

PHOSPHOGYPSUM AS AN ALTERNATIVE BUILDING MATERIAL: PRELIMINARY MODELING AND SIMULATION OF RADON-222 EXHALATION FROM BLOCKS AND INDOOR ACCUMULATION

José Antonio Rabi

Faculty of Animal Science and Food Engineering, University of Sao Paulo – FZEA / USP
Av. Duque de Caxias Norte, 225, 13635-900, Pirassununga, SP, Brazil
jrabi@fzea.usp.br

Nivaldo Carlos da Silva

Poços de Caldas Laboratory, Brazilian Commission on Nuclear Energy – LAPOC / CNEN
Rodovia Poços de Caldas - Andradas, km 13, CP 913, 37701-970, Poços de Caldas, MG, Brazil
ncsilva@cnen.gov.br

Abstract. *Phosphogypsum is a by-product from the phosphate fertilizer industry and its large-scale utilization as an alternative construction material copes with radiological issues related to radon-222. Zero-order models for radon-222 exhalation from phosphogypsum building blocks and its time-varying accumulation in closed domains (e.g. indoor accumulation) presume homogeneous distribution of radon-222 throughout the enclosure. Having in mind radiological protection design, exhalation characterization of a block sample is a valuable parameter for the corresponding building performance simulation and it can be accomplished by placing a test block inside a test chamber together with a suitable nuclear detector (their relative positioning depends on the chamber geometry). As breakdown of the uniform concentration hypothesis is likely to occur, this preliminary work numerically investigates such model oversimplification. Along with emanation and decay processes, the present mathematical model assumes time-dependent two-dimensional diffusion-dominant mass transfer in a domain containing a sample of porous material, namely the phosphogypsum block of finite thickness. Conversely, as the test chamber is quite small and air-tight closed, convective mass transfer is neglected. Numerically simulated results have confirmed that a non-uniform radon-222 distribution takes place, which can obviously influence the position of the nuclear detector (or its primary element), thus affecting its readings.*

Keywords. *modeling and simulation, alternative building material, phosphogypsum, radon-222 exhalation, mass transfer*

1. Introduction

Phosphogypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) is a by-product from phosphate fertilizer industries and its large-scale commercial exploitation might point to a surrogate building material (Mazzilli and Saueia, 1999). Yet, potential radiological impacts related to ^{222}Rn and decay products should be properly controlled and overcome. Such gaseous radionuclide results from the α -decay of ^{226}Ra , an impurity commonly found in phosphogypsum. Hence, ^{226}Ra decays to ^{222}Rn , which is able to percolate the porous matrix, reach up indoor air and be inhaled by occupants. Accordingly, a reliable model for ^{222}Rn exhalation into indoor air can be useful to assess radiation exposure as well as to devise radiological protection based on radon-induced human health risks.

As many concurrent processes are entailed, comprehensive knowledge of ^{222}Rn exhalation from phosphogypsum-bearing materials and its indoor accumulation is likely to rely on numerical simulation. Diffusive-convective models for ^{222}Rn transport within porous media have basically assumed pressure-driven interstitial air flow following Darcy's law (Edwards and Bates, 1980; Loureiro, 1987; Nazaroff, 1992; Yu *et al.*, 1993; Andersen, 2000) while others have also considered pressure fluctuations (Riley *et al.*, 1999) and buoyant effects (Rabi and Mohamad, 2005).

Although sources from soil or building materials are included, time-varying models for ^{222}Rn accumulation in indoor air or building substructures usually provide bulk values (Stranden, 1988; Nazaroff and Teichman, 1990; Jang, *et al.*, 2005). In other words, they are zero-order models with respect to spatial coordinates so that at any instant ^{222}Rn activity concentration is presumably uniform throughout, i.e., it is a representative value for the entire enclosure.

Having in mind spatial variation, an existing simulator has been continuously adapted to provide time-varying ^{222}Rn concentration (Rabi and Mohamad, 2005). In a previous work (Rabi and Silva, 2006), it was adjusted in order to simulate time-varying two-dimensional ^{222}Rn distribution in a square closed chamber with a very thin phosphogypsum housing panel placed tightly against one wall. In the present work, a rather thick phosphogypsum building block is considered instead so that the "mathematical role" of the ^{222}Rn exhaling material is changed accordingly.

2. Mathematical modeling

As a numerical test case, a rectangular phosphogypsum-bearing building block is placed inside a square closed chamber tightly against one vertical wall, as sketched in Fig. 1. Block horizontal and vertical dimensions are L_x and L_y whereas H is the characteristic length of the chamber.

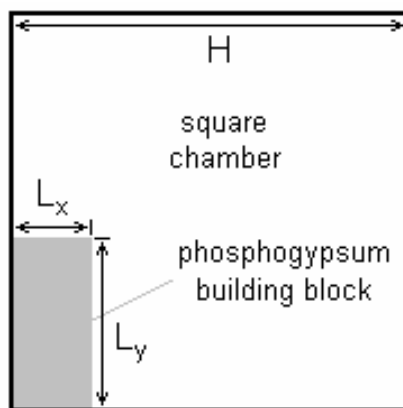


Figure 1. Sketch of a rectangular phosphogypsum building block inside a square closed chamber.

The porous material (i.e., the block) is assumed to have constant porosity ϵ and a homogeneous distribution of ^{226}Ra particles, which renders a uniform generation rate G of ^{222}Rn activity into pore (interstitial) volume. Accounting only for mobile ^{222}Rn activity, the corresponding source term per unit of representative elementary volume (REV) can thus be expressed as $\tilde{G} = \epsilon G$. Additionally, the block is allegedly dry and grain sorption of ^{222}Rn is neglected so that the so-called partition-corrected porosity reduces to the ordinary porosity ϵ (Andersen, 2000).

Bearing in mind that the block is placed in a rather small air-tight closed test chamber at room temperature, thermal effects and air movement (i.e., convective transfer) are both disregarded. Should an entire air-exposed phosphogypsum wall be investigated (instead of a single test block inside a small chamber), continuity, momentum and energy equations (possibly coupled to each other through Boussinesq's approach) would be certainly included in the model framework and numerically solved (Rabi and Mohamad, 2006).

Radon-222 diffusivity in open air D_0 and the corresponding REV value \tilde{D} are assumed to be constant. It is worth noting that such bulk diffusivity \tilde{D} should be used whenever ^{222}Rn flux (in Fick's law) refers to the geometric cross-sectional area of the porous matrix and in this work it is conveniently expressed as:

$$\tilde{D} = \delta D_0 \quad \Leftrightarrow \quad \delta = \tilde{D} / D_0 \quad (1)$$

Recalling that ^{222}Rn sources exist only in the block while ^{222}Rn self-decay (sink) occurs everywhere (but it should be properly corrected to the interstitial volume content inside the block), the time-varying two-dimensional air-borne ^{222}Rn activity concentration $c = c(x,y,t)$ is governed by the following differential equation:

$$\epsilon^n \frac{\partial c}{\partial t} = \delta^n D_0 \left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} \right) + n \tilde{G} - \epsilon^n \lambda c \quad (2)$$

where x and y are Cartesian coordinates, t is time and λ is ^{222}Rn decay constant. The above equation is an extension to the one proposed in (Andersen, 2000) so that the dimensionless parameter n is introduced in order to denote whether the diffusive-dominant ^{222}Rn transfer occurs outside ($n = 0$) or inside ($n = 1$) the porous matrix (Mohamad, 2003).

As an attempt to encompass simultaneous effects of entailed parameters, the previous differential equation is cast in dimensionless form. In view of that, the following set of dimensionless variables is put forward:

$$X = \frac{x}{H} \quad , \quad Y = \frac{y}{H} \quad , \quad \tau = \lambda t \quad , \quad \phi = \frac{c - c_0}{\Delta c} \quad (3)$$

With respect to ^{222}Rn activity concentration, Δc is a scaling factor suitably defined according to the problem physics while c_0 is some reference value. Moreover, if ϕ_0 corresponds to the ϕ value when $c = 0$ then clearly:

$$\phi_0 = -\frac{c_0}{\Delta c} \quad (4)$$

It can be shown that Eq. (2) can be cast into the following dimensionless form:

$$\varepsilon^n \frac{\partial \phi}{\partial \tau} = \frac{\delta^n}{R} \left(\frac{\partial^2 \phi}{\partial X^2} + \frac{\partial^2 \phi}{\partial Y^2} \right) + nM - \varepsilon^n (\phi - \phi_0) \quad (5)$$

or, equivalently,

$$\frac{\partial \phi}{\partial \tau} = \frac{(\delta/\varepsilon)^n}{R} \left(\frac{\partial^2 \phi}{\partial X^2} + \frac{\partial^2 \phi}{\partial Y^2} \right) + \frac{n}{\varepsilon^n} M - (\phi - \phi_0) \quad (6)$$

Accordingly, the following dimensionless groups can be defined:

$$R = \frac{\lambda H^2}{D_o} \quad , \quad S = \frac{\tilde{G} H^2}{D_o \Delta c} \quad , \quad M = \frac{S}{R} = \frac{\tilde{G}}{\lambda \Delta c} \quad (7)$$

Parameters R and S measure the relative importance of decay and of emanation, respectively, in relation to diffusion, whereas the surrogate group $M = S/R$ is interpreted as an emanation-to-decay ratio (Rabi and Mohamad, 2005). In addition, with respect to the chamber characteristic length H , phosphogypsum brick dimensions along the Cartesian coordinates can be expressed respectively as $X_p = L_x/H$ and $Y_p = L_y/H$.

Boundary conditions for the present test case are simply ^{222}Rn no-flux at all impermeable chamber walls, which are expressed as $\partial\phi/\partial X = 0$ (at $Y = 0$ and $Y = 1$) and $\partial\phi/\partial Y = 0$ (at $X = 0$ and $X = 1$). If the reference value c_0 is ^{222}Rn initial activity concentration in the chamber ($c = c_0$ at $t = 0$), then Eq. (3) straightforwardly sets $\phi = 0$ as the initial condition for the dimensionless concentration in the entire domain. For the sake of simplicity, the initial concentration is taken as $c_0 = 0$ (i.e., the chamber initially has no ^{222}Rn activity) so that $\phi_0 = 0$ in accordance to Eq. (4). In Eq. (5) or (6), it is worth bearing in mind that $n = 1$ as long as $0 < X \leq X_p$ and $0 < Y \leq Y_p$; otherwise $n = 0$.

Typical values include $\lambda = 2.098 \times 10^{-6} \text{ s}^{-1}$ and $D_o \cong 1.1 \times 10^{-5} \text{ m}^2 \cdot \text{s}^{-1}$ respectively for ^{222}Rn decay constant and its diffusivity in open air (Yu *et al.*, 1993; UNSCEAR, 2000). As estimates of ^{222}Rn diffusivity in soil point to $\tilde{D} \sim 10^{-6} \text{ m}^2 \cdot \text{s}^{-1}$ (Andersen, 2000; UNSCEAR, 2000), a tentative value $\delta = \tilde{D}/D_o = 0.09$ is here adopted and the same rationale applies for the block porosity, which is fixed at $\varepsilon = 0.5$. For the present test case, physical dimensions were set as $H = 0.5 \text{ m}$, $L_x = 0.1 \text{ m}$ and $L_y = 0.2 \text{ m}$, thus $X_p = 0.2$ and $Y_p = 0.4$. From the previous values, the decay-to-diffusion ratio results $R \cong 0.05$. For convenience (and since Δc is a ^{222}Rn activity concentration scaling factor that can be suitably set in accordance to the investigated scenario), the emanation-to-decay ratio is arbitrarily fixed at $M = 1$ so that Eq. (7) readily establishes $\Delta c = \tilde{G}/\lambda$. As discussed ahead, one is able to assess the generation rate $\tilde{G} = \lambda \Delta c$ by employing Δc values as determined from matching experimental and numerical data.

3. Numerical results and discussion

Numerical solutions were obtained by using an existing finite-volume Fortran program that has been continuously adapted to provide ^{222}Rn distributions. The original simulator has been successfully employed to solve heat and mass transfer problems in media partially or fully filled with porous material and details about it might be found elsewhere (Mohamad, 2003). In the present work, the simulator was particularly adjusted to predict ^{222}Rn accumulation inside the enclosure previously depicted in Fig. 1.

For this preliminary numerical investigation, uniform orthogonal grids were used. Compromise between spatial grid coarseness and numerical accuracy was checked by solving a steady-state simplification of Eq. (5) or (6), namely:

$$\frac{\delta^n}{R} \left(\frac{\partial^2 \phi}{\partial X^2} + \frac{\partial^2 \phi}{\partial Y^2} \right) + nM - \varepsilon^n \phi = 0 \quad (8)$$

or, equivalently,

$$\frac{(\delta/\varepsilon)^n}{R} \left(\frac{\partial^2 \phi}{\partial X^2} + \frac{\partial^2 \phi}{\partial Y^2} \right) + \frac{n}{\varepsilon^n} M - \phi = 0 \quad (9)$$

subjected to the same boundary conditions discussed in the previous section. Figure 2 shows the evolution of simulated values for ϕ at the enclosure midpoint $X = Y = 0.5$ (calculated by interpolation from neighboring values) for increasing number of grid points (including grid points at boundaries), using $\delta = 0.09$, $\varepsilon = 0.5$, $R = 0.05$, $M = 1$, $X_p = 0.2$ and $Y_p =$

0.4. Based on the results obtained, the 100×100 mesh was chosen for further time-varying simulations (dimensionless uniform grid spacing therefore results $\delta X = \delta Y = 0.01$).

In order to set the advancing time step $\delta\tau$ for time-varying simulations, the complete equation, Eq. (5) or (6), was solved at the 100×100 mesh for $0 \leq \tau \leq 1$ (i.e., approximately up to 5.5 days) for diminishing time steps $\delta\tau$. The remaining parameters were held the same (i.e., $\delta = 0.09$, $\varepsilon = 0.5$, $R = 0.05$, $M = 1$, $X_p = 0.2$ and $Y_p = 0.4$). Figure 3 presents obtained values for ϕ at the final instant $\tau = 1$ and at the enclosure midpoint. Having in mind a compromise between numerical accuracy and computational effort, $\delta\tau = 0.001$ was then chosen and results presented hereafter are based on such advancing dimensionless time step.

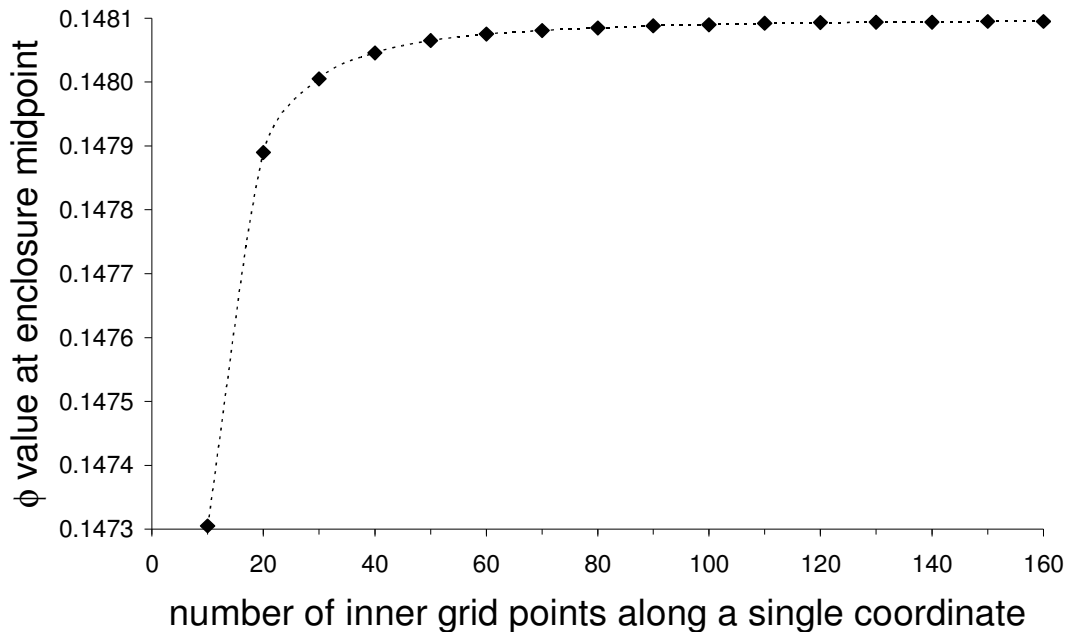


Figure 2. Simulated values for ϕ at the enclosure midpoint for increasing number of grid points.

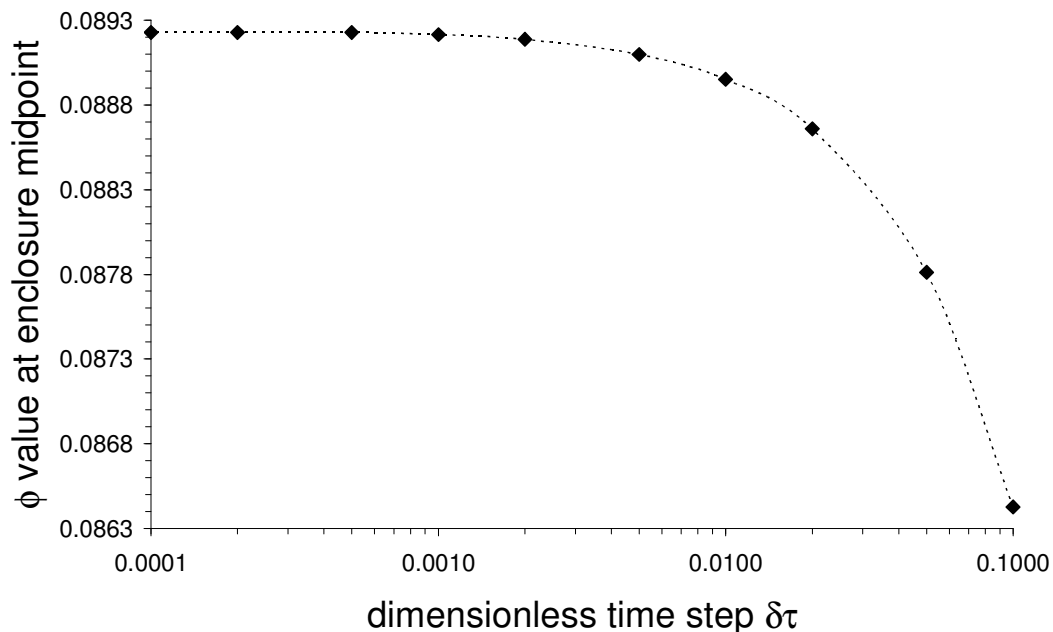


Figure 3. Simulated values for ϕ at the enclosure midpoint and at $\tau = 1$ for distinct time steps.

Accordingly, Figs. 4(a) to 4(d) show dimensionless concentration iso-lines related to the following dimensionless time instants: $\tau = 0.25$, 0.50 , 0.75 and 1.00 (i.e., roughly after 33 h, 66 h, 99 h and 132 h, respectively). The

phosphogypsum block limits are represented as dashed lines. Due to the considerably lower diffusivity inside the porous matrix ($\delta = 0.09$), higher ^{222}Rn concentrations are particularly observed around the lower left corner of the block, i.e., the one in contact with the chamber corner.

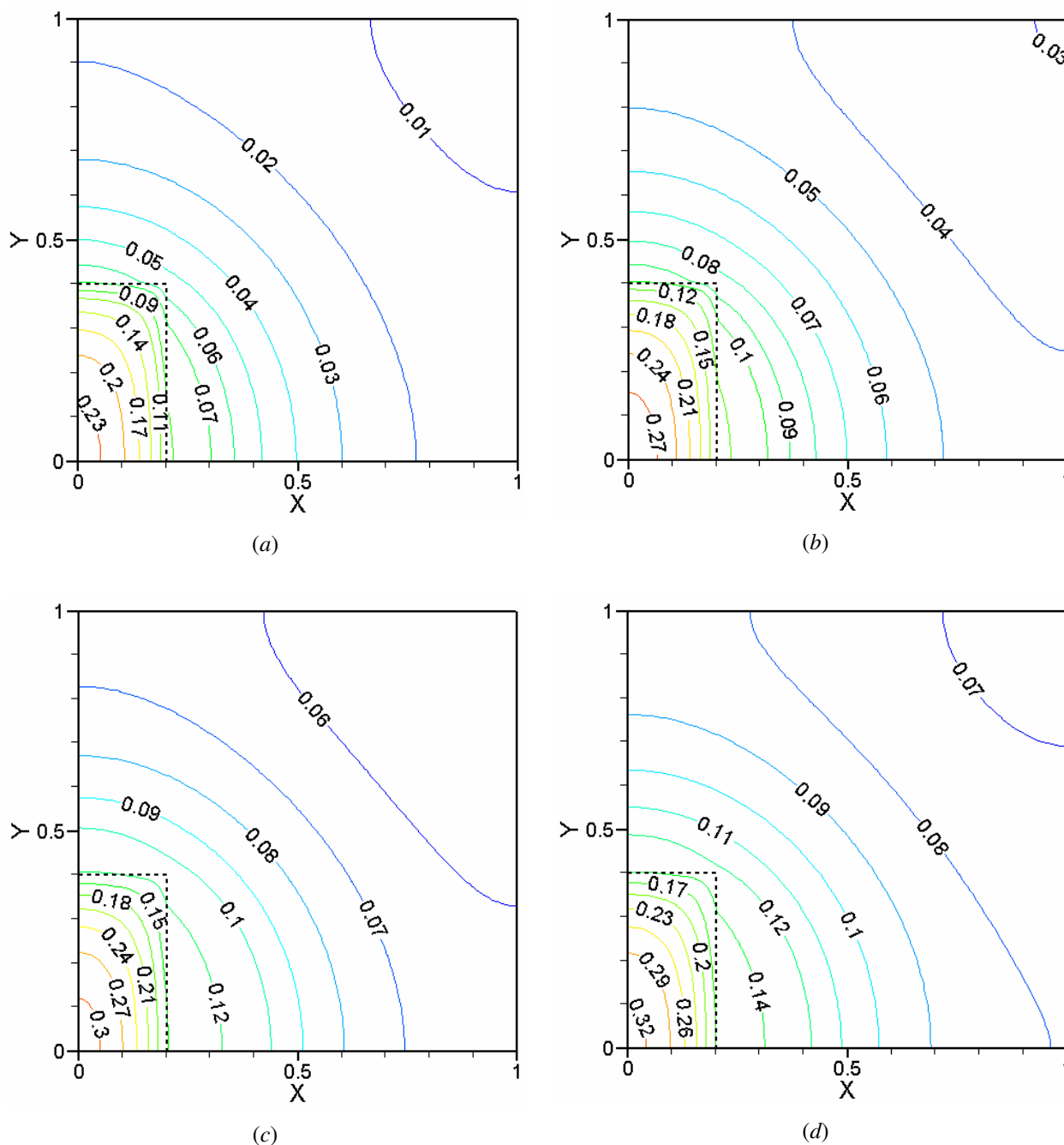


Figure 4. Isoconcentration lines numerically simulated for (a) $\tau = 0.25$, (b) $\tau = 0.50$, (c) $\tau = 0.75$ and (d) $\tau = 1.00$.

Higher concentration levels are observed as time increases but distributions at distinct time instants seem to retain a common pattern. Outside the block, concentrations decrease toward the opposite upper right corner of the chamber and it seems to reduce in a way that the block behaves somehow similar to a point source. Apart from the null initial condition ($\phi = 0$ all over the domain at $\tau = 0$), no homogeneous distribution is verified whatsoever at any further instant during the time interval adopted in the present numerical investigation ($0 \leq \tau \leq 1$). Considering experimental tests, additional precautions concerning the position of the detector (or its primary element) should then be exercised.

This is particularly true if no mixing-up device is introduced within the chamber and it might become decisive for “static” detectors that are simply placed “resting” inside the chamber and later removed for readings. A typical and quite simple device that falls into this category is the so-called SSNTD – solid state nuclear track detector. On one hand, SSNTD is small enough to allow one to treat it as point in the solution domain; on the other hand, its counting system is not automatic and requires human involvement.

A more sophisticated device to measure ^{222}Rn concentration is an active ionizing chamber (e.g. Alfa Guard™ system), which operates under two modes: diffusive or active. In the former, the detection system itself is placed inside the test chamber and ^{222}Rn particles are driven into it solely by mass diffusion. Because the system physical dimensions are significantly large, symmetry breakdown is likely to arise in the solution domain. In the later operation mode, ^{222}Rn particles are forced into the system by means of negative gage pressures, which obviously induce considerable air movement within the test chamber. Hence, if convective transport is to be accounted for, bulk fluid continuity and momentum equations must be included in the model framework, thus increasing problem stiffness.

The later rationale equally applies to air-borne ^{222}Rn accumulation in real rooms with doors and windows, which evidently evokes the abovementioned governing differential equations. Conversely for this scenario, solution domain dimensions are much larger than those of the active ionizing chamber so that its utilization under the assumption that it causes no interference (e.g. symmetry failure or relatively strong air currents) to its surroundings is quite reliable.

Yet, if one compares ^{222}Rn concentrations, for instance, at time $\tau = 0.25$ and positions $(X, Y) = (0.3, 0.1)$ and $(X, Y) = (0.8, 0.7)$, there is roughly a factor 7 between observed levels at these two points. On the other hand, comparing concentration levels at the same points at $\tau = 1$, such factor drops to 2. This result might suggest that heterogeneity of ^{222}Rn concentration could possibly decrease with time, which might thus demand for longer simulation times. Such is an issue for further investigation, particularly if experimental tests should be carried out for a lengthy period.

As suggested by Eq. (3), one may assess the scaling factor $\Delta c_{\text{assessed}}$ for ^{222}Rn activity from numerically simulated values for $\phi(X, Y, \tau) = \phi_{\text{numerical}}$ at a pre-defined position (e.g., chamber midpoint) as well as from their experimentally measured counterparts $c(x, y, t) = c_{\text{experimental}}$. Indeed, manipulating Eq. (3), one obtains:

$$\phi = \frac{c - c_0}{\Delta c} \Rightarrow \Delta c_{\text{assessed}} = \frac{c_{\text{experimental}} - c_0}{\phi_{\text{numerical}}} \quad (10)$$

It is worth recalling that $c_0 = 0$ was assumed in the present work. For $c_0 \neq 0$, such non-zero initial concentration inside the chamber should be experimentally measured in order to be introduced in the previous equation. Besides, as long as $\Delta c = \tilde{G}/\lambda$ as imposed by Eq. (7) after conveniently choosing $M = 1$, one may assess the generation rate $\tilde{G}_{\text{assessed}}$ by employing $\Delta c_{\text{assessed}}$ values provided by Eq. (10) and simply rearranging Eq. (7) as:

$$M = \frac{\tilde{G}}{\lambda \Delta c} \xrightarrow{M=1} \tilde{G}_{\text{assessed}} = \lambda \Delta c_{\text{assessed}} \quad (11)$$

It should be noted that the generation rate \tilde{G} is a desired parameter for radiological protection design.

4. Concluding remarks

Along with its short-lived decay products, ^{222}Rn responds to most of human exposure to radiation from natural sources. Therefore, ^{222}Rn emanation and indoor accumulation are important matters to be accounted for as far as the use of phosphogypsum as an alternative building material is concerned. Although radiological impact assessment claims for comprehensive knowledge of ^{222}Rn release and transport, zero-order models with respect to spatial coordinates can only provide volume-averaged ^{222}Rn concentration levels.

In the present work, such homogeneous distribution constraint was relaxed so as to allow point-to-point variation. Including emanation and decay processes, this work put forward a time-dependent two-dimensional diffusion-dominant model for ^{222}Rn transport within a square chamber containing a rectangular phosphogypsum building block of finite thickness. With the help of an existing finite-volume simulator, numerical investigation of such scenario showed that no homogeneous distribution could be verified indeed during the considered time interval, which might request additional precautions concerning the position of the detector (or its primary element).

It should be stressed that the proposed model framework is somewhat preliminary. Assuming that the test chamber was small and air-tight closed, air movement (convective mass transfer) was neglected. Moreover, thermal effects could also occur so that, accounting for these two extensions, bulk fluid continuity, momentum and energy equations should be simultaneously solved. Along with thermal conditions (e.g. prescribed wall temperature or heat flux), boundary conditions should then include known velocity profiles or conditions at all openings as well as no-slip condition at solid walls. As moisture might exist to some extent inside phosphogypsum building blocks, future investigations point to analyzing effects due to ^{222}Rn partitioning between interstitial air and water. By the same token, further edge effects might be analyzed by extending both the porous matrix and the solution domain up to three dimensions.

5. References

Bordalo, S. N., Ferziger, J. H. and Kline, S. J., 1989, "The Development of Zonal Models for Turbulence", Proceedings of the 10th Brazilian Congress of Mechanical Engineering, Vol.1, Rio de Janeiro, Brazil, pp. 41-44.

- Coimbra, A. L., 1978, "Lessons of Continuum Mechanics", Ed. Edgard Blücher, S.Paulo, Brazil, 428 p.
- Andersen, C. E., 2000, "Radon transport modelling: user's guide to RnMod3d", Riso-R-1201(EN), Riso National Laboratory, Roskilde, Denmark.
- Edwards, J. C. and Bates, R. C., 1980, "Theoretical evaluation of radon emanation under a variety of conditions", *Health Physics*, vol. 39, pp. 263 -274.
- Jang, M., Kang, C.-S. and Moon, J. H., 2005, "Estimation of ^{222}Rn release from the phosphogypsum board used in housing panels", *Journal of Environmental Radioactivity*, vol. 80, pp. 153-160.
- 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, University of Michigan, Ann Arbor, USA.
- Mazzilli, B. and Saueia, C., 1999, "Radiological implications of using phosphogypsum as a building material in Brazil", *Radiation Protection Dosimetry*, vol. 86, pp. 63-67.
- Mohamad, A. A., 2003, "Heat transfer enhancements in heat exchangers fitted with porous media. Part I: constant wall temperature", *International Journal of Thermal Sciences*, vol. 42, pp. 385-395.
- Nazaroff, W. W., 1992, "Radon transport from soil to air", *Reviews of Geophysics*, vol. 30, pp. 137-160.
- Nazaroff W. W. and Teichman, K., 1990, "Indoor radon: exploring US federal policy for controlling human exposures", *Environmental Science and Technology*, vol. 24, pp. 774-782.
- Rabi, J. A. and Mohamad, A. A., 2005, "Radon-222 exhalation rates from phosphogypsum-bearing embankment subjected to constant temperature and fixed activity concentration", *Journal of Porous Media*, vol. 8, pp. 175-191.
- Rabi, J. A. and Mohamad, A. A., 2006, "Parametric modelling and numerical simulation of natural-convective transport of radon-222 from a phosphogypsum stack into open air", *Applied Mathematical Modelling*, vol. 30, pp.1546-1560.
- Rabi, J. A. and Silva, N. C., 2006, "Radon exhalation from phosphogypsum building boards: symmetry constraints, impermeable boundary conditions and numerical simulation of a test case", *Journal of Environmental Radioactivity*, vol. 86, pp. 164-175.
- Riley, W. J., Robinson, A. L., Gadgil, A. J. and Nazaroff, W. W., 1999, "Effects of variable wind speed and direction on radon transport from soil into buildings: model development and exploratory results", *Atmospheric Environment*, vol. 33, pp. 2157-2168.
- Stranden, E., 1988, "Building materials as a source of indoor radon", In: Nazaroff, W. W. and Nero, A. V. (editors), "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.
- Yu, C., Loureiro, C., Cheng, J. J., Jones, L. G., Wang, Y. Y., Chia, Y. P. and Faillace, E., 1993, Data collection handbook to support modeling impacts of radioactive materials in soil, Argonne National Laboratory, Argonne, USA.

6. Copyright Notice

The authors are the only responsible for the printed material included in this paper.