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In predicting hydrogeologic transport of a radionuclide chain flow one usually assumes a transport through a porous medium. However, some rock media surrounding buried wastes are better characterized as inhomogenous fractured media. Groundwater flows mainly through macro fissures but is almost stationary in micropores intersecting these fissures. Molecular diffusion into and out of the micropores can affect the migration of nuclides through the fissures[1]. Here we present the general analytical solution to fissure-flow transport of a radionuclide chain and demonstrate the chromatographic behavior of radionuclides in a three-member decay chain.

Consider a fissure with planar surfaces separated by a width b, through which water flows in the z direction at velocity v. The surrounding rock, with a porosity ε, contains micropores which connect to the fissure surfaces. Dissolved radionuclides do not absorb on the fissure walls, but they can diffuse into and out of the micropores and adsorb on the micropore

surfaces. The governing equations for the transport of radionuclide $\underline{i}$ in the fissure and micropores are:

$$\frac{\partial N_i}{\partial t} + v \frac{\partial N_i}{\partial z} + \lambda_i N_i = -\frac{2}{b} J_i + \lambda_{i-1} N_{i-1} \quad , \; i = 1,2,.... \tag{1}$$

$$\frac{\partial M_i}{\partial t} - \frac{D_i}{K_i} \frac{\partial^2 M_i}{\partial y^2} + \lambda_i M_i = \lambda_{i-1} M_{i-1} \quad , \qquad i = 1,2,.... \tag{2}$$

where $N_i(z,t)$ and $M_i(z,y,t)$ are the aqueous concentrations of nuclide $\underline{i}$ in the fissure and micropores, respectively, D_i is the molecular diffusivity, K_i is the sorption retardation constant, and λ_i is the decay constant. $J_i(z,t)$ is the y diffusive flux into micropores at the surface of the fissure (y=0), given by:

$$J_k(z,t) = -\varepsilon D_i \left. \frac{\partial M_i}{\partial y} \right|_{y=0}, \; z>0, \; t>o, \qquad i = 1,2,.... \tag{3}$$

The initial and boundary conditions are:

$$N_i(z,0) = 0, \qquad z>0, \qquad i = 1,2,3,... \tag{4}$$

$$M_i(z,y,0) = 0, \quad z>0, \; y>0, \qquad i = 1,2,3,... \tag{5}$$

$$N_i(0,t) = N_i^o(t), \; t>0, \qquad i = 1,2,3,... \tag{6}$$

$$M_i(z,0,t) = N_i(z,t), \; t>0, \; y>0, \qquad i = 1,2,3,... \tag{7}$$

where $N_i^o(t)$ is the general time-dependent concentration of $\underline{i}$ at the waste repository.

An exact solution for the first member of a decay chain and an exact

recursive solution for a decay chain of arbitrary length have been obtained[2]. To obtain explicit solutions for the decay chain at early times, we neglect radioactive decay in the micropores, resulting in:

$$N_i(z,t) = e^{-\frac{\lambda i}{v} z} \int_0^{t-\frac{z}{v}} N_i^o\left(t-\tau-\frac{z}{v}\right) P_i(\tau;a_i z)d\tau$$

$$+ \sum_{j=1}^{i-1} \frac{\prod_{\ell=j}^{i-1}(\lambda\ell)}{v^{i-j}} \sum_{\ell=j}^{i} \sum_{\substack{r=j \\ r\neq\ell}}^{i} \frac{A_\ell^{ij}}{\prod_{\substack{\ell=j \\ r\neq\ell}}^{i}(u_{j\ell})} e^{-\frac{\lambda_\ell}{v} z} \int_0^{t-\frac{z}{v}} N_i^o\left(t-\tau-\frac{z}{v}\right) G_{r\ell}(z,t)d\tau \tag{8}$$

$$M_i(z,y,t) = \int_0^t N_i(z,t-\tau)\, P_i(\tau;b_i y)d\tau \tag{9}$$

where

$$P_i(t;a) = \frac{\alpha}{\sqrt{\pi}} \frac{e^{-\frac{\alpha^2}{4t}}}{\sqrt{t^3}} \tag{10}$$

$$G_r(z,t) = \frac{e^{-a_\ell^2 z^2/4t}}{\sqrt{\pi t}} - \delta_{r\ell}\, e^{a_\ell \delta_{r\ell} z + \delta_{r\ell}^2 t}\, \mathrm{erfc}\left(\frac{z}{2\sqrt{t}} + \delta_{r\ell}\sqrt{t}\right) \tag{11}$$

$$\left.\begin{aligned} A_\ell^{ij} &= \prod_{\substack{q=j \\ r\neq\ell \\ q\neq\ell}}^{i} \left(\frac{1}{\delta_{q\ell}-\delta_{r\ell}}\right), \quad \delta_{r\ell} = \frac{\lambda_r-\lambda_\ell}{v(a_r-a_\ell)} \\ a_i &= \frac{z\varepsilon D_i}{bv}\sqrt{\frac{K_i}{D_i}}, \quad b_i = \sqrt{\frac{K_i}{D_i}}, \quad \mu_{r\ell} = a_r-a_\ell \end{aligned}\right\} \tag{12}$$

The space-time-dependent concentration of the i-th species in the fissure

for a step release can be obtained by substituting the time-dependent source concentration $N_i^o(t)$, given by the Bateman equation[4], into the above solution. The concentration for a band release can be obtained from the superposition theorem[3].

The concentration profiles for the radionuclide chain $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra \rightarrow$ are shown as the solid lines in Figure 1. Also shown are the profiles calculated for porous-flow transport. Because of the assumed lack of sorption on the macropore surfaces, the front of the concentration profile in fracture flow moves at the water velocity. Time delays for diffusion into and out of sorbing micropores spreads the concentration profile for fracture flow and considerably attenuates the peak concentrations below those predicted for flow through a porous medium with local sorptive equilibrium. For the parameters assumed here, the peak concentration of ^{226}Ra is a hundred-fold less for fracture flow. An even greater reduction will occur if radioactive decay while in the micropores and sorption on the fissure walls are taken into account. The spreading of the concentration profile due to micropore diffusion is qualitatively similar to, but considerably greater than, spreading by dispersion.

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References

1. I. Neretnieks, "Duffusion in the Rock Matrix, An Important Factor in Radionuclide Transport", J. Geophys. Res., 85, 4379 (1980).

2. T. Kanki, A. Fujita, P.L. Chambré, and T. H. Pigford, "Transport of Radionuclides in Fractured Media", UCB-NE-4009, (1981).

3. M. Foglia, T. H. Pigford, and P. L. Chambré, "The Superposition Solution of the Transport of a Radionuclide Chain Through a Sorbing Medium", Trans. Am. Nucl. Soc., 33, 384 (1979).

4. M. Harada, P. L. Chambré, M. Foglia, K. Higashi, F. Iwamoto, D. Leung, T. H. Pigford and D. Ting; "Migration of Radionuclides Through Sorbing Media, Analytical Solutions I", LBL-10500 (1980).

Figure Caption:

Figure 1. Concentration profiles for the chain $^{234}U \rightarrow {}^{230}Th \rightarrow {}^{226}Ra \rightarrow$ in fissure flow and in porous flow (t = 50,000 yr, no daughter nuclides initially present, leach time = 30,000 yr).

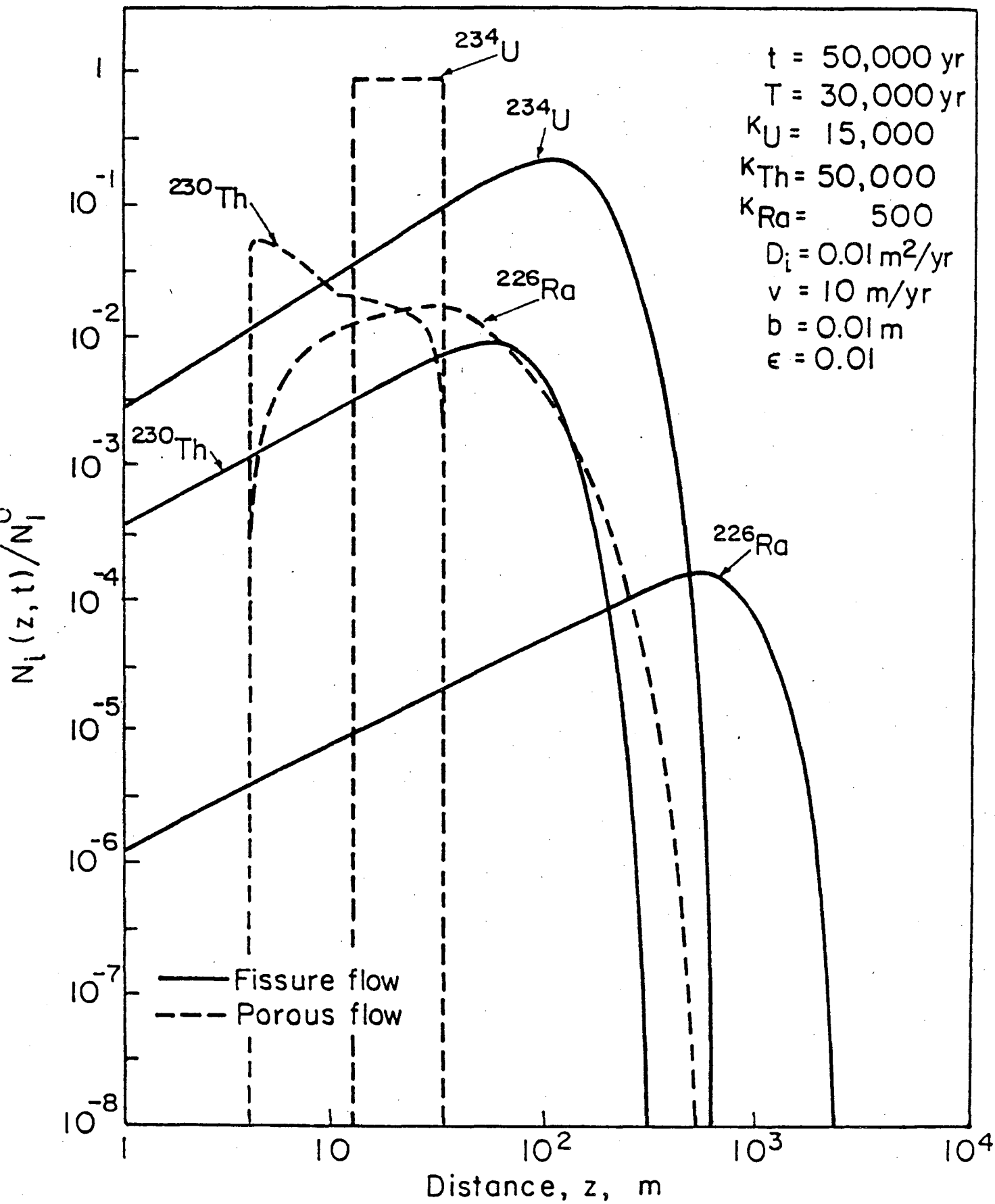
t = 50,000 yr
T = 30,000 yr
K_U = 15,000
K_{Th} = 50,000
K_{Ra} = 500
D_i = 0.01 m²/yr
v = 10 m/yr
b = 0.01 m
ε = 0.01
234U
234U
230Th
226Ra
230Th
226Ra
Fissure flow
Porous flow
$N_i(z,t)/N_i^0$
Distance, z, m
1
10
10^2
10^3
10^4
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
10^{-7}
10^{-8}

Figure 1.

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