Paper CIT04-0442

PREDICTION OF ^{222}Rn EXHALATION RATES FROM PHOSPHOGYPSUM BASED STACKS. PART I: PARAMETRIC MATHEMATICAL MODELING

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Abstract. Radon-222 is a radionuclide exhaled from phosphogypsum by-produced at phosphate fertilizer industries. Alternative large-scale application of this waste may indicate a material substitute for civil engineering provided that environmental issues concerning its disposal and management are overcome. The first part of this paper outlines a steady-state two-dimensional model for ^{222}Rn transport through porous media, inside which emanation (source term) and decay (sink term) exist. Boussinesq approach is evoked for the laminar buoyancy-driven interstitial air flow, which is also modeled according to Darcy-Brinkman formulation. In order to account for simultaneous effects of entailed physical parameters, governing equations are cast into dimensionless form. Apart from usual controlling parameters like Reynolds, Prandtl, Schmidt, Grashof and Darcy numbers, three unconventional dimensionless groups are put forward. Having in mind ^{222}Rn transport in phosphogypsum-bearing porous media, the physical meaning of those newly introduced parameters and representative values for the involved physical parameters are presented. A limiting diffusion-dominated scenario is addressed, for which an analytical solution is deduced for boundary conditions including an impermeable phosphogypsum stack base and a non-zero fixed concentration activity at the stack top. Accordingly, an expression for the average Sherwood number corresponding to the normalized ^{222}Rn exhalation rate is presented.

Keywords. radon transport, phosphogypsum, porous media, heat-mass transfer, mathematical modeling

1. Introduction

Phosphogypsum is a by-product from phosphate fertilizer industries. Since it has presently little or maybe no economic value, it has been simply disposed in stacks. In view of that, huge demands for fertilizers with high P_2O_5 concentration have yielded several piles of such waste material worldwide (Rutherford *et al.*, 1994). Alternative commercial exploitations of phosphogypsum have been sought (Mazzilli *et al.*, 2000). In agriculture, it can be used as soil amendment although only in small amounts. In contrast, its large-scale utilization as a substitute material for civil engineering (e.g., building component, embankment filling and road base) still has to cope with environmental issues, notably as far as ^{222}Rn exhalation rates are concerned.

Along with its short-lived decay products, ^{222}Rn responds to most of human exposure to radiation from natural sources (UNSCEAR, 2000). Such radioactive gas is the most important radon isotope because it is able to reach the environment thanks to its half-life of 3.824 days. If inhaled, its progeny is likely to decay to ^{210}Pb (half-life = 22.3 years) before being removed by lung clearance mechanisms. Although higher lung cancer risk is assigned to the radiation released by those short-lived decay products (Nero, 1988), radiation exposure assessment and radiological protection design are based on ^{222}Rn exhalation rates. In USA for instance, such rates from inactive phosphogypsum stacks are restricted to a maximum of 0.74 Bq·m^{-2}·s^{-1} (US Government, 1998).

Radon-222 belongs to the ^{238}U chain and it results from the α-decay of ^{226}Ra. This latter radionuclide is commonly found in phosphogypsum as an impurity. Hence, ^{226}Ra present in phosphogypsum-bearing material may decay to ^{222}Rn, which is able to travel through the layers, reach up the open atmosphere and finally be inhaled by nearby humans. Consequently, ^{222}Rn entry rates into local air depend intimately on its concentration within some distance from the free surface of the porous matrix. Because many physical factors are involved (e.g. emanation rate, material porosity and permeability, moisture content, temperature and ^{222}Rn diffusivity), analysis of phosphogypsum environmental issues should rely on comprehensive knowledge of ^{222}Rn generation and transport within porous media.

Added risk of lung cancer related to exposures to ^{222}Rn decay products was first recognized in the 1950s among uranium miners while concerns towards indoor ^{222}Rn concentration were brought into light in the 1970s. Several investigations have been conducted to measure such concentrations and to correlate them to known physical parameters. Initial results pointed to soil as a major source of naturally occurring high indoor concentrations though building materials can play an important role (Nero, 1988). This may particularly be true if phosphogypsum is used as an alternative material. Either way, ^{222}Rn transport takes place inside a porous matrix.

Transport phenomena in porous media find many engineering applications (Kaviany, 1994; Nield and Bejan, 1999). Models for ^{222}Rn transport in porous media have considered both diffusion and convection but early models have basically taken into account air flow driven by pre-defined pressures differences and Darcy's law (Loureiro, 1987). This trend has been followed until present day (Andersen, 2000) as it yields a Poisson (transient transport) or a Laplace (steady-state transport) equation to be solved for pressure which may simplify the numerical implementation of the transport equations in a computational program.

Recent contributions include a theoretical study on ^{222}Rn transient diffusion and decay in activated charcoal (Nikezic and Urosevic, 1998) and a simple model for ^{222}Rn and ^{210}Pb transient transport through the atmosphere (Piliposian and Appleby, 2003). Analytical solutions for steady-state diffusion including both decay and emanation processes within phosphogypsum stacks were presented by Rabi and Silva (2002; 2003) for two different boundary conditions prevailing at the top free surface. In order to account for concomitant effects of controlling parameters, their first preliminary analysis was cast into dimensionless form (Rabi and Mohamad, 2004).

The first part of this paper intends to broaden that last mentioned analysis by coupling flow and temperature fields through the inclusion of natural convective effects. Depending on external air flow condition, distinct dimensionless variables can be put forward. An analytical solution for a limiting physical scenario (namely, diffusion-dominated) is presented and discussed.

2. Model assumptions for ^{222}Rn transport in porous media

A representative elementary volume (REV) lying inside a phosphogypsum-based stack may comprise solid grains and interstices filled up with air and water (if wet stack is considered). The vertical cross-section of a stack usually assumes a trapezoidal shape, as sketched in Fig. 1(*a*). Nonetheless, for modeling purposes, it is approximated by a rectangle of (half) width L (m) and height H (m) as depicted in Fig. 1(*b*), which also shows the related Cartesian coordinate system. In this work, the stack is regarded as a porous medium sufficiently large in the direction normal to the plane of Fig. 1, whose porosity ε and permeability K (m^2) are allegedly constant, the latter being further assumed to be isotropic, i.e., it presents no directional dependence.
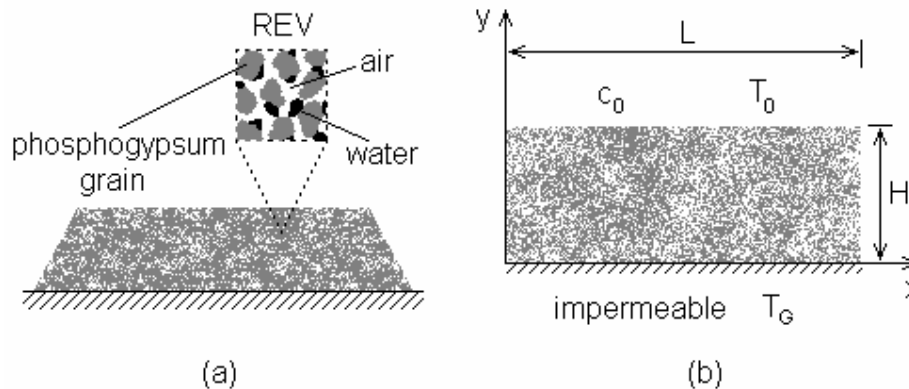

Figure 1. (*a*) Sketch of a phosphogypsum-bearing stack and a representative elementary volume (REV) inside it; (*b*) schematic diagram of a vertical cross-section and coordinate system.

Neglecting internal heat sources (e.g. due to chemical reactions or radioactivity), local thermodynamic equilibrium can be evoked so that the temperature of all phases (phosphogypsum grains, air and water) are the same, i.e., $T_p = T_a = T_w = T$ (K). All thermo-physical properties are considered constant with the exception of air density ρ_a (kg·m^{-3}) in the buoyancy term. In other words, incompressible flow and Boussinesq approximation are assumed to hold and a linear dependence of ρ_a on local temperature T is supposed

$$\rho_a(T) = \rho_0[1 - \beta(T - T_0)] \quad , \quad \text{where} \quad \beta = -\frac{1}{\rho_0}\left(\frac{\partial \rho_a}{\partial T}\right)_0 \tag{1}$$

Density ρ_0 and temperature T_0 are reference state values and β (K^{-1}) is the coefficient of thermal volumetric expansion.

The outstanding feature of ^{222}Rn transport in porous media containing ^{226}Ra particles is the fact that there exist both source and sink terms. They are respectively related to emanation and decay processes and should be properly accounted for in the species concentration equation. This paper assumes that ^{226}Ra impurities are evenly distributed throughout the stack, residing inside phosphogypsum grains only.

Local sinks are directly proportional to the total mobile ^{222}Rn activity dξ_{mob} (Bq) within the volume element dΩ (m^3) of the REV. The proportionality factor is the ^{222}Rn decay constant λ (s^{-1}) so that the sink term becomes

$$\lambda d\xi_{mob} = \lambda\varepsilon_c c_a d\Omega \tag{2}$$

where the dimensionless coefficient ε_c is the so-called partition-corrected porosity (Andersen, 2000) and c_a (Bq·m^{-3}) is the ^{222}Rn activity concentration in the interstitial air.

Internal ^{222}Rn activity sources refer to the ^{226}Ra concentration c_{Ra} (Bq·kg^{-1}), supposedly uniform and expressed on a mass basis. If ρ_p is the phophogypsum density, it has been shown elsewhere (Andersen, 2000) that the ^{222}Rn activity concentration source term within the REV is given by

$$\lambda E(1-\varepsilon)\rho_p d\Omega = \varepsilon G d\Omega \tag{3}$$

where E (Bq·kg^{-1}) is the number of ^{222}Rn particles emanated from ^{226}Ra radionuclides per unit of time and per unit of phosphogypsum-grain mass (on a dry basis) whereas the quantity

$$G = \lambda E\frac{1-\varepsilon}{\varepsilon}\rho_p \tag{4}$$

is interpreted as the ^{222}Rn activity generation rate per unit of interstitial volume (Bq·m^{-3}·s^{-1}).

It is common to express E in terms of an emanation coefficient f, also known as emanating fraction or emanating power (Nazaroff *et al.*, 1988), and the concentration c_{Ra} as

$$E = f\, c_{Ra} \tag{5}$$

With the help of Eq. (4) and if $\tilde{\rho}_{dry} = (1-\varepsilon)\rho_p$ is the REV bulk density on a dry-basis, the ^{222}Rn activity generation rate per unit of REV bulk volume $\tilde{G}$ (Bq·m^{-3}·s^{-1}) can be defined as

$$\tilde{G} = \varepsilon G = \lambda\, f\, c_{Ra}\tilde{\rho}_{dry} \tag{6}$$

3. Governing equations for ^{222}Rn transport in porous media

3.1. Primitive variables formulation

Under the aforementioned model assumptions and adopting Darcy-Brinkman-Boussinesq formulation including convective inertia terms, the steady-state 2-D Cartesian governing equations for bulk air mass, momentum, energy and species (^{222}Rn activity) concentration inside the porous matrix can be expressed respectively as

$$\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} = 0 \tag{7}$$

$$u\frac{\partial u}{\partial x} + v\frac{\partial u}{\partial y} = \tilde{\upsilon}\left(\frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2}\right) - \frac{1}{\rho_0}\frac{\partial p}{\partial x} - \frac{\upsilon}{K}u \tag{8}$$

$$u\frac{\partial v}{\partial x} + v\frac{\partial v}{\partial y} = \tilde{\upsilon}\left(\frac{\partial^2 v}{\partial x^2} + \frac{\partial^2 v}{\partial y^2}\right) - \frac{1}{\rho_0}\frac{\partial p}{\partial y} - \frac{\upsilon}{K}v + g\beta(T - T_0) \tag{9}$$

$$u\frac{\partial T}{\partial x} + v\frac{\partial T}{\partial y} = \tilde{\alpha}\left(\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2}\right) \tag{10}$$

$$u\frac{\partial c_a}{\partial x} + v\frac{\partial c_a}{\partial y} = \tilde{D}\left(\frac{\partial^2 c_a}{\partial x^2} + \frac{\partial^2 c_a}{\partial y^2}\right) + \tilde{G} - \varepsilon_c\lambda c_a \tag{11}$$

where u and v are the velocity components (m·s^{-1}) respectively in x and y directions, p (Pa) is the pressure, υ (m^2·s^{-1}) is the kinematic viscosity (REV bulk values wherever needed), g (m·s^{-2}) is the gravity acceleration, $\tilde{\alpha}$ (m^2·s^{-1}) is the REV

bulk thermal diffusivity and $\tilde{D}$ (m^2·s^{-1}) is the ^{222}Rn REV bulk mass diffusivity. It is worth noting that ^{222}Rn activity sources and sinks are accounted for in the species equation, Eq. (11).

At this point, comments about the nomenclature are meaningful. The bulk diffusivity $\tilde{D}$ is to be used whenever the ^{222}Rn flux refers to the geometric (bulk) cross-sectional area. If Fick's law of diffusion is set up in terms of the pore system cross-sectional area, the so-called "interstitial" diffusivity should be used (Nazaroff *et al.*, 1988; Yu *et al.*, 1993). In these two last references, such coefficient is also called "effective" diffusivity, although such terminology may bring about confusion with the usual meaning attributed to the term "effective" in the literature on porous media. Accordingly, "interstitial" diffusivity should be preferred as it is unique.

3.2. Dimensionless governing equations and parameters: laminar external flow

The previous governing equations can be written in dimensionless form as an attempt to deal with concurrent effects of physical parameters influencing the ^{222}Rn transport. As far as pressure p and velocity components u and v are concerned, different dimensionless variables may be defined depending on the condition of the external air flow.

One possibility is external air flowing in laminar regime (e.g. breeze or mild wind). It may be further assumed that the incoming flow is parallel to the ground, i.e. the x axis in Fig. 1(*b*), so that u_0 (m·s^{-1}) is the free stream velocity. The following set of dimensionless variables is put forward:

$$X = \frac{x}{H} \quad , \quad Y = \frac{y}{H} \quad , \quad U = \frac{u}{u_0} \quad , \quad V = \frac{v}{u_0} \quad , \quad P = \frac{p}{\rho_0 u_0^2} \quad , \quad \theta = \frac{T - T_0}{\Delta T} \quad , \quad \phi = \frac{c_a - c_0}{\Delta c} \quad (12)$$

Scaling reference values ΔT and Δc can be properly defined depending on the physics of the problem.

Moreover, a dimensionless parameter n can be introduced to indicate whether the flow takes place inside or outside the porous matrix. In line with (Mohamad, 2003), $n = 1$ corresponds to flow within porous medium whereas $n = 0$ indicates flow in a region without porous material. Insertion of Eqs. (12) into the governing equations, Eqs. (7) to (11), yields the following set of dimensionless equations:

$$\frac{\partial U}{\partial X} + \frac{\partial V}{\partial Y} = 0 \tag{13}$$

$$U \frac{\partial U}{\partial X} + V \frac{\partial U}{\partial Y} = \frac{\Gamma^n}{Re}\left(\frac{\partial^2 U}{\partial X^2} + \frac{\partial^2 U}{\partial Y^2} \right) - \frac{\partial P}{\partial X} - \frac{n\,U}{Re\,Da} \tag{14}$$

$$U \frac{\partial V}{\partial X} + V \frac{\partial V}{\partial Y} = \frac{\Gamma^n}{Re}\left(\frac{\partial^2 V}{\partial X^2} + \frac{\partial^2 V}{\partial Y^2} \right) - \frac{\partial P}{\partial Y} - \frac{n\,V}{Re\,Da} + \frac{Gr}{Re^2}\theta \tag{15}$$

$$U \frac{\partial \theta}{\partial X} + V \frac{\partial \theta}{\partial Y} = \frac{\Lambda^n}{Re\,Pr}\left(\frac{\partial^2 \theta}{\partial X^2} + \frac{\partial^2 \theta}{\partial Y^2} \right) \tag{16}$$

$$U \frac{\partial \phi}{\partial X} + V \frac{\partial \phi}{\partial Y} = \frac{\Psi^n}{Re\,Sc}\left(\frac{\partial^2 \phi}{\partial X^2} + \frac{\partial^2 \phi}{\partial Y^2} \right) + \frac{1}{Re\,Sc}\left[nS - \varepsilon_c^n R(\phi - \phi_0) \right] \tag{17}$$

Dimensionless groups typically encountered in natural-convective heat-mass transfer in porous media arise as expected, like Reynolds, Darcy, Grashof, Prandtl and Schmidt numbers:

$$Re = \frac{u_0 H}{\upsilon} \quad , \quad Da = \frac{K}{H^2} \quad , \quad Gr = \frac{g\,\beta\,\Delta T\,H^3}{\upsilon^2} \quad , \quad Pr = \frac{\upsilon}{\alpha} \quad , \quad Sc = \frac{\upsilon}{D_o} \tag{18}$$

Further dimensionless parameters characterizing heat and mass transfer in porous media are bulk-to-fluid property ratios, in this case for kinematic viscosity $\Gamma = \tilde{\upsilon}/\upsilon$, thermal diffusivity $\Lambda = \tilde{\alpha}/\alpha$ and mass diffusivity $\Psi = \tilde{D}/D_o$. With respect to the last of Eqs. (12), the dimensionless activity level ϕ_0 in Eq. (17) corresponds to the value of ϕ when the ^{222}Rn activity concentration in air is $c_a = 0$, i.e. $\phi_0 = -c_0/\Delta c$, where c_0 is the local outdoor activity concentration.

Apart from the usual controlling parameters above, Eq. (17) puts forward two unconventional dimensionless groups R and S directly related to decay and emanation processes. They are defined according to

$$R = \frac{\lambda H^2}{D_o} \quad \text{and} \quad S = \frac{\tilde{G} H^2}{D_o \Delta c} \tag{19}$$

While R measures the relative importance of decay in relation to diffusion, S is here referred to as an emanation-to-diffusion ratio. A surrogate dimensionless group M can be proposed as

$$M = \frac{S}{R} = \frac{\tilde{G}}{\lambda \Delta c} \tag{20}$$

which can be interpreted as an emanation-to-decay ratio. Although this parameter is still sensitive to the reference value Δc for activity concentration, it has the advantage of being independent from both the open-air diffusivity D_o and the characteristic length H. In terms of R and M, Eq. (17) is readily recast as follow

$$U \frac{\partial \phi}{\partial X} + V \frac{\partial \phi}{\partial Y} = \frac{\Psi^n}{\text{Re Sc}} \left(\frac{\partial^2 \phi}{\partial X^2} + \frac{\partial^2 \phi}{\partial Y^2} \right) + \frac{R}{\text{Re Sc}} \left[n M - \varepsilon_c^n (\phi - \phi_0) \right] \tag{21}$$

3.3. Dimensionless governing equations and parameters: negligible external flow

Another possibility refers to the situation when wind effects are disregarded, so that u_0 is comparatively small. Having in mind such boundary condition, the set of dimensionless variables is:

$$X = \frac{x}{H} \quad , \quad Y = \frac{y}{H} \quad , \quad U = \frac{uH}{\upsilon} \quad , \quad V = \frac{vH}{\upsilon} \quad , \quad P = \frac{pH^2}{\rho_0 \upsilon^2} \quad , \quad \theta = \frac{T - T_0}{\Delta T} \quad , \quad \phi = \frac{c_a - c_0}{\Delta c} \tag{22}$$

where U, V and P are properly redefined. In this case, after introducing Eqs. (22) into Eqs. (7) to (11), the dimensionless governing equations become:

$$\frac{\partial U}{\partial X} + \frac{\partial V}{\partial Y} = 0 \tag{23}$$

$$U \frac{\partial U}{\partial X} + V \frac{\partial U}{\partial Y} = \Gamma^n \left(\frac{\partial^2 U}{\partial X^2} + \frac{\partial^2 U}{\partial Y^2} \right) - \frac{\partial P}{\partial X} - n \frac{U}{\text{Da}} \tag{24}$$

$$U \frac{\partial V}{\partial X} + V \frac{\partial V}{\partial Y} = \Gamma^n \left(\frac{\partial^2 V}{\partial X^2} + \frac{\partial^2 V}{\partial Y^2} \right) - \frac{\partial P}{\partial Y} - n \frac{V}{\text{Da}} + \text{Gr}\theta \tag{25}$$

$$U \frac{\partial \theta}{\partial X} + V \frac{\partial \theta}{\partial Y} = \frac{\Lambda^n}{\text{Pr}} \left(\frac{\partial^2 \theta}{\partial X^2} + \frac{\partial^2 \theta}{\partial Y^2} \right) \tag{26}$$

$$U \frac{\partial \phi}{\partial X} + V \frac{\partial \phi}{\partial Y} = \frac{\Psi^n}{\text{Sc}} \left(\frac{\partial^2 \phi}{\partial X^2} + \frac{\partial^2 \phi}{\partial Y^2} \right) + \frac{1}{\text{Sc}} \left[n S - \varepsilon_c^n R (\phi - \phi_0) \right] \tag{27}$$

while Eq. (21) is replaced by

$$U \frac{\partial \phi}{\partial X} + V \frac{\partial \phi}{\partial Y} = \frac{\Psi^n}{\text{Sc}} \left(\frac{\partial^2 \phi}{\partial X^2} + \frac{\partial^2 \phi}{\partial Y^2} \right) + \frac{R}{\text{Sc}} \left[n M - \varepsilon_c^n (\phi - \phi_0) \right] \tag{28}$$

It is worth mentioning that Nazaroff *et al.* (1988), analyzing transient pressure-driven [222]Rn migration in soil, likewise introduced two dimensionless groups in the species ([222]Rn activity) concentration equation. They defined a Péclet number Pe for mass transfer in porous media and a convection-to-decay ratio N. Bearing in mind that the Péclet number measures the relative importance of convection with respect to diffusion, it is interesting to verify that

$$\frac{\text{Pe}}{\text{N}} = \frac{convection/diffusion}{convection/decay} = \frac{decay}{diffusion} \quad \Rightarrow \quad \frac{\text{Pe}}{\text{N}} \cong \text{R} \tag{29}$$

as confirmed by recalling the proposed definitions for Pe and N.

4. Radon transport within phosphogypsum

4.1. Representative values of entailed physical parameters

The number of physical parameters concurrently influencing the ^{222}Rn transport in porous media, particularly in phosphogypsum, is quite large. As far as boundary conditions are concerned, a further parameter for 2-D domains is introduced – the aspect ratio $A = L/H$ – while initial conditions are needed if problems are set in transient formulation. The analysis can be simplified if governing equations are written in dimensionless form and physical parameters are conveniently grouped. Still, it is impractical to address systematically the effects of each controlling dimensionless group. Conversely, values for some parameters can be inferred for particular physical scenarios.

The ^{222}Rn decay constant is $\lambda = 2.098 \times 10^{-6}$ s^{-1} (UNSCEAR, 2000) and its mass diffusivity in open air is reported as $D_o = 1.2 \times 10^{-5}$ m$^2 \cdot$s^{-1} (Nazaroff *et al.*, 1988) and as $D_o = 1.1 \times 10^{-5}$ m$^2 \cdot$s^{-1} (Yu *et al.*, 1993). Correlations to evaluate $\tilde{D}$ from D_o are also found in these two references. A somewhat lower bulk value $\tilde{D}$ is expected inside porous media and typical values for ^{222}Rn transport in soil are of the order $\tilde{D} \cong 10^{-6}$ m$^2 \cdot$s^{-1} (Nazaroff *et al.*, 1988; Andersen, 2000; UNSCEAR, 2000).

If air is the saturating fluid, $\text{Pr} \cong 0.71$ and $\upsilon \cong 1.5 \times 10^{-5}$ m$^2 \cdot$s^{-1} are representative values if room temperature is considered (Kays and Crawford, 1993). A simplifying assumption concerning bulk-to-fluid property ratios is to assume $\Gamma = \Lambda = 1$ (Mohamad and Bennacer, 2001; Mohamad and Bennacer, 2002; Bennacer *et al.*, 2003) but the same cannot hold for Ψ, as discussed in the previous paragraph.

If ε_a and ε_w are the air-based and the water-based porosities, the partition-corrected porosity ε_c is defined as

$$\varepsilon_c = \varepsilon_a + \kappa_w \varepsilon_w + \kappa_s \rho_{dry} \tag{30}$$

where κ_s and κ_w are referred to as radon surface sorption coefficient and Ostwald partitioning coefficient respectively (Andersen, 2000). The latter is temperature dependent, ranging from 0.5249 at 0°C down to 0.1797 at 35°C. For a dry medium ($\varepsilon_w = 0$) without grain sorption ($\kappa_s = 0$), it follows that the partition-corrected porosity reduces to $\varepsilon_c = \varepsilon_a = \varepsilon$.

In general, quite low permeability values are reported for phosphogypsum stacks (May and Sweeney, 1982; Braz *et al.*, 2002) but they may depend upon the deposition process. Braz *et al.* (2002) determined the phosphogypsum density ρ_p from two distinct samples as 2270 and 2330 kg$\cdot$m^{-3}, both within the range presented by Rutherford *et al.* (1994). Based on their figures, they estimated the phosphogypsum stack porosity as $\varepsilon \sim 0.5$. For soil applications, permeability and porosity may show some variation, mainly the former (Kaviany, 1994; Nield and Bejan, 1999).

Braz *et al.* (2002) also determined the ^{222}Rn emanation coefficient as $f = 0.035$, consistent with values shown by Rutherford *et al.* (1995) but lower than the range $0.245 - 0.340$ obtained by Hartley and Freeman (1986). Mazzilli *et al.* (2000) measured the ^{226}Ra activity concentration c_{Ra} within different phosphogypsum samples and results ranged from 22 to 695 Bq$\cdot$kg^{-1} while references suggesting an appreciably higher range for c_{Ra} are cited by Stranden (1988). Recent outdoors ^{222}Rn concentration measurements estimate $c_0 \cong 10$ Bq$\cdot$m^{-3} (UNSCEAR, 2000).

Nevertheless, it is worth bearing in mind that local physical conditions (e.g. temperature, humidity and pressure) may alter the values of parameters presented above. This adds more complexity to the problem and the solution process is likely to rely on numerical methods.

4.2. Limiting scenario: diffusion-dominated ^{222}Rn transport

Apart from possibly varying physical parameters and the presence of source and sink terms in the species equation, a comprehensive model for ^{222}Rn transport within porous media may include natural convective effects, transient phenomena and be extended up to 3-D domains. Such ^{222}Rn transport model does not render itself a simple formulation since bulk mass, momentum and energy equations are inevitably evoked. Nazaroff *et al.* (1988) already pointed at two distinct approaches to this problem: approximate analysis and numerical modeling. In the second part of this paper, a test case for ^{222}Rn transport in a phosphogypsum stack is numerically solved with the help of an existing simulator for heat and mass transfer in porous media.

Analytical solutions are only feasible for limiting physical scenarios. Nonetheless, they can play a major role as far as numerical validation is concerned. A common simplification is to neglect one of the transport processes (diffusion or convection) with respect to the other. Given the low permeability of phosphogypsum stacks, one possibility is to disregard interstitial air flow, $U \approx V \approx 0$. In other words, the interstitial ^{222}Rn transfer becomes diffusion-dominated.

As a first approach, external air is supposed to be at rest, $u_0 = 0$. In view of that, it is readily seen that Eq. (27), or equally Eq. (28), is the single governing equation whose left-hand side reduces to zero. For a stack having its horizontal width L much larger then its vertical height H, in connection with Fig. 1(b), it is reasonable to suppose that the most significant ^{222}Rn transfer rates occur along the direction normal to the top free surface. It is implied that

$$\frac{\partial \phi}{\partial Y} \gg \frac{\partial \phi}{\partial X} \tag{31}$$

The same rationale can be applied for porous media vertically stratified with respect to the ^{222}Rn activity concentration so that larger ϕ gradients are established in the Y direction. Either way, Eqs. (27) and (28) simplify to

$$\frac{d^2 \phi}{dY^2} + S - \varepsilon_c R(\phi - \phi_0) = 0 \quad \Leftrightarrow \quad \frac{d^2 \phi}{dY^2} + R[M - \varepsilon_c(\phi - \phi_0)] = 0 \tag{32}$$

Next, an analytical solution is deduced for the first form of the above linear second-order non-homogeneous O.D.E. (the one in terms of S and R). A variable change of the form $\Phi = S - R_c(\phi - \phi_0)$, where $R_c = \varepsilon_c R$, converts the first of Eqs. (32) into the following homogeneous O.D.E.:

$$\frac{d^2 \Phi}{dY^2} - R_c \Phi = 0 \tag{33}$$

With reference to Fig. 1(b), boundary conditions adopted are impermeability at the stack base ($Y = 0$) whereas the top ($Y = 1$) is subjected to fixed ^{222}Rn activity concentration c_0. These two conditions are expressed respectively as

$$\left(\frac{d\phi}{dY}\right)_{Y=0} = 0 \quad \Rightarrow \quad \left(\frac{d\Phi}{dY}\right)_{Y=0} = 0 \quad \text{and} \quad \phi_{Y=1} = 0 \quad \Rightarrow \quad \Phi_{Y=1} = S + R_c \phi_0 \tag{34}$$

The solution of Eq. (33) subjected to the previous boundary conditions is

$$\Phi = \Phi(Y) = (S + R_c \phi_0) \frac{\cosh(Y\sqrt{R_c})}{\cosh(\sqrt{R_c})} \tag{35}$$

or, back in terms of the dimensionless activity concentration,

$$\phi = \phi(Y) = \left(\frac{S}{\varepsilon_c R} + \phi_0\right)\left[1 - \frac{\cosh(Y\sqrt{\varepsilon_c R})}{\cosh(\sqrt{\varepsilon_c R})}\right] = \left(\frac{M}{\varepsilon_c} + \phi_0\right)\left[1 - \frac{\cosh(Y\sqrt{\varepsilon_c R})}{\cosh(\sqrt{\varepsilon_c R})}\right] \tag{36}$$

This problem can be further simplified assuming null external concentration, i.e., $c_0 = 0 \Rightarrow \phi_0 = 0$.

4.3. ^{222}Rn exhalation rates

As mentioned, ^{222}Rn exhalation rates are essential for radiation exposure assessment and radiological protection design. Yet, care should be exercised when presenting and analyzing results since radiological impact concerns not only with diffusive fluxes j_{diff} (Bq·m^{-2}·s^{-1}) but also with ^{222}Rn activity advected out of the stack j_{conv} (Nero, 1988; Andersen, 2000). Local ^{222}Rn exhalation rate from the phosphogypsum stack top corresponds to the total flux j_x, which can be given as a product of a ^{222}Rn activity transfer coefficient γ_x (m·s^{-1}) and the activity concentration reference value Δc as

$$j_x = j_{\text{conv,x}} + j_{\text{diff,x}} = \gamma_x \Delta c \tag{37}$$

Dimensionless mass transfer rates are usually expressed via Sherwood number and $\Delta j = \tilde{D}\Delta c / H$ can be a scale for the previous fluxes. Evoking Fick's law, a normalized local diffusive flux $J_{\text{diff,x}}$ from the stack top is evaluated as

$$J_{\text{diff,x}} = \frac{j_{\text{diff,x}}}{\Delta j} = \frac{1}{\Delta j}\left[-\tilde{D}\left(\frac{\partial c_a}{\partial y}\right)_{y=H}\right] \quad \Rightarrow \quad J_{\text{diff,x}} = -\left(\frac{\partial \phi}{\partial Y}\right)_{Y=1} \tag{38}$$

Conversely, with the help of Eqs. (18) and (22) and recalling that $\phi_0 = -c_0/\Delta c$, a normalized local convective flux $J_{conv,x}$ from the stack top can be inferred as

$$J_{conv,x} = \frac{j_{conv,x}}{\Delta j} = \frac{1}{\Delta j}(vc_a)_{y=H} \quad \Rightarrow \quad J_{conv,x} = \frac{Sc}{\Psi}[V(\phi - \phi_0)]_{Y=1} \tag{39}$$

Having in mind the definition for Δj, insertion of (38) and (39) into (37) leads to

$$\gamma_x = \frac{\tilde{D}}{H}(J_{conv,x} + J_{diff,x}) = \frac{\tilde{D}}{H}\left[\frac{Sc}{\Psi}V(\phi - \phi_0) - \frac{\partial \phi}{\partial Y}\right]_{Y=1} \tag{40}$$

or, in terms of the local Sherwood number Sh_x,

$$Sh_x = \frac{x\gamma_x}{\tilde{D}} = X\left[\frac{Sc}{\Psi}V(\phi - \phi_0) - \frac{\partial \phi}{\partial Y}\right]_{Y=1} \tag{41}$$

It can be readily verified that the average Sherwood number Sh is given by

$$\gamma = \frac{1}{L}\int_0^L \gamma_x dx \quad \Rightarrow \quad Sh = \frac{H\gamma}{\tilde{D}} = \frac{1}{A}\int_0^A \left[\frac{Sc}{\Psi}V(\phi - \phi_0) - \frac{\partial \phi}{\partial Y}\right]_{Y=1} dX = J_{conv} + J_{diff} \tag{42}$$

In other words, the average Sherwood number encompasses both average advective and diffusive ^{222}Rn activity fluxes.

Finally, the analytical solution $\phi(Y)$ for the phosphogypsum stack analyzed in the preceding section, Eq. (36), can be introduced in the above expression. Recalling that interstitial fluid flow was neglected (so that $U \approx V \approx 0$), the expression to be integrated no longer depends on X so that the average Sherwood number related to the average dimensionless ^{222}Rn exhalation rate from the stack top results to

$$Sh = \left(\frac{S}{\varepsilon_c R} + \phi_0\right)\sqrt{\varepsilon_c R}\tanh(\sqrt{\varepsilon_c R}) = \left(\frac{M}{\varepsilon_c} + \phi_0\right)\sqrt{\varepsilon_c R}\tanh(\sqrt{\varepsilon_c R}) \tag{43}$$

5. Concluding remarks

The first part of this paper presented the dimensionless model equations for steady-state 2-D ^{222}Rn transport within porous media including natural convection according to Darcy-Brinkman-Boussinesq formulation. Depending on the external air flow condition, dimensionless variables can be defined distinctly as far as pressure and velocity components are concerned. Apart from usual controlling dimensionless groups like Re, Pr, Sc, Gr and Da numbers, three unconventional dimensionless parameters were put forward, namely, a decay-to-diffusion ratio, an emanation-to-diffusion ratio and a surrogate emanation-to-decay ratio.

Diffusion-dominated ^{222}Rn transport is a limiting physical scenario in which negligible interstitial air flow is a reasonable assumption. Accordingly, an analytical solution was deduced for boundary conditions including non-zero fixed concentration activity at the top of the phosphogypsum stack and no-flux condition at its base. For this particular case, it is possible to infer a formal expression for the average Sherwood number, which is a measure of the normalized ^{222}Rn exhalation rate from the stack top.

Numerous physical parameters are involved in the ^{222}Rn transport within porous media, as for instance inside phosphogypsum-bearing materials. As shown in this paper, a promising way to account for their concurrent effects is to cast the governing equations into dimensionless form. Yet, analogous to many heat and mass transfer problems in porous media, a comprehensive approach for ^{222}Rn transport in phosphogypsum stacks is likely to rely on numerical simulation.

6. Acknowledgement

7. References

Andersen C.E., 2000, "Radon Transport Modelling: User's Guide to RnMod3d", Riso-R-1201(EN), Riso National Laboratory, Roskilde, Denmark.

Bennacer R., Beji H., Mohamad A.A., 2003, "Double diffusive convection in a vertical enclosure inserted with two saturated porous layers confining a fluid layer", Int. J. Thermal Sciences, vol. 42, pp. 141-151.

Braz C.E., Silva N.C., Macacini J.F., Ribeiro F.B., Taddei M.H.T., 2002, "Determinação dos parâmetros necessários para avaliação do impacto ambiental decorrente da utilização do fosfogesso", Research Development Braz. J., vol. 4, n. 3, pp. 1204 -1209.

Hartley J.N., Freeman H.D., 1986, "Radon Flux Measurements on Gardinier and Royster Phosphogypsum Piles near Tampa and Mulberry, Florida", EPA 520/5-85-029, US Environmental Protection Agency, Office of Radiation Programs, 38 p.

Kaviany M., 1994, "Principles of Heat Transfer in Porous Media", Springer, New York, USA.

Kays W.M., Crawford M.E., 1993, "Convective Heat and Mass Transfer", McGraw-Hill, New York, USA.

Loureiro C.O., 1987, "Simulation of the steady-state transport of radon from soil into houses with basements under constant negative pressure", Ph.D. thesis, Environmental Health Sciences, University of Michigan, Ann Arbor, USA.

May A., Sweeney J.W., 1982, "Assessment of Environmental Impacts Associated with Phosphogypsum in Florida", Report of Investigations 8639, US Department of the Interior, Bureau of Mines, 19 p.

Mazzilli B., Palmiro V., Saucia C., Nisti M.B., 2000, "Radiochemical characterization of Brazilian phosphogypsum", J. Environmental Radioactivity, vol. 49, pp. 113-122.

Mohamad A.A., 2003, "Heat transfer enhancements in heat exchangers fitted with porous media Part I: constant wall temperature", Int. J. Thermal Sciences, vol. 42, pp. 385-395.

Mohamad A.A., Bennacer R., 2001, "Natural convection in a confined saturated porous medium with horizontal temperature and vertical solutal gradients", Int. J. Thermal Sciences, vol. 40, pp. 82-93.

Mohamad A.A., Bennacer R., 2002, "Double diffusion, natural convection in an enclosure filled with saturated porous medium subjected to cross gradients; stably stratified fluid", Int. J. Heat Mass Transfer, vol. 45, pp. 3725-3740.

Nazaroff W.W., Moed B.A., Sextro, R.G., 1988, "Soil as a source of indoor radon: generation, migration and entry". IN: Nazaroff W.W., Nero A.V. (eds.), "Radon and its Decay Products in Indoor Air", John Wiley & Sons, New York, USA.

Nero A.V., 1988, "Radon and its decay products in indoor air: an overview". IN: Nazaroff W.W., Nero A.V. (eds.), "Radon and its Decay Products in Indoor Air", John Wiley & Sons, New York, USA.

Nield D.A., Bejan A., 1999, "Convection in Porous Medium", Springer, New York, USA.

Nikezic D., Urosevic V., 1998, "A theoretical study of radon measurements with activated charcoal", Nuclear Instr. Meth. Phys. Research A, vol. 406, pp. 486-498.

Piliposian G.T., Applebly P.G., 2003, "A simple model for the origin and transport of ^{222}Rn and ^{210}Pb in the atmosphere", Continuum Mech. Thermodyn., vol. 15, pp. 503-518.

Rabi J.A., Mohamad A.A., 2004, "A parametric approach for the prediction of ^{222}Rn exhalation rates from phosphogypsum-based embankments", Proc. 2nd ICAPM – Int. Conf. Applications of Porous Media, Evora, Portugal, May 24-27.

Rabi J.A., Silva N.C., 2002, "Prediction of Rn-222 exhalation rates from phosphogypsum-based embankments. Part I: physical-mathematical modeling", Research Development Braz. J., vol. 4, n. 3, pp. 672 – 676.

Rabi J.A., Silva N.C., 2003, "Rn-222 exhalation rates from phosphogypsum embankments subjected to fixed external activity", Proc. 18th ICTT – Int. Conf. Transport Theory, Rio de Janeiro, Brazil, July 20-25.

Rutherford P.M., Dudas M.J., Arocena J.M., 1995, "Radon emanation coefficients for phosphogypsum", Health Physics, vol. 69, pp. 513-520.

Rutherford P.M., Dudas M.J., Samek R.A., 1994, "Environmental impacts of phosphogypsum", Science of the Total Environment, vol. 149, pp. 1-38.

Stranden E., 1988, "Building materials as a source of indoor radon", IN: Nazaroff W.W., Nero A.V. (eds.), "Radon and its Decay Products in Indoor Air", John Wiley & Sons, New York, USA.

UNSCEAR – United Nations Scientific Committee on the Effects of Atomic Radiation, 2000, "Sources and Effects of Ionizing Radiation", New York, U.N.

U.S. Government, 1998, Code of Federal Regulations, title 40, v. 7, pt. 61.202, 61.204 (40CFR61.202, 40CFR61.204).

Yu C., Loureiro C., Cheng J.J., Jones L.G., Wang Y.Y., Chia Y.P., Faillace E., 1993, "Data Collection Handbook to Support Modeling Impacts of Radioactive Materials in Soil", Environmental Assessment and Information Sciences Division, Argonne National Laboratory, Argonne, USA.