Dispersal Rate of Radon-219 from Aqueous Radium-223 Solution Containing Sodium Chloride/Citrate

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The airborne dispersion of 223Ra and its descendent nuclides from an aqueous 223Ra solution containing sodium chloride/citrate was investigated. The dispersion of 223Ra was not detected, and γ-rays of 211Pb and 211Bi caused by the 219Rn dispersion were observed by means of γ-ray spectrometry. The detected radioactivity of 211Pb was found to decrease as the inner diameter of the vessel containing the 223Ra solution was decreased because of the decreasing surface area of the solution exposed to the air. The dispersal rate of 219Rn from the aqueous solution was determined by the ratio of the number of the dispersed 219Rn atoms calculated from the detected radioactivity of 211Pb on the samples. The values at room temperature were in the range of \((1.6 \pm 0.1) \times 10^{-3} - (2.0 \pm 0.1) \times 10^{-2}\). These rates were found to decrease with decreasing exposed surface area of the vessel used.

Key Words: Radium-223, Radon-219, Targeted alpha therapy, Dispersal rate

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1. Introduction

Radium-223 is an α-particle emitter commonly used in targeted alpha therapy.1) In radionuclide therapy, α-particles have an advantage over β-particles because α-particles efficiently attack targeted cells such tumor cells by high linear energy transfer and do not damage the surrounding normal cells due to the low penetration depth of alpha particles (2–10 cells lengths).2) In fact, 223RaCl₂ has been approved in Japan as a drug (Xofigo®) for metastatic castrate-resistant prostate cancer that is metastatic to bone.3) The clinical application of 223Ra has rapidly grown.4) For instance, the use of 223Ra in Japan accounts for approximately one-third of the radionuclide therapy usage in 2017.5) Recently, it has been reported that 211Bi, a daughter nuclide of 223Ra, was detected from exhaled breath and surroundings of patients administered Xofigo®.6) To the best of our knowledge, information on the dispersal rate of 223Ra and its descendant nuclides from an aqueous solution is still rare. The dispersal data relating to 223Ra and its descendant nuclides such as dispersal rates from an aqueous solution having the same chemical composition with Xofigo® are of great significance from the view point of radiation safety management. Figure 1 shows the radioactive decay series of 223Ra. A nuclide of 223Ra finally disintegrates to stable 207Pb via four α- and two β-particle-emission processes. 219Rn is a noble-gas isotope with a half-life of 3.96 s and is generated by the decay of 223Ra as an intermediate descendent in the decay series

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of $^{223}$Ra. Therefore, it is important to determine the dispersal rates of $^{223}$Ra and $^{219}$Rn. In the present study, we investigated the airborne dispersion of $^{219}$Rn and $^{223}$Ra from an aqueous solution of sodium chloride/citrate using a radioactive-materials-collection apparatus composed of a cylinder and a container in which filter papers and charcoal cartridges were loaded. The dispersion of $^{223}$Ra was below the detectable level, and the radioactivity of $^{211}$Pb originating from the dispersion of $^{219}$Rn was identified by the $\gamma$-ray spectrometry. The dispersal rate from the aqueous solution was evaluated by the ratio of the dispersed $^{219}$Rn atoms, which was calculated from the radioactivity of $^{211}$Pb atoms detected in the parts of the collection apparatus, to the number of $^{219}$Rn atoms in the aqueous solution during the collection time. This ratio is used because it is difficult to directly measure the dispersal rate of $^{219}$Rn due to its short lifetime. The dependence of the dispersal rate of $^{219}$Rn on the inner diameter of the vessel was also investigated.

2. Experimental Section

2-1. Materials

Aqueous solutions (6 mL each) containing $^{223}$Ra$^{2+}$, 0.1 mol/L of NaCl, and 0.03 mol/L of sodium citrate, which were the same composition with Xofigo®, were kindly supplied by Bayer HealthCare Pharmaceuticals Inc.

2-2. Setup of the radioactive-materials-collection apparatus and experiments for collection of dispersed radionuclides

The experimental setup and the filter papers and cartridge container used for the collection of radioactive materials are shown in Figures 2 and 3, respectively. The container was custom-made based on the structure of the radioactive iodine sampling holder, SIBATA model RI-55.

The position of the exhaust pipe was modified in the custom-made container. The body of the container is exhaust pipe, and air inlet are made of duralumin, brass, and SUS304 mesh (mesh size: 1.9 mm). In the container, filter papers and cartridges were placed downstream from the air inlet to match the order of a glass fiber filter paper (AP-2005500, Merck), a charcoal impregnated filter paper (CP-20, ADVANTEC), and two charcoal cartridges (CHC-50, ADVANTEC). The particle retention efficiency for the glass fiber filter paper AP-2005500 is >99.9% for a particulate matter (PM2.5). The collection efficiencies of CP-20 for I$_2$, the mixture of organic (20%) and inorganic iodine (80%), and the mixture of organic (80%) and inorganic iodine (20%) were reported to 90%, avg. 65%, and avg. 25%, respectively. The collection efficiencies of CHC-50 were reported more than 96% for the mixture of organic (80%) and inorganic iodine (20%), and more than 95% for volatile phosphorus, sulfur, and mercury. Viton O-rings were used to seal the filter papers and cartridges in the container. The container was set at the top of an acrylic cylinder (70 mm I.D., 80 mm O.D., and 480 mm length) and the interspace between the container and the acrylic cylinder was sealed with a Viton O-ring. The whole inside area of the acrylic cylinder was covered with pieces of the polyethylene terephthalate (PET) film of 100 µm in thickness (T60, TORAY). Two 6-mL solutions of $^{223}$Ra$^{2+}$ were mixed, and a 1.0-mL aliquot of the solutions was subjected to $\gamma$-ray spectrometry with a Ge detector to quantify the radioactivity of $^{223}$Ra. Then, the entire solution (12 mL) was poured into a 100-mL, 50-mL, or 20-mL beaker, two 10-mL beakers, or seven microtubes (I.D.: 8 mm). When the 100-mL beaker was used, the outside of the beaker was covered with the paraffin film (PARAFILM® (PM-996), Bemis Flexible Packaging). When using a 50-mL, 20-mL, or 10-mL beaker(s) or the microtubes, each sample was set in a 100-mL beaker used as a sample holder, the outside and inside surfaces of which were covered with the paraffin film. The vessel was put on a magnetic stirrer at the bottom of the acrylic cylinder. The distance between the bottom of the cylinder and...
the magnetic stirrer (CHPS-170DS, AS ONE) whose top face was covered with the paraffin film was set to 30 mm. The solution(s) were continuously stirred with PTFE stirring bar(s) using the magnetic stirrer. A low volume suction pump (MP-Σ300NII, SIBATA or LV-40BW, SIBATA) and the sampling container were connected with a polyvinyl chloride tube. The air in the cylinder was suctioned for 1 h with an exhaust rate of 1–4 L/min using the MP-Σ300NII pump and 5–30 L/min using the LV-40BW pump at room temperature (23 ± 2°C), and the dispersed radioactive materials were collected in parts of the collection apparatus such as the PET film on the inside wall of the cylinder, filter papers, and cartridges.
2-3. Measurement of radiation

A high purity Ge detector (BE2020, CANBERRA) was used for γ-ray spectrometric measurements. All the signals from the Ge detector were converted from analog to digital and were sorted by an all-in-one multichannel analyzer with 4096 channels (MCA-7a, SEIKO EG&G). After the collection of radioactive materials, the γ-rays of the PET films, the glass fiber filter paper, the charcoal impregnated filter paper, the two charcoal cartridges, the O-rings, and the paraffin film pieces that covered the glass vessel and the magnetic stirrer were separately measured to quantify the amounts of dispersed radioactive nuclides. The radioactivity of 211Pb and 211Bi at the end time of the collection was evaluated from γ-rays observed at 404.9 keV for 211Pb and at 351.1 keV for 211Bi. The radioactivity of 223Ra was determined using the γ-ray peak at 154.2 keV. For the energy calibration and calculation of the detection efficiency of the Ge detector, a nine-nuclide mixed wide area activity standard gamma source (MX421, 60 mmϕ, Japan Radioisotope Association) was used. After the dispersal experiment, each sample was placed in a polyethylene bag (size: 60 × 60 mm) and set at a distance of 5 cm from the Ge detector, and then γ-ray measurement was performed for 200–10000 s. The radioactivity in each sample determined by averaging data collected from twice experiments.

3. Results and Discussion

3-1. Identification of dispersed radionuclide

Figure 4 shows typical γ-ray spectra of the original 223Ra solution and the glass fiber filter paper after dispersion for 1 h. Table 1 summarizes the radioactivity of 211Pb collected on the pieces of the film and in the filter papers and cartridges. The γ-ray peaks of 211Pb and 211Bi were found in the γ-ray spectra, while the γ-peaks of 223Ra were not observed. The radioactivity of the 223Ra solution before and after the dispersal experiment, was equal within the error. These results indicate that the dispersion of 223Ra is below the detectable level. In the experiment using a 100-mL beaker, the upper limit of the dispersal rate of 223Ra was 2.4 × 10^-6 which was calculated from the measurable limit of the Ge detector used. The decay of the radioactivity of 211Pb and that of 211Bi show linear correlations between the logarithms of the radioactivity and the elapsed time. The half-lives determined from the slopes fitted by least-squares analysis are 35.8 ± 0.3 min for 211Pb and 36.2 ± 0.2 min for 211Bi. These values are in good agreement with the half-life of 211Pb (T1/2 = 36.1 min). 211Pb is generated by the decay of 223Ra into 219Rn and 215Po (See Figure 1). 211Pb decayed with its own half-life and not with that of 223Ra. This indicates that the detected 211Pb on parts of the collection apparatus is not in radioactive equilibrium with 223Ra, and the

![Fig. 4. γ-ray spectra of (a) the glass fiber filter after the collection of the dispersed radionuclides and (b) the aqueous 223Ra²⁺ solution.](image-url)
dispersion of $^{211}\text{Pb}$ does not originate from the dispersion of $^{223}\text{Ra}$. $^{219}\text{Rn}$ and $^{215}\text{Po}$ were not detected on the parts of the collection apparatus because it immediately decayed out to give $^{211}\text{Pb}$ during and after the experiment due to the very short half-lives of $^{219}\text{Rn}$ (3.96 s) and $^{215}\text{Po}$ (1.78 ms). Rn is well known to form a gaseous single-atom molecule under normal conditions. In practice, the dispersion of $^{222}\text{Rn}$ from water has been previously reported.\(^{10}\) Although $^{215}\text{Po}$ may be also dispersed, its half-life is too short to contribute to the dispersion in the present study.\(^{11}\) The direct dispersion of $^{211}\text{Pb}$ and $^{211}\text{Bi}$ is negligible based on the fact that the most stable oxidation states of Pb and Bi in aqueous solutions are Pb$^{2+}$ and Bi$^{3+}$, respectively\(^{12,13}\) because it is known that volatility of an ionic species is quite small compared with that of a neutral species.\(^{14}\) Therefore, the detection of $^{211}\text{Pb}$ and $^{211}\text{Bi}$ on the parts of the collection apparatus is due to the dispersion of $^{219}\text{Rn}$ molecules from the aqueous solution.

### 3-2. Distribution of $^{211}\text{Pb}$ in the radioactive-materials-collection apparatus

The distribution of $^{211}\text{Pb}$ in the collection apparatus was investigated by varying the exhaust rate of the suction pump from 1 to 30 L/min at room temperature using a 100-mL glass beaker. Table 1 lists the radioactivity of $^{211}\text{Pb}$ found on the parts of the collection apparatus. At 1 L/min, the sum of detected $^{211}\text{Pb}$ radioactivity calculated at the end of the air exhausting experiment is $(1.1 \pm 0.1) \times 10^2$ kBq, of which $15 \pm 4$ kBq is in the glass filter paper and $99 \pm 6$ kBq is on the film on the cylinder inside wall, while the other parts are $< 1$ kBq. At 30 L/min, the sum is $(1.4 \pm 0.4) \times 10^2$ kBq, and the radioactivity is $63 \pm 25$ kBq in the glass fiber filter paper, $30 \pm 17$ kBq on the cylinder inside wall, $30 \pm 4$ kBq in the first charcoal cartridge, and $17 \pm 2$ kBq in the charcoal impregnated filter paper. Rn is likely to pass through the glass fiber filter paper as it is a gaseous neutral molecule. The nuclides generated after the disintegration of $^{219}\text{Rn}$ become trapped within the filters and are adsorbed on the film on the inside wall of the cylinder because the produced atoms exist as non-volatile ions. The linear flow rate, $V_f$ [mm/s], is calculated as follows:

$$V_f = \frac{10^6 V_e}{\pi \left(\frac{D}{2}\right)}.$$

(1)

where $V_e$ [L/s] is the exhaust rate and $D$ [mm] is the inner diameter of the cylinder. The time required for an atom dispersed from the solution to reach the air inlet of the container ($T_R$ [s]) is expressed as

$$T_R = \frac{L}{V_f}.$$

(2)

where $L$ [mm] is the length from the upper end of the vessel to the air inlet of the container. The ratio ($r$) of the number of the dispersed and disintegrated atoms ($N_o$) to the number of dispersed atoms ($N_0$) within the elapsed $T_R$ is derived as

| Exhaust rate [L/min] | 1       | 4       | 15      | 30      |
|----------------------|---------|---------|---------|---------|
| Radioactivity of $^{223}\text{Ra}$ [MBq] | 8.2 ± 0.1 | 8.2 ± 0.2 | 8.5 ± 0.1 | 8.7 ± 0.1 |
| Radioactivity of $^{211}\text{Pb}$ [kBq] |         |         |         |         |
| PET film             | 99 ± 6  | 72 ± 9  | 41 ± 2  | 30 ± 17 |
| Glass fiber filter paper | 15 ± 4 | 59 ± 5  | 63 ± 1  | 63 ± 25 |
| Charcoal impregnated filter paper | 0.22 ± 0.18 | 0.66 ± 0.44 | 7.2 ± 1.1 | 17 ± 2  |
| Charcoal cartridge 1 | 0.017 ± 0.002 | 0.20 ± 0.11 | 3.8 ± 0.1 | 30 ± 4  |
| Charcoal cartridge 2 | 0.041 ± 0.031 | 0.10 ± 0.02 | 0.068 ± 0.007 | 0.12 ± 0.04 |
| O-ring               | 0.15 ± 0.10 | 0.33 ± 0.02 | 0.47 ± 0.06 | 0.51 ± 0.10 |
| Paraffin film (stirrer) | 0.025 ± 0.014 | 0.052 ± 0.004 | 0.14 ± 0.11 | 0.15 ± 0.07 |
| Paraffin film (beaker) | 0.18 ± 0.11 | 0.18 ± 0.12 | 0.17 ± 0.05 | 0.098 ± 0.048 |
| Sum                  | $(1.1 \pm 0.1) \times 10^2$ | $(1.3 \pm 0.1) \times 10^2$ | $(1.2 \pm 0.1) \times 10^2$ | $(1.4 \pm 0.4) \times 10^2$ |
The $T_R$ value is calculated to be $1.0 \times 10^2$ s with $V_e = 1.0$ L/min, $D = 70$ mm, and $L = 450$ mm for the cylinder of the collection apparatus from Equation 2. The $S$ value is calculated to be $1.3 \times 10^{-8}$ with the $T_{1/2}$ of $^{219}$Rn (3.96 s) from Equation 3. Therefore, at an exhaust rate of 1 L/min, more than 99.99% of $^{219}$Rn atoms decay in the cylinder before reaching the air inlet of the container. This indicates that all of the dispersed $^{219}$Rn atoms and their descendant nuclides are trapped on the parts of the collection apparatus at an exhaust rate of 1 L/min. It is calculated that 30% and 55% of the dispersed $^{219}$Rn atoms in the cylinder are calculated to reach the cartridge at exhaust rate of 15 and 30 L/min, respectively, before the decay of $^{219}$Rn. Therefore, some of $^{219}$Rn atoms might pass through the cartridge at exhaust rate of 15 and 30 L/min, although the sums of the $^{211}$Pb radioactivity values are similar to one another in the range of exhaust rates of 1–30 L/min as listed in Table 1. The distribution of the radioactivity in the collection apparatus varies somewhat by exhaust rate. At 1 L/min, the almost all radioactivity is on the PET film on the inside wall of the cylinder. This is because the dispersed Rn atoms disintegrated in the cylinder and the generated ions of the descendant nuclides are adsorbed on the film on the inside wall of the cylinder. At 30 L/min, radioactivity is detected in the other parts including the charcoal impregnated filter paper and the charcoal cartridge which are set downstream of the collection apparatus, and the amount distributed on the film on the inside wall of the cylinder is rather small. This suggests that the generated ions of the descendant nuclides reach the glass fiber filter paper before being adsorbed on the film on the inside wall of the cylinder due to the high exhaust rate and are filtered, and that many of the Rn atoms pass through the cylinder inner space before their disintegration and are trapped in the charcoal impregnated filter paper or the charcoal cartridge at an exhaust rate of 30 L/min. At 4 and 15 L/min, the $^{211}$Pb distribution was intermediate between those at 1 and 30 L/min.

3-3. Dependence of distribution of $^{211}$Pb by $^{219}$Rn dispersion on the exposed surface area of the solution

As shown in the previous section, the dispersion of $^{219}$Rn can be tracked by the amount of detected $^{211}$Pb in the collection apparatus. The dependence of the dispersion of $^{219}$Rn on the surface area of the $^{223}$Ra solution was investigated based on the detection of the radioactivity of $^{211}$Pb with the microtubes and 10, 20, 50, and 100-mL beakers with 12 mL of the aqueous solution containing 6.0–8.7 MBq of $^{223}$Ra. In the experiments using the microtubes and 10-mL beaker, the solution was equally divided and was added to 7 microtubes or two 10-mL beakers to keep the volume of the $^{223}$Ra solution constant. The exhaust rate was set at 1 L/min in all the experiments. Under all of the studied conditions, the most $^{211}$Pb is adsorbed on the film on the inner wall of the cylinder and the paraffin film on the

| Used vessel    | Inner diameter of the vessels [mm] | The number of vessels | Total exposed surface area of the solution [mm$^2$/10$^3$] | Radioactivity of $^{223}$Ra used [MBq] | Radioactivity of $^{211}$Pb detected in the collection apparatus [kBq] | Rate coefficient of the dispersed $^{219}$Rn atoms, $k$ [10$^4$ s$^{-1}$] | Dispersal rate of $^{219}$Rn [10$^{-3}$] |
|---------------|-----------------------------------|----------------------|-------------------------------------------------|----------------------------------------|-----------------------------------------------------------------|--------------------------------------|-----------------|
| 100 mL beaker | 51                                | 1                    | 2.0                                             | 8.2 ± 0.1                             | (1.1 ± 0.1) × 10$^2$                                              | 16 ± 1                               | 20 ± 1         |
| 50 mL beaker  | 39                                | 1                    | 1.2                                             | 8.1 ± 0.1                             | 36 ± 15                                                          | 5.2 ± 1.7                             | 6.4 ± 2.1      |
| 20 mL beaker  | 31                                | 1                    | 0.75                                            | 6.2 ± 0.1                             | 17 ± 2                                                           | 2.5 ± 0.2                             | 4.0 ± 0.4      |
| 10 mL beakers | 22                                | 2                    | 0.76                                            | 6.1 ± 0.1                             | 10 ± 1                                                           | 1.5 ± 0.1                             | 2.5 ± 0.2      |
| micro tubes   | 8                                 | 7                    | 0.35                                            | 6.0 ± 0.1                             | 6.8 ± 0.1                                                        | 1.0 ± 0.1                             | 1.6 ± 0.1      |
inner wall of the sample holder (a 100-mL beaker). Table 2 summarizes the total amount of radioactivity for \(^{211}\)Pb with each vessel used. The sum of the detected radioactivity for \(^{211}\)Pb in the experiment using the 100-mL beaker is \((1.1 \pm 0.1) \times 10^2\) kBq and that using the microtubes is \(6.8 \pm 0.1\) kBq. The detected radioactivity of \(^{211}\)Pb decreases as the inner diameter of the vessel containing the \(^{223}\)Ra solution decreases. This is probably because the exposed surface area of the solution to the air decreases with the decreasing inner diameter of the vessel.

3-4. Dispersal rate of \(^{219}\)Rn derived from the ratio of the dispersed radioactivity detected as \(^{211}\)Pb to the radioactivity of \(^{219}\)Rn in the solution

It is presumed that a small fraction of \(^{219}\)Rn continually disperses from the solution to the air. All of the \(^{211}\)Pb nuclides that are produced by the nuclear decay of the dispersed \(^{219}\)Rn are adsorbed on or filtered by the parts of the collection apparatus (the PET film on the inside wall of the cylinder, filters, cartridges, O-rings, and paraffin films) at the exhaust rate of 1 L/min as described above. Thus, the dispersal rate of \(^{219}\)Rn from the solution to the air at an exhaust rate of 1 L/min could be derived. In the present study, the dispersal rate of \(^{219}\)Rn is defined based on the ratio of the number of dispersed \(^{219}\)Rn atoms, which is calculated from the total radioactivity of \(^{211}\)Pb detected using the collection apparatus, to the number of \(^{219}\)Rn atoms in the solution in 1 h. The rate of the number of \(^{219}\)Rn atoms dispersed from the solution to the air is

\[
\frac{dN_{Rn(Ra)}}{dt} = k = \lambda_{Rn} N_{D(Rn)},
\]

where \(N_{D(Ra)}\) is the number of dispersed \(^{219}\)Rn atoms, \(k\) is the rate coefficient of the dispersed \(^{219}\)Rn atoms from the solution \([s^{-1}]\), and \(\lambda_{Rn}\) is the decay constant of \(^{219}\)Rn \([s^{-1}]\). Integration of Equation 4 gives

\[
N_{D(Rn)} = \frac{k}{\lambda_{Rn}} (1 - e^{-\lambda_{Rn} t}).
\]

As the branching ratio of the disintegration from \(^{215}\)Po to \(^{211}\)Pb is \(> 0.99\), as shown in Figure 1, the ratio approximates to 1. The rate of change of the number of \(^{215}\)Po atoms can be written as follows,

\[
\frac{dN_{Po}}{dt} = \lambda_{Rn} N_{D(Rn)} - \lambda_{Po} N_{Po} = k(1 - e^{-\lambda_{Rn} t}) - \lambda_{Po} N_{Po},
\]

where \(N_{Po}\) is the number of \(^{215}\)Po atoms and \(\lambda_{Po}\) is the decay constant of \(^{215}\)Po \([s^{-1}]\). Integration of Equation 6 yields

\[
N_{Po} = k \left\{ \frac{1}{\lambda_{Po}} (1 - e^{-\lambda_{Po} t}) - \frac{1}{\lambda_{Rn} - \lambda_{Po}} (e^{-\lambda_{Rn} t} - e^{-\lambda_{Po} t}) \right\}.
\]

In the stepwise disintegrations, the approximation \(\lambda_{Po} \approx \lambda_{Rn}\) \(\lambda_{Rn}\) can be employed. Equation 7 is then simplified to

\[
N_{Po} = k \left\{ \frac{1}{\lambda_{Po}} (1 - e^{-\lambda_{Po} t}) \right\}.
\]

The rate of change of the number of \(^{211}\)Pb atoms collected on the parts of the apparatus can be written as follows,

\[
\frac{dN_{pb}}{dt} = \lambda_{Po} N_{Po} - \lambda_{pb} N_{pb} = k(1 - e^{-\lambda_{Rn} t}) - \lambda_{Po} N_{Po}.
\]

where \(N_{pb}\) is the number of \(^{211}\)Pb atoms collected on the parts of the apparatus and \(\lambda_{pb}\) is the decay constant of \(^{211}\)Pb \([s^{-1}]\). Integration of Equation 9 gives

\[
N_{pb} = k \left\{ \frac{1}{\lambda_{pb}} (1 - e^{-\lambda_{pb} t}) - \frac{1}{\lambda_{Rn} - \lambda_{Po}} (e^{-\lambda_{Rn} t} - e^{-\lambda_{Po} t}) \right\}.
\]

The \(k\) value is calculated from Equation 10 using the value of \(N_{pb}\) value which is derived from the radioactivity of \(^{211}\)Pb measured at the end of the collection. The calculated \(k\) values listed in Table 2 are in the range of \((1.0 \pm 0.1) \times 10^4 - (16 \pm 1) \times 10^4\) s\(^{-1}\).

As the half-life of \(^{219}\)Rn is short, the secular radioactive equilibrium between \(^{223}\)Ra and \(^{219}\)Rn has established in the solution during the collection of the dispersed \(^{219}\)Rn atoms, the number of \(^{219}\)Rn atoms in the solution \((N_{S(Rn)})\) is given by the following relationship:

\[
N_{S(Rn)} = \frac{\lambda_{Po}}{\lambda_{Rn}} N_{S(Ra)}.
\]

where \(\lambda_{Ra}\) and \(N_{S(Ra)}\) represent the decay constant of \(^{223}\)Ra \([s^{-1}]\) and the number of \(^{223}\)Ra atoms in the solution. The dispersal rate of \(^{219}\)Rn \((R_{Rn})\) for 1 h is derived as follows:

\[
R_{Rn} = \int_0^{600} \frac{N_{D(Rn)}dt}{N_{S(Ra)}} = k \int_0^{600} \frac{N_{D(Rn)}dt}{N_{S(Ra)}} = \frac{k}{\lambda_{Rn} N_{S(Ra)}} \int_0^{600} \frac{dt}{\lambda_{Rn} N_{S(Ra)}} = \frac{1}{\lambda_{Rn} N_{S(Ra)}} \int_0^{600} \frac{dt}{\lambda_{Rn} N_{S(Ra)}}.
\]
Table 2 also shows the $R_{Ru}$ values of the various vessels used. The values of the dispersal rate for $^{219}$Rn are in the range of $(1.6 \pm 0.1) \times 10^{-3} - (2.0 \pm 0.1) \times 10^{-2}$, which decrease significantly with the decreasing inner diameter of the vessel used. The dispersion of $^{222}$Rn is known to depend on the exposed surface area of the dispersal source.\textsuperscript{15} Thus, the decrease in the dispersal rate should be primarily due to the decrease in the exposed surface area of the solution. The reported solubility for a neutral single-atom molecule of Rn is $0.2268 \text{ mL per 1 mL of water under standard conditions.}$\textsuperscript{16} The calculated cubic volume of $^{219}$Rn in radioactive equilibrium with $^{222}$Ra in the solution under the conditions of the present study is $1.4 \times 10^{-12} - 2.0 \times 10^{-12} \text{ mL per 12 mL of the aqueous solution; namely, the ratio is } 1.2 \times 10^{-13} - 1.7 \times 10^{-15} \text{ mL/mL.}$ This value is quite small compared the solubility value of Rn. The large solubility of Rn in water would result in a small decrease in the dispersal rate of $^{223}$Ra. The values of the dispersal rate for $^{219}$Rn are in the range of $1.2 \times 10^{-13} - 1.7 \times 10^{-15} \text{ mL/mL.}$ The values of the dispersal rate for $^{219}$Rn from the aqueous $^{223}$Ra solution containing sodium chloride/citrate is $1.4 \times 10^{-3} - (2.0 \pm 0.1) \times 10^{-2}$. These rates were found to decrease with decreasing exposed surface area of the vessel used.

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