



Research note

Analytical solution to the diffusion, sorption and decay chain equation in a saturated porous medium between two reservoirs



Juan Guzman ^{a,*}, Serguei Maximov ^{b,c}, Rafael Escarela-Perez ^a, Irvin López-García ^a, Mario Moranchel ^d

^a Departamento de Energía, Universidad Autónoma Metropolitana-Azcapotzalco, Av. San Pablo 180 Col. Reynosa Tamaulipas, México, D.F. 02200, Mexico

^b Instituto Tecnológico de Morelia, Av. Tecnológico #1500, Lomas de Santiaguito, Morelia, Michoacán 58120, Mexico

^c On sabbatical leave, Universidad Autónoma Metropolitana, Azcapotzalco, México, D.F. 02200, Mexico

^d Escuela Superior de Física y Matemáticas, Instituto Politécnico Nacional, Av. Instituto Politécnico Nacional s/n, U. P. Adolfo López Mateos, Col. San Pedro Zacatenco, México, D.F. 07738, Mexico

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ABSTRACT

The diffusion and distribution coefficients are important parameters in the design of barrier systems used in radioactive repositories. These coefficients can be determined using a two-reservoir configuration, where a saturated porous medium is allocated between two reservoirs filled by stagnant water. One of the reservoirs contains a high concentration of radioisotopes. The goal of this work is to obtain an analytical solution for the concentration of all radioisotopes in the decay chain of a two-reservoir configuration. The analytical solution must be obtained by taking into account the diffusion and sorption processes. Concepts such as overvalued concentration, diffusion and decay factors are employed to this end. It is analytically proven that a factor of the solution is identical for all chains (considering a time scaling factor), if certain parameters do not change. In addition, it is proven that the concentration sensitivity, due to the distribution coefficient variation, depends of the porous medium thickness, which is practically insensitive for small porous medium thicknesses. The analytical solution for the radioisotope concentration is compared with experimental and numerical results available in literature.

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1. Introduction

The determination of diffusion and distribution coefficients is important in the design of barrier systems that are used in radioactive repositories. Among the available experimental setups, the two-reservoir configuration is commonly used to determine diffusion and distribution coefficients. This configuration consists of a saturated porous medium surrounded by two reservoirs: one of the reservoirs, called injective reservoir (IR), contains a high concentration of radioisotopes and the other, called diffusive reservoir (DR), is initially free of radioisotopes.

The goal of this work is to present an analytical solution for the radioisotope concentration in the two reservoir configuration. The analytical solution must be obtained by taking into account the

diffusion and sorption processes. It is demonstrated that the solution has a factor that is invariant in time.

There are at least three ways of analyzing a two-reservoir configuration: a) analytical, b) experimental and c) finite element methods (FEM). Chen et al. (2012) found radioisotope concentrations and the diffusion coefficient using multi-compartment methods. Moridis (1999) obtained analytical solutions for the diffusion, sorption and decay equations of radioisotope concentrations in the Laplace domain. Guzmán et al. (2014), employed the finite element method (FEM) for determining diffusion and distribution coefficients. Lü and Ahl (2005) and Lü and Viljanen (2002) derived an analytical expression for the diffusion coefficient in the steady state. Pérez Guerrero et al. (2009, 2010) found an analytical expression for the concentration using classical integral transform techniques. An approximated analytical solution for the concentration in the diffusive reservoir is obtained in the work of Crank (1975) using a separation of variables technique. Shackelford (1991) reviews classical methods aimed at finding diffusion coefficients.

Alongside the analytical and numerical methods, experimental methods are a powerful tool for determining diffusion and

Abbreviations: IR, Injective reservoir; DR, Diffusive reservoir; DC, Diffusion coefficient; DIC, Distribution coefficient.

* Corresponding author. Tel.: +52 55 53189047; fax: +52 55 53947378.

E-mail address: maestro_juan_rafael@hotmail.com (J. Guzman).

distribution coefficients. Yamaguchi and Nakayama (1998), experimentally found the diffusion coefficient (DC) and the distribution coefficient (DIC) for uranium-233 and plutonium-239 of Inada granite. Yamaguchi et al. (1997) determined DC and DIC for the uranyl ion and Yamaguchi et al. (1993) for strontium in Inada granite. Aldaba et al. (2010) determined DC and DIC of cesium and strontium in unsaturated soils from Spain. García-Gutiérrez et al. (2001) established DC and DIC of strontium and Eriksen et al. (1999) obtained DC and DIC of cations Na^+ , Cs^+ , Co^{2+} , and Sr^{2+} in compacted bentonite. DC and DIC of tritiated water in cement pastes were obtained in the work of Tits et al. (2003).

An important concept that simplifies the analysis of the decay chain is the overvalued concentration (Guzmán et al., 2014). An overvalued concentration is an estimation of the maximum of the actual radioisotope concentration, which is obtained by replacing the molecular diffusion and distribution coefficients by the maximum value of the molecular diffusion coefficient and the minimum distribution coefficient of all the isotopes in the decay chain, respectively. Since the overvalued radioisotope concentration is greater than its actual concentration, then, if the estimated overvalued concentration does not harm human health, then the real concentration does not do it either. In Guzmán et al. (2014), it is shown that such overvalued concentration for any isotope of the decay chain and the actual concentration for the first isotope can be factorized in two terms: a decay factor that describes decay without diffusion, and a diffusion factor, which describes diffusion without decay. Equation for the decay factor is solved analytically whereas the diffusion factor is obtained numerically by means of FEM.

In present work, the diffusion factor is determined analytically by using a separation of variables technique (Fourier method). For validation purposes, the obtained analytical solution is compared with the numerical solution obtained by Guzmán et al. (2014) and with the experimental data from Yamaguchi et al. (1998). It is shown that the diffusion factor is invariant in time, that is, the factor is the same for all the decay chain (considering a time scaling factor), provided that the same dimensions, porosities and distribution factors are employed. Moreover, it is demonstrated analytically that the diffusion factor is practically insensitive to the distribution coefficient variation if the porous medium thickness is small enough.

The advantages and applications of the analytical solution are: 1) they provide rigorous and sound solutions that become very useful for practical design and data analysis problems. Indeed, analytical methods provide explicit functions of parameters, leading to easy application of optimization tools and data analysis. Moreover, analytical functions may significantly reduce computational costs. For instance, finite-element approaches require tailor-made software and heavy computer resources (memory and cpu), whereas an analytical solution can even be treated and analyzed without a computer in many cases. 2) The demonstration of the time invariance of the diffusion factor allows the easy determination of the concentrations and distribution and diffusion coefficients using a time rescaling. 3) It has also been proven that the diffusion factor is almost insensitive to the distribution coefficient variation in situations where the porous medium thickness is small, whereas for a wide thickness, the effect is opposite. This means that the experimental determination of the diffusion coefficient can be easily obtained using thin porous media.

2. Model

In order to simplify the analysis, the half life of the radioisotope is considered large enough so that the isotope decay can be neglected. The isotope decay is considered in Section 5.

Consider a cylinder \mathbf{S} with a cross section of an arbitrary form and an area A and a height l , as illustrated in Fig. 1. The cylinder is filled with an absorbing porous material and an incompressible fluid (water), which is stagnated in a homogeneous saturated porous medium. The dissolved-in-water radioisotope can be absorbed by the porous medium. Two reservoirs \mathbf{S}_{IR} and \mathbf{S}_{DR} of arbitrary geometry and volumes V_{IR} and V_{DR} respectively, are connected by a cylinder \mathbf{S} . The reservoirs \mathbf{S}_{IR} and \mathbf{S}_{DR} are filled with water and the dissolved radioisotope. Mixing mechanisms are mounted inside the reservoirs, such that the isotope concentrations $c_{\text{IR}}(t)$ and $c_{\text{DR}}(t)$ are maintained constant throughout the entire volumes of the reservoirs \mathbf{S}_{IR} and \mathbf{S}_{DR} . The isotope penetrates from one cavity to the other one through the absorbing porous material inside cylinder \mathbf{S} . The dynamics of penetration is described by the diffusion and absorption equation (Yamaguchi et al., 1993):

$$\alpha \frac{\partial c}{\partial t} = D_e \frac{\partial^2 c}{\partial x^2} \quad (1)$$

where $c(x, t)$ is the concentration of the radioisotope per unit volume of water at the position x and time t in the porous medium; D_e is the effective diffusion coefficient of the radioisotope; α is the capacity factor in region $\mathbf{S} = \{x : 0 \leq x \leq l\}$. The effective diffusion coefficient D_e is given by:

$$D_e = \theta T D_m \quad (2)$$

and, the capacity factor α is:

$$\alpha = \theta + K\rho \quad (3)$$

where, θ and ρ are the porosity and bulk density of the porous medium, respectively; D_m and K are the molecular diffusion and distribution coefficients of the radioisotope, respectively; T is the tortuosity factor.

The boundary conditions for Equation (1) consist of the flux continuity through the boundary separating region \mathbf{S}_{IR} from \mathbf{S} and region \mathbf{S}_{DR} from \mathbf{S} , i.e., at points $x = 0$ and $x = l$:

$$V_{\text{IR}} \frac{dc_{\text{IR}}}{dt} = A D_e \frac{\partial c}{\partial x} \Big|_{x=0}, \quad V_{\text{DR}} \frac{dc_{\text{DR}}}{dt} = -A D_e \frac{\partial c}{\partial x} \Big|_{x=l} \quad (4)$$

and the concentration continuity on the same surfaces (boundaries):

$$c_{\text{IR}}(t) = c|_{x=0}, \quad c_{\text{DR}}(t) = c|_{x=l} \quad (5)$$

Also the radioisotope concentration in the porous region \mathbf{S} should satisfy the following initial condition:

$$c(x, 0) = f(x) \quad (6)$$

where, the initial conditions for the radioisotope concentrations in regions \mathbf{S}_{IR} and \mathbf{S}_{DR} have the form:

$$c_{\text{IR}}|_{t=0} = f(0), \quad c_{\text{DR}}|_{t=0} = f(l) \quad (7)$$

due to the continuity conditions (5) and (6).

3. Analytical solution

3.1. Solution methodology

First, it can be noticed that concentrations $c_{\text{IR}}(t)$ and $c_{\text{DR}}(t)$ can be excluded from conditions (4) and (5) by substituting (5) into (4). As a result the following boundary conditions for the isotope concentration $c(x, t)$ in the porous medium can be obtained:

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