Radon diffusion in an anhydrous andesitic melt: a finite difference solution

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1. Introduction

Radon (Rn) is the heaviest element of the noble gas group. Unlike other gases of this chemical family, it has no stable isotopes and the three found in nature, 222Rn, 220Rn and 219Rn, are short-lived. These isotopes belong to the natural decay chains of 238U, 232Th and 235U, respectively. The longest-lived among them is 222Rn with half-life of 3.825 days (Gill et al., 1985). In other lava types, 222Rn loss was shown to be important when arbitrary initial and boundary conditions are required. We have shown that the explicit finite difference method is effective and accurate for solving equations that describe 222Rn diffusion in andesitic melts, which is especially important when arbitrary initial and boundary conditions are required.
where \( a \) is the half-width of the slab. In order to take into account the radioactivity that is not negligible in view of the duration of heating in the experiments, equation (1) has to be modified by adding a term for the production of 222Rn from its parent 226Ra, and a decay term, which leads to:

\[
\frac{\partial C_{Rn}}{\partial t} = \lambda_{Rn} C_{Rn} - \lambda_{Ra} C_{Ra} + D \frac{\partial^2 C_{Rn}}{\partial z^2} + \frac{4C_{Rn}^{eq}}{\pi} \sum_{n=0}^{\infty} \left[ \frac{(-1)^n \exp \left( -\frac{(2n+1)^2 \pi^2 z}{4a^2} + \lambda_{Rn} t \right)}{(2n+1) \left[ 1 + \frac{4\lambda_{Rn} a^2}{(2n+1)^2 \pi^2 D} \right]} \right] \times \cos \left( \frac{(2n+1) \pi z}{2a} \right)
\]

(3)

where \( C_{Rn} \) and \( C_{Ra} \) represent concentrations (in atoms \( \times g^{-1} \)) of 222Rn and 226Ra, respectively; \( \lambda_{Rn} \) and \( \lambda_{Ra} \) are the decay constants of 226Ra and 222Rn, respectively. For initial and boundary conditions \( C_{Rn} = C_{Rn}^{eq} = C_{Ra} (\lambda_{Rn}/\lambda_{Ra}) \) at \( t = 0, -a < z < a \) and \( C_{Ra} = 0 \) at \( t > 0, z = \pm a \), the solution of (3) is given by:

\[
C_{Rn}(z, t) = C_{Rn}^{eq} - \frac{4C_{Rn}^{eq}}{\pi} \sum_{n=0}^{\infty} \left[ \frac{(-1)^n \exp \left( -\frac{(2n+1)^2 \pi^2 z}{4a^2} + \lambda_{Rn} t \right)}{(2n+1) \left[ 1 + \frac{4\lambda_{Rn} a^2}{(2n+1)^2 \pi^2 D} \right]} \right] \times \cos \left( \frac{(2n+1) \pi z}{2a} \right)
\]

(4)

Multiplying both sides of equation (4) by \( \lambda_{Rn} \), one obtains the activity of 222Rn:

\[
(\text{Rn})(z, t) = (\text{Ra}) - (\text{Ra}) \frac{\cosh \left( \frac{z \sqrt{\lambda_{Rn}/D}}{D} \right)}{\cosh \left( \frac{a \sqrt{\lambda_{Rn}/D}}{D} \right)} \times \frac{4C_{Rn}^{eq}}{\pi} \sum_{n=0}^{\infty} \left[ \frac{(-1)^n \exp \left( -\frac{(2n+1)^2 \pi^2 z}{4a^2} + \lambda_{Rn} t \right)}{(2n+1) \left[ 1 + \frac{4\lambda_{Rn} a^2}{(2n+1)^2 \pi^2 D} \right]} \right] \times \cos \left( \frac{(2n+1) \pi z}{2a} \right)
\]

(5)

where \( (\text{Ra}) \) and \( (\text{Rn}) \) are activities of 226Ra and 222Rn, respectively. The initial and boundary conditions become \((\text{Rn}) = (\text{Rn})_{Ba} = (\text{Ra}) \) at \( t = 0, -a < z < a \) and \((\text{Ra}) = 0 \) at \( t > 0, z = \pm a \). Knowing the function \((\text{Rn})(z, t)\) allows calculation of the 222Rn concentration at each point of the slab for a given diffusion coefficient and at a given time. Steady state profiles are governed by the following equation:

\[
(\text{Rn})(z, \infty) = (\text{Ra}) - (\text{Ra}) \frac{\cosh \left( \frac{z \sqrt{\lambda_{Rn}/D}}{D} \right)}{\cosh \left( \frac{a \sqrt{\lambda_{Rn}/D}}{D} \right)}
\]

(6)

which is strongly dependent on \( \zeta = a \sqrt{\lambda_{Rn}/D} \).

3. Numerical method

Because analytical solutions (2) and (4) of respective equations (1) and (3) are infinite series, employing numerical methods offers flexibility especially for arbitrary initial distribution and boundary conditions. In this work, the explicit finite difference method (EFDM) is used to solve the diffusion equations (1) and (3). For both

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**Fig. 1.** Schematic representation of the sample geometry (Gauthier et al., 1999).
cases, the central difference scheme was used to represent the term \(\frac{\partial^2 C}{\partial z^2}\) and \(\frac{\partial C}{\partial t}\), and a forward difference scheme for the derivative term \(\frac{\partial C}{\partial z}\) and \(\frac{\partial C}{\partial t}\) (Anderson, 1995). With these substitutions, equation (1) transforms into:

\[
C_{i,j}^{n+1} = \frac{D\Delta t}{\Delta z^2}C_{i,j}^{n-1} + \left(1 - \frac{2D\Delta t}{\Delta z^2}\right)C_{i,j}^{n} + \frac{D\Delta t}{\Delta z^2}C_{i,j}^{n+1}
\]

and equation (3) transforms into:

\[
C_{i,Rn}^{n+1} = \frac{D\Delta t}{\Delta z^2}C_{i,Rn}^{n} + \left(1 - \lambda_{Rn}\Delta t - \frac{2D\Delta t}{\Delta z^2}\right)C_{i,Rn}^{n} + \frac{D\Delta t}{\Delta z^2}C_{i,Rn}^{n+1} + \lambda_{Rn}\Delta t C_{i,Rn}^{n+1}
\]

where indexes \(i\) and \(j\) refer to the discrete step lengths \(\Delta z\) and \(\Delta t\) for the coordinate \(z\) and time \(t\), respectively. Equations (7) and (8) represent formulas for \(C_{i,j}^{n+1}\) and \(C_{i,Rn}^{n+1}\), respectively, at the \((i,j+1)\)th mesh point in terms of known values along the \(j\)th time row. The truncation error for the difference equations (7) and (8) is \(O(\Delta t, \Delta z^2)\). Using a small-enough value of \(\Delta t\) and \(\Delta z\), the truncation error can be reduced until the accuracy achieved is within the error tolerance (Anderson, 1995).

In the difference form, the initial condition for equation (1) can be expressed as:

\[
C_{i}^{0} = C_{0} \quad \text{for} \quad t = 0; \quad -a < z < a
\]

The boundary conditions are:

\[
C_{N}^{j} = 0 \quad \text{for} \quad z = \pm a; \quad t > 0
\]

where \(N = a/\Delta z\) is the total grid dimension of the slab in the \(z\) direction. The initial condition for equation (3) can be expressed as:

\[
C_{i,Rn}^{n} = C_{Rn}^{eq,0} = \frac{\lambda_{Rn}}{\lambda_{Rn}}C_{i,Rn}^{0} \quad \text{for} \quad t = 0; \quad -a < z < a
\]

and the boundary conditions are:

\[
C_{i}^{n} = 0 \quad \text{for} \quad z = \pm a; \quad t > 0
\]

4. Analytical and numerical results

To facilitate the comparison of results, we applied our method of \(^{222}\text{Rn}\) diffusion calculation to the same cylindrical geometry used in experiments by Gauthier et al. (1999), Fig. 1. Shown in Fig. 2 are numerical results obtained by solving the diffusion equations (1) and (3) by EFDM. In the calculations, the step lengths \(\Delta z = 0.05\text{ cm}\) and \(\Delta t = 0.01\text{ s}\) have been used to achieve the stability of the finite difference scheme.

Gauthier et al. (1999) calculated from Henry’s law a gas concentration in andesitic melts at 1500 °C under atmospheric conditions close to \(1.5 \times 10^{-26}\) mol/g. The equilibrium \(^{222}\text{Rn}\) activity in their sample was equivalent to a concentration of about \(2.7 \times 10^{-18}\) mol/g, which is considerable higher (by ten orders of magnitude) than a gas concentration in andesitic melts at 1500 °C under atmospheric conditions: \(^{222}\text{Rn}\) was thus grossly supersaturated in their melt.

Fig. 2(a) shows diffusion profiles derived from equation (2) and our numerical solution of equation (1) for stable element, for \(0 < z < a (a = 10^{-2}\text{ m})\) with \(D = 10^{-10}\text{ m}^2\text{s}^{-1}\). The curves represent solution of the diffusion equation (1) obtained using EFDM (solid squares) and analytical solution (2) (solid lines) with \(a = 10^{-2}\text{ m}\) and \(D = 10^{-10}\text{ m}^2\text{s}^{-1}\) for different diffusion times \(t\) (in seconds). 2(b): Curves represent solution of the diffusion equation (3) obtained using EFDM (solid squares) and analytical solution (5) (solid lines) with \(a = 10^{-2}\text{ m}\) and \(D = 10^{-10}\text{ m}^2\text{s}^{-1}\) for different diffusion times \(t\) (in seconds). 2(c): Steady state diffusion profiles obtained using EFDM (solid squares) for \(t = 10^6\text{ s}\) and analytical steady state solution (6) (solid line). Numbers on diffusion curves refer to values of \((1 - \alpha/\lambda_{Rn}/D)\) (please refer to the text).

(Fig. 2(a)) when \(t\) tends to infinity, the concentration pattern becomes zero in the entire slab. Fig. 2(b) shows that with increasing diffusion time the effect of \(^{222}\text{Rn}\) radioactive ingrowth becomes more dominant, counterbalancing the effect of diffusion. In fact, a steady state is reached, for which diffusion is exactly balanced by \(^{222}\text{Rn}\) production from \(^{226}\text{Ra}\).

In Fig. 2(c), steady state diffusion profiles obtained analytically from equation (6) \((t \rightarrow \infty)\) are compared with the numerical solution of equation (3) for \(t = 10^6\text{ s}\). For small (\(\zeta < 0.1\)), the \(^{222}\text{Rn}\) radioactive ingrowth is negligible and \(^{222}\text{Rn}\) behaves like a stable element. For large \((\zeta > 50)\), diffusion processes are too slow and balanced by \(^{226}\text{Ra}\) decay, leading to very small, if any, \(^{222}\text{Rn}\) depletion in the slab.

5. Conclusion

Numerical solutions are reported for the diffusion equations employed to investigate the process of \(^{222}\text{Rn}\) transport in a slab of anhydrous andesitic melt. The solutions of the relevant diffusion equations were by the explicit finite difference method. Results were compared to analytical solutions available in the literature.
and good agreement was found. For a stable element when $t$ tends to infinity, the concentration pattern becomes zero in the entire slab. If the element is not stable, the effect of $^{222}$Rn radioactive ingrowth becomes more dominant with the lapse of diffusion time, counterbalancing the effect of diffusion. In fact, a steady state is reached for which diffusion is exactly balanced by $^{222}$Rn production from $^{226}$Ra. On the other hand, for small $\zeta(<0.1)$, the $^{222}$Rn radioactive ingrowth is negligible and $^{222}$Rn behaves like a stable element. For large $\zeta(>50)$, diffusion processes are too slow and balanced by $^{226}$Ra decay, leading to very small, if any, $^{222}$Rn depletion in the slab.

Finally, we have shown that explicit finite difference method is effective and accurate for solving diffusion equations that describe $^{222}$Rn diffusion in andesitic melts, which is especially important when arbitrary initial and boundary conditions are required.

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References


