

Identification of radon transfer velocity coefficient between liquid and gaseous phases

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Abstract

Radon transfer between a liquid phase and a gaseous phase is modelled by a Robin's condition (radon flux at the common interface is expressed as function of radon concentrations in the two phases). This condition involves two constants: Ostwald's coefficient (α) and the transfer velocity coefficient (β). Assuming the value of α is known, a method is proposed to determinate the value of β , by studying the radon transfer phenomenon at the laboratory scale. Knowing the initial radon concentrations, the experiment consists in measuring how long the radon flux passes through the common interface. In this stabilisation time radon transport is governed in each phase by diffusion and disintegration. Then, determination of β is equivalent to solving an inverse problem formulated using measured data. A numerical procedure is developed to solve this problem. *To cite this article: D.-G. Calugaru, J.-M. Crolet, C. R. Mecanique 330 (2002) 377–382.* © 2002 Académie des sciences/Éditions scientifiques et médicales Elsevier SAS

granular media / interface / Robin's condition / radon / Ostwald's coefficient / inverse problem

Identification de la vitesse de transfert de radon entre une phase liquide et une phase gazeuse

Résumé

Le transfert de radon entre une phase liquide et une phase gazeuse est modélisé par une condition de Robin (le flux de radon à l'interface commune est exprimé en fonction des concentrations de radon dans les deux phases). Cette condition implique deux constantes : le coefficient d'Ostwald (α) et la vitesse de transfert (β). En supposant que la valeur de α est connue, une méthode est proposée pour déterminer la valeur de β , en étudiant le phénomène de transfert du radon à l'échelle du laboratoire. En connaissant les concentrations initiales de radon, l'expérience consiste à mesurer la durée pendant laquelle il existe un flux de radon à travers l'interface commune. Pendant cette période de stabilisation, le transport de radon est gouverné dans chaque phase par la diffusion et la désintégration. Alors, la détermination de β équivaut à la résolution d'un problème inverse formulé en utilisant les données mesurées. Une procédure numérique est développée pour résoudre ce problème. *Pour citer cet article : D.-G. Calugaru, J.-M. Crolet, C. R. Mecanique 330 (2002) 377–382.* © 2002 Académie des sciences/Éditions scientifiques et médicales Elsevier SAS

milieux granulaires / interface / condition Robin / radon / coefficient d'Ostwald / problème inverse

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Version française abrégée

Des phénomènes de transport au travers d'une l'interface entre une phase liquide et une phase gazeuse apparaissent dans les Sciences de la Terre (au niveau de la surface libre d'un aquifère non confiné) [1,2], ou dans l'étude des polluants atmosphériques (à l'interface lac/atmosphère) ou pour estimer l'augmentation de la concentration du radon dans l'habitat, lors de l'utilisation de l'eau ménagère [3].

Dans ces applications, la condition de transfert appropriée entre la phase aqueuse et la phase gazeuse est de type Robin (Équation (2b)) : elle relie le flux de radon à l'interface commune et les concentrations de radon dans les deux phases. L'utilisation de cette condition nécessite la connaissance de deux constantes physiques : le coefficient d'Ostwald (noté α) [4] et la « vitesse » de transfert (notée β) [5]. La difficulté vient de la méconnaissance du coefficient β .

On n'étudie pas le problème physique dans son intégralité, mais le seul processus de transfert de radon entre une masse d'eau et une masse d'air à l'échelle du laboratoire. Le système physique considéré est montré dans une coupe verticale à la Fig. 1.

Physiquement, la période de stabilisation est une fonction décroissante de la valeur du coefficient β . Alors, la détermination de ce paramètre équivaut à la résolution d'un problème inverse : trouver la valeur de β telle que la période de stabilisation lui correspondant soit égale à celle qui a été mesurée.

Une procédure numérique est développée pour résoudre ce problème : la solution est approchée par une suite construite en trois étapes : une étape d'initialisation, où un itéré initial est déterminé par la formule (7), une étape de localisation, où l'on cherche un intervalle qui contient la solution, et une dernière étape où les itérés sont construits par dichotomie [6]. A chaque itération, il faut résoudre le problème direct i.e. déterminer la période de stabilisation correspondant à l'itéré courant. La résolution numérique du problème direct est faite en résolvant, à chaque pas du temps, deux schémas découplés (4) obtenus en discrétisant la condition de Robin par un schéma explicite. Un test numérique illustre la convergence de la suite des itérés (Tableau 1).

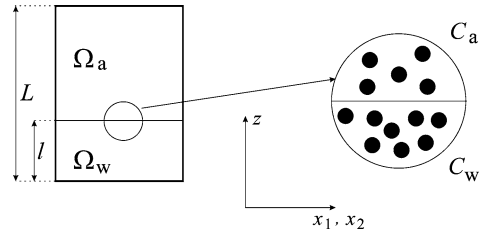
1. Introduction: Physical problem

Transport phenomena at an interface between two phases are to be taken into account in several physical problems. For instance, it is the case of radon transfert at the free surface of an unconfined aquifer [1,2]. Another situation when a similar phenomenon appears is in the evaluation of human radiation exposure due to radon coming from water sources [3]. In previous applications, the radon transfer between the aqueous phase and the gaseous phase can be modelled by a Robin's condition in which the radon flux at the common interface is expressed as a function of the radon concentrations in the two phases.

The transfert phenomenon is studied at the laboratory scale. The physical domain of the experiment is shown in Fig. 1 in a vertical plane. It contains two phases: below, a mass of water (Ω_w) and above, a mass of air (Ω_a). Let us introduce an orthogonal coordinate system (x_1, x_2, z) , where z is the vertical coordinate, while (x_1, x_2) are coordinates of the horizontal plane. Then, the domain dimensions are: L height of the domain, l height of the water domain and S surface of an horizontal cut. The system is supposed closed, in gravitational and isothermal equilibrium, and without any convective phenomenon. Then, radon transport is only governed in each phase by diffusion and radioactive decay. Neglecting capillary phenomena on the walls, the common interface is horizontal.

At the initial state, radon is assumed to be uniformly distributed in each phase and radon concentrations are known (C_w^{in} and C_a^{in}). We point out that radon concentration is given in Becquerel per m^3 (a concentration of $1 \text{ Bq}/\text{m}^3$ corresponds to approximately 476000 atoms). The experiment consists in detecting at each moment if a radon flux through the common interface exists. When no flux is recorded, the equilibrium state is reached. The measured stabilisation period is noted T_m^{st} . The measured concentrations at $t = T_m^{\text{st}}$ are respectively noted C_w^{st} and C_a^{st} . After stabilisation, the radon is again uniformly distributed in each phase.

Figure 1. Experimental domain.
Figure 1. *Le domaine expérimental.*



2. Mathematical modelling

Because radon remains always uniformly distributed in any horizontal plane, the process can be studied in a one-dimensional framework, with the following unknowns: radon concentrations in water phase (C_w) and in air phase (C_a), considered as functions of the vertical coordinate.

Even if the radon concentrations are constant in both phases at initial and final time, they are not constant at intermediary instants. Noting d_w and d_a the diffusion coefficients in two phases and λ the decay rate constant, radon transport in the two subdomains and conditions of no external perturbation are modelled by Eqs. (1). In order to obtain a well posed problem, one should impose some conditions at common interface, i.e. at $z = l$. A first condition is the continuity of radon flux (noted F) which is expressed by Eq. (2a). The radon flux F is given in Bq/(m²·s) and is defined as the number of atoms which pass through a unity of area per unity of time. Since there are two different phases, the continuity of concentration is not valid and must be replaced by a discontinuity condition. This condition is given by Eq. (2b).

$$\begin{cases} \frac{\partial C_w}{\partial t} - d_w \frac{\partial^2 C_w}{\partial z^2} + \lambda C_w = 0, & z \in (0, l), \\ \frac{\partial C_w}{\partial z} = 0, & z = 0, \end{cases} \quad \begin{cases} \frac{\partial C_a}{\partial t} - d_a \frac{\partial^2 C_a}{\partial z^2} + \lambda C_a = 0, & z \in (l, L) \\ \frac{\partial C_a}{\partial z} = 0, & z = L \end{cases} \quad (1)$$

$$F = -d_w \frac{\partial C_w}{\partial z} \Big|_{z=l_-} = -d_a \frac{\partial C_a}{\partial z} \Big|_{z=l_+} \quad (2a)$$

$$F = \beta (C_w(l_-) - \alpha C_a(l_+)) \quad (2b)$$

Since $C_w(l_-)$ and $C_a(l_+)$ evolve in time, the corresponding value of F depends also on time, α and β being two constants. Eq. (2b) shows that if for a given instant, $C_w/C_a > \alpha$ on the common interface, then $F > 0$, i.e. the radon flux is oriented from water to air and inversely, a radon flux from air to water is present if $C_w/C_a < \alpha$. We note that coefficient α can be obtained using radon concentrations in water and in air at equilibrium state: $\alpha = C_w^{st}/C_a^{st}$ (at such an instant, we have no exchange between the two phases, and then $F = 0$). This partition coefficient of radon between water and air is called Ostwald's coefficient. Its value depends on the temperature, varying from 0.525 at 0 °C to 0.226 at 25 °C [4].

While α gives the orientation of radon flux, the intensity is given by β . The value of this coefficient is much more difficult to estimate. Unlike Ostwald's coefficient which is adimensional, β has the dimension of a velocity and is called "transfer radon velocity" between the two phases [5]. Indeed, it determines effectively the transfer rate, and stabilisation period is a monotone decreasing function of β .

The method proposed in this paper is based on this idea. Assuming Ostwald's coefficient is known, one determinates the value of β from measured stabilisation period in the former experiment. Noting β_* the searched value and if measured data contain no error, then the problem of β_* determination is equal to solving the following inverse problem:

$$\text{To find } \beta_* \in \mathbf{R}^+ \quad \text{so that} \quad T^{st}(\beta_*) = T_m^{st} \quad (3)$$

3. Numerical resolution of direct problem

The direct problem associated to the problem (3) consists in finding the stabilisation period for a given value of β parameter. The problem (1), (2) is simulated by an iterative scheme in time: if radon concentrations are known at time t_k in water (C_w^k) and in air (C_a^k), then one computes the concentrations at time $t_{k+1} = t_k + dt$ in water (C_w^{k+1}) and in air (C_a^{k+1}), by solving the two following problems posed in Ω_w and respectively in Ω_a :

$$\left\{ \begin{array}{l} \frac{C_w^{k+1} - C_w^k}{dt} - d_w \frac{\partial^2 C_w^{k+1}}{\partial z^2} + \lambda C_w^{k+1} = 0, \\ \frac{\partial C_w^{k+1}}{\partial z} = 0, \quad z = 0, \\ -d_w \frac{\partial C_w^{k+1}}{\partial z} = F^k, \quad z = l, \end{array} \right. \quad \left\{ \begin{array}{l} \frac{C_a^{k+1} - C_a^k}{dt} - d_a \frac{\partial^2 C_a^{k+1}}{\partial z^2} + \lambda C_a^{k+1} = 0 \\ \frac{\partial C_a^{k+1}}{\partial z} = 0, \quad z = L \\ -d_a \frac{\partial C_a^{k+1}}{\partial z} = F^k, \quad z = l \end{array} \right. \quad (4)$$

The system is discretised in time, for each subdomain, based on fully implicit finite differences, but with an explicit discretisation of the boundary condition at the air/water interface: on the interval (t_k, t_{k+1}) , the radon flux has been approximated by its value at time t_k . Then, the two problems are uncoupled and can be solved independently using an appropriate spatial discretisation (we have also used finite differences, with a mesh refinement around the common interface).

The direct problem is simulated until the equilibrium is reached, i.e. when there is no flux between the two phases. We assume that equilibrium is reached if the stopping criteria $F^k \leq \varepsilon$ is verified, where ε is a small constant. The value of ε must correspond to the limit of detectable flux in the used measurement technique. In this situation, we define $T^{st}(\beta)$ as t_k .

4. Numerical algorithm for the inverse problem

To determine the solution β_* of inverse problem (3), an iterative algorithm has been developed, by constructing a sequence $(\beta_k)_{k \geq 0}$ which converges to β_* . The construction of this sequence is made in three steps: (i) initialisation of β , (ii) localisation of an interval I containing the solution, and (iii) bisection iterations in the interval I .

Initialisation step consists in calculating a good initial iterate in order to assure a fast convergence of the algorithm. A reasonable choice of β_0 is determined by introducing some supplementary approximations. Firstly, time evolution of radon concentrations at the common interface is approximated by:

$$C_w(l-; t) \simeq a_1 t + b_1 \exp(-\lambda t) \quad \text{and} \quad C_a(l+; t) \simeq a_2 t + b_2 \exp(-\lambda t) \quad (5)$$

where the constants $a_i, b_i, i = 1, 2$, can be computed by taking into account the values of radon concentrations on the interface at initial and stabilisation times. The former relations generalise two particular situations: firstly, if the decay is neglected ($\lambda = 0$), then they become linear laws and secondly, if there is no exchange between the two phases, then they lead to decay laws.

If $A_a(t)$ denotes total radon activity in air phase at time t , it verifies the following ordinary differential equation:

$$\frac{\partial A_a}{\partial t} = S \cdot F(t) - \lambda A_a(t)$$

This equation is integrated over $(0, T_m^{st})$. At initial and stabilisation times, radon concentrations in air are available and radon is uniformly distributed in both phases. Then, we obtain:

$$(C_a^{st} - C_a^{in})(L - l)S = \int_0^{T_m^{st}} (S \cdot F(t) - \lambda A_a(t)) dt \quad (6)$$

For the right term, we use Eq. (2b) to express $F(t)$ in function of radon concentrations at the common interface which are also replaced by Eqs. (5). An approximation of $A_a(t)$ can be obtained noting that diffusion phenomenon in the gaseous phase is much faster than in liquid phase. Then, a second

approximation is used: radon concentration in air is approximated by its value on interface, which is replaced once again by Eqs. (5). Therefore, from (6) we get the initial iterate:

$$\beta_0 = \frac{\lambda(L-l)(2 + \lambda T_m^{\text{st}})(C_a^{\text{st}} - C_a^{\text{in}} \exp(-\lambda T_m^{\text{st}}))}{(C_w^{\text{in}} - \alpha C_a^{\text{in}})(2 - \exp(-\lambda T_m^{\text{st}})(2 + \lambda T_m^{\text{st}}))} \quad (7)$$

Localisation step consists in finding two consecutive iterations such that the unknown value β_* is in the interval limited by them: if the iterate β_k is already determined, the value $T^{\text{st}}(\beta_k)$ is evaluated solving direct problem and the next iterate β_{k+1} will be defined as $2\beta_k$ if $T^{\text{st}}(\beta_k) > T_m^{\text{st}}$ and $\beta_k/2$ if $T^{\text{st}}(\beta_k) < T_m^{\text{st}}$. This step is stopped when for some k_l , we obtain $(T^{\text{st}}(\beta_{k_l}) - T_m^{\text{st}})(T^{\text{st}}(\beta_{k_l-1}) - T_m^{\text{st}}) \leq 0$ which means β_* is in the interval I delimited by β_{k_l} and β_{k_l-1} . We note (a, b) this search interval.

The last step follows the bisection method [6] to solve equation (3) on this interval. A new iterate is then obtained as $\beta_k = (a + b)/2$ and a new definition of a and b is made according to the value of $T^{\text{st}}(\beta_k)$ compared to T_m^{st} . Therefore, we obtain an interval containing β_* which is twice as small as before. This algorithm is stopped when the interval become smaller than an arbitrary value.

5. Numerical test

Numerical procedure described above has been applied for a domain with dimensions $L = 10$ cm, $l = 5$ cm, $S = 1$ m² and with initial concentrations $C_w^{\text{in}} = 100000$ Bq/m³ and $C_a^{\text{in}} = 10$ Bq/m³. Ostwald's coefficient $\alpha = 0.25$ has been used. Other used values are $d_w = 1.4 \times 10^{-9}$ m²/s, $d_w = 1.03 \times 10^{-5}$ m²/s and $\lambda = 2.1 \times 10^{-6}$ s⁻¹. The “measured” data have been obtained by solving direct problem for a given value of β parameter. Thus, for $\beta_* = 10^{-6}$ m/s, it has been found $T_m^{\text{st}} = 8.15$ days, $C_w^{\text{st}}(l_-) = 4563$ Bq/m³ and $C_a^{\text{st}}(l_+) = 18232$ Bq/m³. The ratio of these concentrations is approximately 0.25 (the used Ostwald coefficient). The value $\varepsilon = 5 \times 10^{-6}$ Bq/(m²s) has been used as a stopping criteria.

For the previous considered values, the initial iterate, obtained by Eq. (7), is $\beta_0 = 0.055 \times 10^{-6}$. Numerical convergence of algorithm is presented in Table 1. From previous initial value β_0 , it has been found that the “searched” value is between $\beta_4 = 0.88 \times 10^{-6}$ and $\beta_5 = 1.76 \times 10^{-6}$ (localisation step). The convergence to β_* is assured by the bisection method. The algorithm is stopped when $(\beta_{k+1} - \beta_k)/\beta_k \leq$

Table 1. Iterates in localisation step (1–5) and by bisection method (6–15).
Tableau 1. Les itérés dans l'étape de localisation (1–5) et par dichotomie (6–15).

Iteration (k)	β_k [$\times 10^{-6}$ m/s]	$T^{\text{st}}(\beta_k)$ [days]	$(T^{\text{st}}(\beta_k) - T_m^{\text{st}})(T^{\text{st}}(\beta_{k-1}) - T_m^{\text{st}})$
0	0.055	76.50	–
1	0.110	40.24	+2193.35
2	0.220	22.15	+449.26
3	0.441	13.15	+70.00
4	0.882	8.67	+2.60
5	1.765	6.40	–0.91
6	1.323	7.17	–
7	1.103	7.77	–
...	–
14	1.001	8.14	–
15	1.000	8.15	–

10^{-3} and the iterate $\beta_{15} = 1.0006 \times 10^{-6}$ is obtained, which is a very good approximation of “searched” value $\beta_* = 10^{-6}$.

6. Conclusion

A numerical method allowing to evaluate the transfer radon velocity between two phases has been proposed. It is based on analysis of recorded data in an experiment which consists in measuring the stabilisation time. Mathematical modelling and numerical tools are then used to estimate the transfer coefficient which governs radon exchanges between aqueous phase and gaseous phase.

However, the method can be also adapted when data are available long time before stabilisation state. Moreover, even if the method is described for a mass of water in contact with a mass of air, it can be adapted for a saturated natural porous medium in contact with a dry porous medium by considering the process (at laboratory scale) with samples of two media.

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