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PREDICTION OF RADON CONCENTRATIONS AT CONCRETING IN CONSTRUCTION SITES WITH LIMITED VENTILATION

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INTRODUCTION

It is known that radon exhalation rate can achieve extremely high values during cement hydration. Radon contained in trace amounts in hardening material, as other inert gases, does not interact with the surrounding media chemically, and hence does not interfere with chemical reactions accompanying cement hydration. Therefore, the unique properties of radon as a noble gas can be used for monitoring cement hydration and microstructural transformations in cementitious system.

The first attempt to measure radon exhalation in hardening cementitious materials and use radon as an indicator of microstructural changes occurring during cement hydration was made in Czechoslovakia by Balek and Donhálek (1983). Balek and Donhálek attributed the changes in radon exhalation rate, mainly, to the changes in surface area and microporosity occurring in the hardening material during hydration. The authors suggested labeling the sample of cement powder before hydration by the parent isotopes of radon. They impregnated cement powder with a non-aqueous (acetone) solution containing ^{228}Th or ^{224}Ra in the concentration of 1011 Bq m^{-3}. The radionuclides were adsorbed on the surfaces of cement particles and the non-aqueous solution was evaporated. The specific radioactivity of the labeled cement was 106 – 107 Bq kg^{-1}, i.e. higher by 5-6 orders of magnitude than that before labeling, therefore the sensitivity of the method was tremendously enhanced.

The disadvantage of the labeling is that working with very high radioactivity concentrations requires specific safety precautions. In addition, labeling cement particles for a certain depth, which is independent of cement particle size, leads to uncertainty with the interpretation of the results, especially when the effect of cement fineness is studied.

The more detailed review of the publications on using radon exhalation method for monitoring cement hydration and microstructural transformations in hardening cementitious materials is available in the State-of-the-Art Report of RILEM (International Union of Laboratories and Experts in Construction Materials, Systems, and Structures) Technical Committee TC-185 ATC "Advanced Testing of Cement-Based Materials during Setting and Hardening" (Kovler, 2005).

Potentially, high radon exhalation rates can be a concern for construction workers employed on daily basis at concreting in sites with limited ventilation. The goal of the present work was to measure the kinetics of radon exhalation rate during Portland cement hydration in the laboratory and to estimate generated radon concentrations, which can develop in construction sites with different ventilation rates.

METHODOLOGY

In general, radon concentration *C(t)* (Bq m^{-3}) generated in the given time *t* (s) in the hermetically closed chamber can be determined as

$$C(t) = C_i e^{-\lambda_{eq} t} + \frac{ES + qC_o}{V_{eq} \cdot \lambda_{eq}} (1 - e^{-\lambda_{eq} t}) \tag{1}$$

where

C_i - initial radon concentration in the chamber (Bq m^{-3});
C_0) - background radon concentration in the room (Bq m^{-3});
E - radon exhalation rate of the source, per unit surface area or mass (Bq m^{-2} s^{-1} or Bq kg^{-1} s^{-1});
S - characteristic of the radon source, which can be either mass M (kg) of the specimen in the test chamber or its surface area A (m^2);
V_{eq} - equivalent volume occupied by air in the test chamber (volume of the chamber plus volume of the circuit pipes of the radon monitor instrument connected to the chamber minus volume of the specimen, m^3);
q - radon leakage (m^3 s^{-1}). Note: in a hermitically closed chamber, this term is zero.
λ_{eq} – equivalent radon decay constant (s^{-1}), which is a sum of radon decay constant λ_{Rn} ($2.1 \cdot 10^{-6}$ s^{-1}), back diffusion coefficient λ_{bd} (s^{-1}) and ventilation rate λ_{vent} (s^{-1}):

$$\lambda_{eq} = \lambda_{Rn} + \lambda_{bd} + \lambda_{vent} \tag{2}$$

We have to emphasize that the concentration is not a characteristic of the material, because it increases with time until achieving a constant value C_∞ corresponding to the saturation condition. At infinite time $t \to \infty$, assuming that $C_0 = C_i = 0$, we obtain that maximum radon concentration in the hermetically closed space under saturation C_∞ (Bq m^{-3}) is:

$$C_\infty = \frac{ES}{V_{eq} \cdot \lambda_{eq}} \tag{3}$$

Radon concentration *C(t)* at a given time is related to maximum radon concentration C_∞ as follows:

$$C(t) = C_\infty (1 - e^{-\lambda_{eq} t}) \tag{4}$$

The method of determination of radon exhalation rate of the building material via radon concentration measured in the hermetically closed chamber has been described by several authors, and in particular, by Kovler et al. (2005). It is based on measuring the radon concentration at a given time $C(t)$ and calculating C_∞, for determination of the radon exhalation rate E:

$$E = C_\infty \lambda_{eq} V_{eq} / S \tag{5}$$

However, the described method is valid only under assumption of constant radon exhalation rate of the sample in time. For materials, which change their properties in time, the radon exhalation rate E is time-dependent, so the formula is modified:

$$E(t) = \frac{V_{eq}}{S}\left[\frac{dC}{dt} + \lambda_{eq} C(t)\right] \tag{6}$$

EXPERIMENTAL RESULTS AND DISCUSSION

Concentration of radium ^{226}Ra in Portland cement used in the current study was 64.2 ± 2 Bq kg^{-1}. It was determined in cement powder sample by gamma-ray spectroscopy, using NaI detector (NPP Doza). The sample was dried at 105°C for 24 hours before testing. Cement was measured as a sample of 200 cm^3 volume in sealed cylindrical polyethylene container after 30 days, to achieve secular equilibrium of the ^{226}Ra progeny.

Cement paste specimens of 5 kg mass were mixed in a pan mixer, cast in the glass desiccator chamber of 6 liter. This experimental arrangement (the specimen mass to volume of the chamber ratio) represented a reasonable compromise to fulfill both requirements: to have acceptably small back diffusion and small experimental scatter (Kovler, 2006). The desiccator was hermetically closed and the radon concentrations generated in the chamber were measured by means of continuous radon gas monitor RAD-7, Durridge Company Inc. Such a small chamber enabled to obtain high radon concentrations within a short time of the test. The duration of each test was a few days.

The radon exhalation of unhydrated cement was compared with that of hydrating cement paste, made with water to cement ratio of 0.33. The radon concentrations were measured at 20°C. In order to compare the measurements data, the relative radon concentrations (per 1 kg of cement) were calculated. These values are presented on a logarithm scale in Figure 1. The low-value range is a characteristic of unhydrated cement. The radon concentration curve for hydrating cement paste starts from zero (background) concentrations, similarly to unhydrated cement. However, the radon concentrations developed at the end of the test in the desiccator containing cement paste were significantly higher than those of unhydrated cement. This fact proves the importance of microstructural transformations taking place in the process of cement hydration, in comparison with the radon emanation from cement particle, which is a time-stable material.

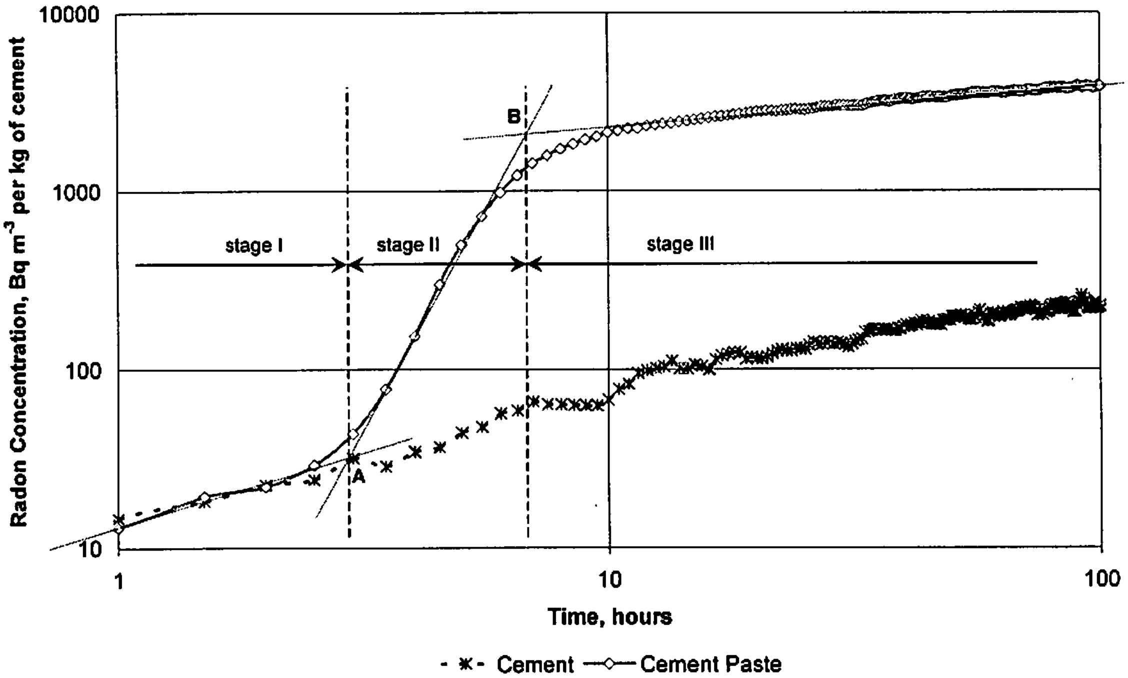

Figure 1 - Dependence of relative radon concentration in the desiccator (in Bq m^{-3} per 1 kg of cement) vs. time for unhydrated cement and hydrating cement paste

To explain the enormous difference in the amount of radon atoms exhaling from the materials containing the same number of radium atoms, we have to take into account several important mechanisms occurring in the course of cement hydration. For this reason, it would be convenient to distinguish between three main stages, which are readily seen in Figure 1 and correspond with stages in cement hydration and microstructural development: stage I (dormant period), stage II responsible for setting, when intensive microstructural transformations occur, and stage III (the densification of the structure and drying).

PREDICTION OF RADON CONCENTRATIONS IN CONSTRUCTION SITE

The dramatic increase of the radon exhalation rate up to the maximum observed in a few hours after mixing with water is one of the main findings reported and analyzed in the previous section. Our experiments show that the maximum value of radon exhalation rate observed for hydrating cement pastes made with different water to cement ratios, with and without different chemical and mineral admixtures, can reach 0.6 and sometimes exceeds 1.0 mBq kg^{-1} s^{-1}. Such extremely high values of radon exhalation rate significantly exceed all *E*-values known from the previous literature dealing with radon exhalation from cementitious materials. Potentially, enormous radon exhalation rate when concrete sets may lead to the development of high radon concentrations in construction sites at closed spaces. This phenomenon may be of radiological concern for construction workers, which are employed in casting concrete in a routine form (by daily or weekly basis), and consequently are exposed to enhanced radon concentrations in work places, especially where ventilation is poor.

Assuming that the time dependence of radon exhalation rate in-situ remains similar to that determined in the laboratory, the radiological consequences of the extremely high values of radon exhalation rate developing a few, hours after mixing cement with water, should be evaluated in terms of predicted radon concentrations for construction site. The result of this analysis is reported hereafter.

The ratio M/V for the real construction conditions is different from those tested in the lab. Let us consider the case of casting concrete floor each square meter of which contains 50 kg of cement paste. The rest of concrete composition are aggregates, entrained and entrapped air and admixtures, which almost do not contribute in radon exhalation of concrete floor. Let us assume that the height of air column above each square meter of concrete floor in the space, where concreting is executed, is 2.5 m. It means that *M/V* ratio for this case is 50/2.5 = 20 kg m^{-3} only. In addition, the space should have some air exchange; that is why the back diffusion can be neglected for the calculations of radon concentrations in situ.

For deriving the expression for radon concentrations developing during concreting in closed spaces, equation (6) is solved as "First order linear differential equation":

$$C(t) = e^{-\lambda_{eq} t} \frac{S}{V_{eq}} \int e^{\lambda_{eq} t} E(t) dt \quad \text{or} \quad C(t) = e^{-\lambda_{eq} t} \frac{M}{V_{eq}} \int e^{\lambda_{eq} t} E(t) dt \qquad (7)$$

The results of the calculation by formula (7), assuming that the radon exhalation rate develops according to the curve obtained in the laboratory (the case of *M/V* = 439 kg m^{-3}), but in the smoothed and approximated form, makes possible to predict the development of radon concentrations on construction site. The curves of radon concentration calculated for different ventilation conditions (from the space closed hermetically to the cases of air exchange rate of 0.5 hr^{-1}) are shown in Figure 2.

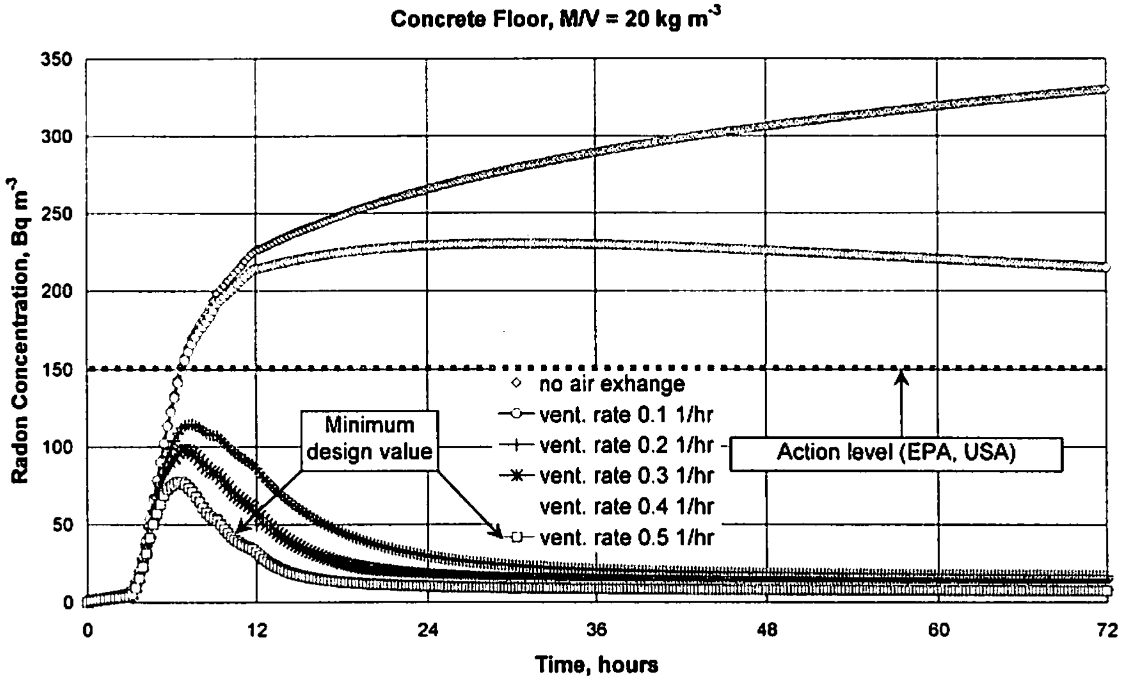

Figure 2 - Predicted radon concentrations at casting concrete floor under different ventilation conditions (hermetically closed space and air exchange rate of 0.1 - 0.5 hr^{-1})

Let us assume that the action level in dwellings is 150 Bq m^{-3}, according to the EPA (USA). This limit is shown by dashed line parallel to the time axis. It can be seen that in the cases of extremely poor ventilation (λ_{vent} = 0.1 hr^{-1} and less) the radon concentrations exceed 150 Bq m^{-3}. Moreover, the peak concentrations can increase, when taking into account the background in the given place. In this case, the curves shown in Figure 2 will shift up. However, from the radiological point of view, such shifting is also not of any concern, because the local maximum of radon concentration is observed only when concrete sets (4-10 hours after casting). Then the concentrations drop down to the level, which depends on the background and usual radon entry rate from hardened building materials and other possible sources.

It can be seen that increasing ventilation a little, from λ_{vent} = 0.1 hr^{-1} to 0.2 hr^{-1} only, improves significantly the situation. The difference in radon concentrations between λ_{vent} = 0.1 hr^{-1} and λ_{vent} = 0.2 hr^{-1} is rather impressive. In any case, the minimum ventilation rate accepted in the design practice is 0.5 hr^{-1}, which guarantees that the concentrations in most cases will not exceed the action level and that they are not of any radiological concern for construction workers employed in concreting in closed spaces.

This conclusion is drawn with a high safety factor. The elevated radon concentrations developing in a few hours after mixing cement with water, are very short-lived, so comparing with an action level which is meant to be applied to average long-term concentrations is fraught with danger. In addition, construction sites are usually very draughty, and have much activity going on which will increase air exchange, so the true values are likely to be on the lower side. Also, the workers move from one construction site to another, so it is unlikely that they are always exposed to the low-air-exchange conditions.

CONCLUSIONS

1. The maximum value of radon exhalation rate observed in hydrating cementitious material can reach 0.6 and sometimes exceeds 1.0 mBq kg^{-1} s^{-1}. These values are extremely high and exceed significantly those known before about cementitious materials, both unhydrated and hardened (well-hydrated).
2. At the same time, the elevated radon concentrations developing in a few hours after mixing cement with water are short-lived. The minimum ventilation rate accepted in the design practice (0.5 hr^{-1}), guarantees that the concentrations expected to develop at concreting in close spaces will not exceed the action level in most of the cases and that they are not of any radiological concern for construction workers employed in routine daily concreting in poorly ventilated sites.

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