Journal of Environmental Radioactivity 135 (2014) 100–107

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Diffusion and decay chain of radioisotopes in stagnant water in saturated porous media

Juan Guzmán [a,*], Jose Alvarez-Ramirez [b], Rafael Escarela-Pérez [a], Raúl Alejandro Vargas [c]

[a] *Departamento de Energía, Universidad Autónoma Metropolitana-Azcapotzalco, Av. San Pablo 180 Col. Reynosa Tamaulipas, México, D.F. 02200, Mexico*
[b] *División de Ciencias Básicas e Ingeniería, Universidad Autónoma Metropolitana-Iztapalapa, Av. San Rafael Atlixco 186 Col. Vicentina, México, D.F. 09340, Mexico*
[c] *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*

A R T I C L E I N F O

Article history:
Received 29 November 2013
Received in revised form
21 March 2014
Accepted 21 March 2014
Available online 8 May 2014

Keywords:
Radioisotope
Dispersion
Porous
Medium
Diffusion
Decay

A B S T R A C T

The analysis of the diffusion of radioisotopes in stagnant water in saturated porous media is important to validate the performance of barrier systems used in radioactive repositories. In this work a methodology is developed to determine the radioisotope concentration in a two-reservoir configuration: a saturated porous medium with stagnant water is surrounded by two reservoirs. The concentrations are obtained for all the radioisotopes of the decay chain using the concept of overvalued concentration. A methodology, based on the variable separation method, is proposed for the solution of the transport equation. The novelty of the proposed methodology involves the factorization of the overvalued concentration in two factors: one that describes the diffusion without decay and another one that describes the decay without diffusion. It is possible with the proposed methodology to determine the required time to obtain equal injective and diffusive concentrations in reservoirs. In fact, this time is inversely proportional to the diffusion coefficient. In addition, the proposed methodology allows finding the required time to get a linear and constant space distribution of the concentration in porous mediums. This time is inversely proportional to the diffusion coefficient. In order to validate the proposed methodology, the distributions in the radioisotope concentrations are compared with other experimental and numerical works.

1. Introduction

The barrier systems used in radioactive repositories prevent the release of radioactive substances to the ground surface. In order to avoid this release, the radioactive substances are vitrified and overpacked for a certain period of time. However, this containment of substances is impossible for all time. In order to minimize the possible liberation of substances, a low permeability material is allocated between the overpack and the host rock so that water is stagnant in this material. This means that the release of substances is limited by diffusion rather than advection in this material (Malekifarsani and Skachek, 2009). Therefore, it is important to analyze the diffusion and decay of substances dissolved in stagnant water in the two-reservoir configuration. The configuration consists of a specimen (saturated porous medium) surrounded by two reservoirs: a reservoir consists of a high concentration of radioisotopes (called injective reservoir, IR) and other is free of radioisotopes (called diffusive reservoir, DR).

Several studies have examined the diffusion of substances in water, which is stagnant in saturated porous media. In the works of Chen et al. (2012), Aldaba et al. (2010), García-Gutiérrez et al. (2001) and Shackelford (1991), the two-reservoir configuration is analyzed to determine the diffusion coefficients. Eriksen et al. (1999), Lü and Ahl (2005), Lü and Viljanen (2002), Tits et al. (2003), Yamaguchi and Nakayama (1998) determine diffusion coefficients for situations where the concentration of radioisotopes in IR is constant. While, Bharat et al. (2009), Lake and Rowe (2004), and Moridis (1999) find diffusion coefficients for situations where the concentration in the IR is variable.

In this work, a methodology is developed to determine the radioisotope concentration in water, which is stagnant in saturated porous media for a two-reservoir configuration. The concentrations are obtained for all the radioisotopes of the decay chain using the concept of overvalued concentration. A methodology based, on the

Abbreviations: IR, injective reservoir; DR, diffusive reservoir; DSD, diffusion, sorption and decay equation; DD, diffusion and decay equation.

* Corresponding author. Tel.: +52 55 53189047; fax: +52 55 53947378.
E-mail address: doctor_guzman@hotmail.com (J. Guzmán).

http://dx.doi.org/10.1016/j.jenvrad.2014.03.015
0265-931X/

variable separation method, has been proposed for the solution of the radioisotope transport equation. This methodology proposes to factorize the overvalued concentration in two terms: a factor that describes the diffusion without decay and other factor that describes the decay without diffusion.

It is considered that the radioisotopes can be absorbed by the porous medium (sorption process) and that these radioisotopes can decay. Henceforth, the transport of radioisotopes is examined by solving the diffusion, sorption and decay equations (DSD equations). It is considered that the sorption process follows a linear relationship, so that DSD equations are reduced to equations only involving diffusion and decay (DD equations). In order to solve the DD equations, a separation of variables is employed. A decoupling of the DD equations is then achievable, leading to an equation that describes the diffusion and other equation that incorporates decay. The resulting equations of the decay are then handled through the Laplace Integral Transform Technique (Arfken and Weber, 2005), which provides analytical expressions. The diffusion equation can be analytically or numerically solved, depending on the complexity of the problem. In this work, a numerical solution is established using the finite element method and a commercial software package (COMSOL, 2008). In order to validate the proposed methodology, our solution of the radioisotope concentration is compared with the solution of Moridis (1999) and Chen et al. (2012). Moridis (1999)have modeled the radioisotope transport using the Laplace transform technique for the decay of the parent radioisotope. The decay of the descendents of the parent has not been modeled. However, the diffusion and decay of the radioisotope and its first descendent are modeled in the work of Chen. The effect of the decay of other descendents was not considered. In this work, the transport of all descendents of the radioisotope is fully considered.

2. Preliminaries

Water is modeled by an incompressible fluid, which is stagnated in a homogeneous saturated porous medium. Henceforth, the advective part in the transport of radioisotopes is not considered. The transport of radioisotopes dissolved in water is produced by diffusion. It is considered that the radioisotopes can be absorbed (sorption process) by the porous medium. Also, the radioisotopes can decay into other radioisotopes, as is illustrated in Fig. 1. In Fig. 1, the radioisotope labeled 1 is the first radioisotope in the decay chain and decays in the radioisotope labeled 2, which in turn decays in the radioisotope labeled 3, and so on. In Fig. 1, the decay chain is formed by N radioisotopes.

Using the mass balance (Bear and Cheng, 2010), the equations that describe the radioisotopes transport due to diffusion, sorption and decay in a porous medium are obtained:

$$\frac{\partial C_1(x,t)}{\partial t} = D_{m1}\frac{\partial^2 C_1(x,t)}{\partial^2 x} - \frac{\rho_b}{\Phi}\frac{\partial S_1}{\partial t} - \frac{\rho_b}{\Phi}\lambda_1 S_1 - \lambda_1 C_1(x,t), \qquad (1)$$

and

$$\frac{\partial C_i(x,t)}{\partial t} = D_{mi}\frac{\partial^2 C_i(x,t)}{\partial x^2} - \frac{\rho_b}{\Phi}\frac{\partial S_i}{\partial t} - \lambda_i C_i(x,t) - \frac{\rho_b}{\Phi}\lambda_i S_i + \lambda_{i-1}C_{i-1}(x,t) + \frac{\rho_b}{\Phi}\lambda_{i-1}S_{i-1} \quad i = 2,\ldots,N, \qquad (2)$$

where $C_i(x,t)$ is the aqueous concentration per unit volume of water of the radioisotope i in the decay chain formed by N radioisotopes, at the position x and time t; C_{i-1} is the aqueous concentration of father radioisotope of i per unit volume of the water; λ_i and λ_{i-1} are the decay constant of the radioisotope i and its parent radioisotope,

$$1 \underset{\lambda_1}{\longrightarrow} 2 \underset{\lambda_2}{\longrightarrow} 3 \underset{\lambda_3}{\longrightarrow} \cdots \underset{\lambda_{N-1}}{\longrightarrow} N \underset{\lambda_N}{\longrightarrow}$$

Fig. 1. Radioisotopes intervening in the decay chain.

respectively; D_{mi} is the molecular diffusion coefficient of the radioisotope i; S_i is the absorbed mass of the radioisotope i in the soil matrix per unit bulk dry mass of the porous medium; Φ is the porosity; ρ_b is the bulk dry density of the porous medium (bulk soil density).

Eqs. (1) and (2) can be recast into one equation given by:

$$\frac{\partial C_i(x,t)}{\partial t} = D_{mi}\frac{\partial^2 C_i(x,t)}{\partial x^2} - \frac{\rho_b}{\Phi}\frac{\partial S_i}{\partial t} - \lambda_i C_i(x,t) - \frac{\rho_b}{\Phi}\lambda_i S_i + \lambda_{i-1}C_{i-1}(x,t) + \frac{\rho_b}{\Phi}\lambda_{i-1}S_{i-1} \quad i = 1,\ldots,N, \qquad (3)$$

This equation is not consistent with Eq. (1), since it contains the fictitious radioisotope $C_0(x,t)$. As a result, constant λ_0 is defined as $\lambda_0 = 0$, so as to make them consistent.

The first term on the left-hand side of Eq. (3) describes the diffusion of water in pores. The second term describes the absorption processes of movable substances: sorption and precipitation. If a movable substance is precipitated in a porous medium, this substance becomes immovable since porosity is small enough. Therefore, the precipitation effect is described by means of absorption of movable substances. The remaining terms in Eq. (3) describes decay of the radioisotope and its parent, respectively.

It is considered that the sorption process follows a linear relationship. That is, $S_i = K_i C_i$, where K_i is the distribution coefficient of radioisotope i. Therefore, Eq. (3) is given by:

$$\frac{\partial C_i(x,t)}{\partial t} = D_i\frac{\partial^2 C_i(x,t)}{\partial x^2} - \lambda'_i C_i(x,t) + \lambda'_{i-1}C_{i-1}(x,t) \quad i = 1,\ldots,N, \qquad (4)$$

where $\lambda_1^{'} = \lambda_1, \lambda'_{i-1} = (R_{i-1}/R_i)\lambda_{i-1} \quad (i\neq 2), R_i = 1 + (\rho_b/\Phi)K_i$ is the retardation factor of radioisotope i, D_i is the apparent diffusion coefficient of radioisotope i and given by:

$$D_i = \frac{D_{mi}}{R_i}. \qquad (5)$$

In order to validate the proposed methodology, proven solutions, provided in the works of Moridis (1999) and Chen et al. (2012), have been employed to. In these works, the two-reservoir method is used, where the saturated porous medium is surrounded by: a reservoir containing a high radioisotope concentration (called injective reservoir) and other reservoir that is initially free of radioisotopes (called diffusive reservoir), as shown in Fig. 2. A one-dimensional geometry is considered, where variable x describes position. In Fig. 2, $C_i(x,t)$ represents the concentration of the radioisotope i in the porous medium at position x and time t; $C_{IR,i}(t)$

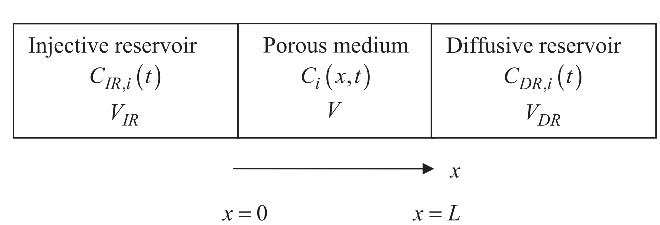

Fig. 2. Scheme of the two reservoir method.