor is applied with high voltage. Set the suitable sampling time, the radon concentration inside the device is evenly mixed. In the period of $t_1 \sim t_2$, the reading of RAD7 is the Po-218 concentration (Tan et al. 2015). Assuming m is the total Po-218 count of RAD7 in the measuring period of $t_1 \sim t_2$:

$$m = \int_{t_1}^{t_2} C_{Po}(t) dt = \frac{C_0 V_2^2 X \lambda_{Po} (e^{-\frac{L+LX}{V_2} \frac{V_2}{V_1} t_1} - e^{-\frac{L+LX}{V_2} \frac{V_2}{V_1} t_2})}{L^2 (1 + \frac{V_2}{V_1} X)^2 (1 + X \frac{V_2}{V_1} - \frac{V_2 \lambda_{Po}}{L})} - \frac{C_0 X \times (t_1 - t_2)}{1 + X \frac{V_2}{V_1}} - \frac{C_0 X (e^{-\lambda_{Po} t_1} - e^{-\lambda_{Po} t_2})}{\lambda_{Po} (1 + X \frac{V_2}{V_1} - \frac{V_2 \lambda_{Po}}{L})} \quad (8)$$

n is the average value measured by the RAD7 during $t_1 \sim t_2$:

$$n = \frac{\int_{t_1}^{t_2} C_{Po}(t) dt}{t_2 - t_1} = \frac{m}{t_2 - t_1} = \frac{\frac{C_0 V_2^2 X \lambda_{Po} (e^{-\frac{L+LX}{V_2} \frac{V_2}{V_1} t_1} - e^{-\frac{L+LX}{V_2} \frac{V_2}{V_1} t_2})}{L^2 (1 + \frac{V_2}{V_1} X)^2 (1 + X \frac{V_2}{V_1} - \frac{V_2 \lambda_{Po}}{L})} - \frac{C_0 X \times (t_1 - t_2)}{1 + X \frac{V_2}{V_1}} - \frac{C_0 X (e^{-\lambda_{Po} t_1} - e^{-\lambda_{Po} t_2})}{\lambda_{Po} (1 + X \frac{V_2}{V_1} - \frac{V_2 \lambda_{Po}}{L})}}{t_2 - t_1} \quad (9)$$

k is the conversion coefficient, and it is equal to the ratio of the initial concentration C_0 of the radon in water to the average value measured by the RAD7

142 within $t_1 \sim t_2$:

$$\begin{aligned}
 k &= \frac{C_0}{n} = \frac{C_0 \times (t_2 - t_1)}{m} \\
 &= \frac{(t_2 - t_1)}{\frac{X \lambda_{p_o} (e^{-\frac{L+LX}{V_2} \frac{V_2}{V_1} t_1} - e^{-\frac{L+LX}{V_2} \frac{V_2}{V_1} t_2})}{\left(\frac{L}{V_2}\right)^2 (1 + \frac{V_2}{V_1} X)^2 (1 + X \frac{V_2}{V_1} - \frac{V_2 \lambda_{p_o}}{L})} - \frac{X \times (t_1 - t_2)}{1 + X \frac{V_2}{V_1}} - \frac{X (e^{-\lambda_{p_o} t_1} - e^{-\lambda_{p_o} t_2})}{\lambda_{p_o} (1 + X \frac{V_2}{V_1} - \frac{V_2 \lambda_{p_o}}{L})}} \quad (10)
 \end{aligned}$$

144

145 If the measurement time is much long, that is, $t_2 \gg t_1$, the equation (10) can be

146 simplified as:

$$\begin{aligned}
 k &\approx \frac{(t_2 - t_1)}{\frac{X \lambda_{p_o} e^{-\frac{L+LX}{V_2} \frac{V_2}{V_1} t_1}}{\left(\frac{L}{V_2}\right)^2 (1 + \frac{V_2}{V_1} X)^2 (1 + V_2 (\frac{X}{V_1} - \frac{\lambda_{p_o}}{L}))} - \frac{X \times (t_1 - t_2)}{1 + X \frac{V_2}{V_1}} - \frac{X e^{-\lambda_{p_o} t_1}}{\lambda_{p_o} (1 + V_2 (\frac{X}{V_1} - \frac{\lambda_{p_o}}{L}))}} \quad (11)
 \end{aligned}$$

148

149 If L is large, $\frac{X}{V_1} \gg \frac{\lambda_{p_o}}{L}$, $\frac{X}{V_1} - \frac{\lambda_{p_o}}{L}$ is approximately equal to $\frac{X}{V_1}$, and the

150 equation (11) can be simplified to:

$$\begin{aligned}
 k &\approx \frac{(t_2 - t_1)}{\frac{X \lambda_{p_o} e^{-\frac{L+LX}{V_2} \frac{V_2}{V_1} t_1}}{\left(\frac{L}{V_2}\right)^2 (1 + \frac{V_2}{V_1} X)^3} - \frac{X \times (t_1 - t_2)}{1 + X \frac{V_2}{V_1}} - \frac{X e^{-\lambda_{p_o} t_1}}{\lambda_{p_o} (1 + X \frac{V_2}{V_1})}} \quad (12)
 \end{aligned}$$

152 If $V_2 \gg V_1$, $\frac{V_2}{V_1} X \gg 1$, and $1 + \frac{V_2}{V_1} X$ are approximately equal to $\frac{V_2}{V_1} X$. The

153 equation (12) can be simplified to:

$$k \approx \frac{(t_2 - t_1)}{\frac{\lambda_{Po} e^{-\lambda_{Po} t_1}}{L^2 \frac{V_2}{V_1^3} X^2} - \frac{(t_1 - t_2)}{\frac{V_2}{V_1}} - \frac{e^{-\lambda_{Po} t_1}}{\lambda_{Po} (\frac{V_2}{V_1})}} \quad (13)$$

155 If L is large enough, $\frac{\lambda_{Po} e^{-\lambda_{Po} t_1}}{L^2 \frac{V_2}{V_1^3} X^2}$ approaches 0, and the equation (13) can be

156 simplified to:

$$k \approx \frac{(t_2 - t_1)}{\frac{(t_2 - t_1)}{\frac{V_2}{V_1}} - \frac{e^{-\lambda_{Po} t_1}}{\lambda_{Po} (\frac{V_2}{V_1})}} \quad (14)$$

158 The decay constant of Po-218 is 0.228 min^{-1} , when $t_2 - t_1 \gg \frac{e^{-\lambda_{Po} t_1}}{\lambda_{Po}}$, the
159 equation (14) can be simplified to:

$$k \approx \frac{(t_2 - t_1)}{\frac{(t_2 - t_1)}{\frac{V_2}{V_1}}} = \frac{V_2}{V_1} \quad (15)$$

161 For different sample bottle, assuming $k_1 = \frac{V_{21}}{V_{11}}$, $k_2 = \frac{V_{22}}{V_{12}}$, if $V_{21} = V_{22}$, then

162 $\frac{k_1}{k_2} = \frac{V_{12}}{V_{11}}$. According to the H₂O accessory manual, the ratio of the conversion

163 coefficient between 40 ml and 250 ml sample bottle is 25 to 4, and it is consistent
164 with the equation (15).

165 In summary, equation (10) can be used to calculate the radon concentration in the
166 water sample. When the corresponding conditions are suitable, the water radon
167 concentration can be calculated by equation (11) or (12) or (13) or (14) or (15).

168

Results and Discussion

Three experiments are performed. 200ml and 500ml water sample bottle are used in these experiments. Take out a bucket of water from the lake and pour it into two kind water sample bottles in each experiment.

The volume of the gas circuit of the 200ml water sample bottle device is 1.467L, and the volume of the gas circuit of the 500ml water sample bottle device is 1.456L. The pump flow rate of RAD7 is 0.7L/min. The setting of the RAD7 is: mode, sniff; cycle, 5minutes; recycle, 8.

The pump works for 15 minutes, which can make the gas in the pipeline circulate 7 times, so that the gas in the pipeline is evenly mixed. The measurement is completed within 15-40min. Table 1 lists all the measurement data.

Table 1 RAD7 readings in three experiments

Experiment No.	Sample bottle volume	15-20min Reading (Bq/m ³)	20-25min Reading (Bq/m ³)	25-30min Reading (Bq/m ³)	30-35min Reading (Bq/m ³)	35-40min Reading (Bq/m ³)
1	200ml	40±180	110±220	100±200	100±200	40±170
1	500ml	180±290	150±240	260±280	150±280	220±270
2	200ml	206±250	34.2±165	137±222	137±222	240±263
2	500ml	309±286	309±286	446±326	240±263	343±296
3	200ml	68.3±187	68.3±187	34.2±165	68.3±187	103±205
3	500ml	205±249	171±236	34.2±165	275±275	171±236

According to the equation (2), if T=20°C, $K_{w/air} \approx 0.25$, then $X = \frac{C_{air}(t)}{C_w(t)} = 4$.

The conversion coefficient and the radon concentration in water can be obtained by Eq. (10). Table 2 lists the radon concentration in water and the conversion coefficient in the three experiments.

Table 2 Radon concentration in water and the conversion coefficient

Experiment No.	Sample bottle volume	Conversion coefficient(k)	Measured average radon concentration $\pm 2\sigma$ (Bq/m^3)	Radon concentration in water $\pm 2\sigma$ (Bq/m^3)
1	200ml	7.63	78 ± 66	595 ± 504
1	500ml	3.18	192 ± 85	611 ± 270
2	200ml	7.63	151 ± 141	1152 ± 1076
2	500ml	3.18	329 ± 134	1046 ± 426
3	200ml	7.63	68 ± 44	519 ± 336
3	500ml	3.18	171 ± 153	544 ± 487

189 The results listed in Table 3 show that the radon concentrations in water in two
 190 kind bottles are similar in each experiment. The differences of the radon
 191 concentrations in water among the three experiments are caused by difference of the
 192 sampling time and location of the lake.

193 Assuming C_0 is $600 \text{ Bq}/\text{m}^3$, Fig. 2 and 3 illustrate the concentration change $C_1(t)$,
 194 $C_2(t)$, and $C_3(t)$ in the 200ml and 500ml sample bottle by equation (5), (6) and (7).

195

196

Fig. 2 Concentration change of 200ml water sample bottle

Fig. 3 Concentration change of 500ml water sample bottle

According the manual of Durrige Company, the conversion coefficients for the 250ml and 40ml water sample bottles are 4 and 25 respectively, the volume of the gas circuit is 0.88L, and $\frac{C_{air}(t)}{C_w(t)} = 4$. Substituting the same parameters into equation (10), the k values are 4.15 and 24.47 respectively, which are approximately equal to the conversion coefficients 4 and 25 from the manual of Durrige Company.

Conclusion

The H₂O accessory of Durrige Company based on the principle of bubbling method is wide used in the world for measuring radon concentration in water. However, there are only 40ml and 250ml water sample bottle, and the accessory is expensive. Moreover, the drying tube of the accessory is so small, which requires frequent replacement of the desiccant. A novel model is proposed to determine the concentration of radon in water by using any volume of sample bottle and drying tube, which will broaden the application of the device. Substituting the parameters of the H₂O accessory of Durrige Company into the novel model, the conversion coefficient

calculated by the novel model is similar to that from the manual of Durrige Company.
It means that the conversion coefficient obtained by the novel model is accurate and
reliable.

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Availability of data and materials All data generated or analyzed during this study are
included in this published article.

Competing interests The authors declare that they have no competing interests

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