Pergamon

Appl. Radiat. Isot. Vol. 48, No. 7, pp. 1003–1007, 1997

Printed in Great Britain
PII: S0969-8043(97)00021-3
0969-8043/97 $17.00 + 0.00

Radon Emanation from Concrete Surfaces and the Effect of the Curing Period, Pulverized Fuel Ash (PFA) Substitution and Age

K. N. YU[1], E. C. M. YOUNG[1], M. J. STOKES[1], M. K. KWAN[1] and R. V. BALENDRAN[2]

[1]Department of Physics and Materials Science, City University of Hong Kong, Tat Chee Avenue, Kowloon, Hong Kong and [2]Department of Building and Construction, City University of Hong Kong, Tat Chee Avenue, Kowloon, Hong Kong

(Received 30 January 1997)

Radon emanation rates from 48 concrete blocks have been monitored throughout a period in excess of 1 yr. These blocks have been produced for three distinct mixes of materials, i.e. mix A with 25% by weight substitution of type A pulverized fuel ash (PFA), mix B with 25% by weight substitution of type B PFA and mix C without PFA. Each mix is represented by four sets of concrete blocks with curing periods of 1, 3, 7 and 28 days. Every set comprises of four blocks each with the same composition and curing period. From the results, rates of change of radon emanation have been identified for two distinct periods, i.e. for the first month following curing and for the period following the first month. The different emanation rates are linked to the role of the superficial and inner pores of the concrete. It is also seen that the radon emanation rate tends to decrease with the age of the blocks for curing periods of 1, 3 and 7 days, but tends to increase for a curing period of 28 days, all of which can be rationalized in terms of the gradual dehydration of concrete as it ages. The patterns are similar for the different mixes, although the exact effect of PFA upon radon emanation rates remains unresolved. © 1997 Elsevier Science Ltd

Introduction

In recent years, interest in radon (by which in this work is meant the radioisotope ^{222}Rn) has become a focus of research in environmental physics and the built environment. The factors affecting the variation of radon emanation from concrete surfaces are important in understanding the basis of radon levels and their mitigation. A recent cross-sectional study of the variation of radon emanation rates from concrete surfaces of buildings in Hong Kong has shown that the emanation rate decreases with the age of a building (Yu *et al.*, 1995). Since the variation of radon emanation rate from concrete and soil has already been suggested to be due to water content (Stranden *et al.*, 1984; Fleischer, 1987), it is natural to assert that the phenomenon observed in the cross-sectional study of buildings is also similarly due to the change of water content of the concrete with ageing (Yu *et al.*, 1995).

Three objectives have been identified for the present investigation. The first is to conduct a long-term longitudinal study of the radon emanation rate from concrete surfaces. The second is to examine the effects of the curing period upon the radon emanation rates from concrete surfaces. The third is to identify whether the inclusion of pulverized fuel ash (PFA) in concrete blocks affects the emanation rates. PFA has been included to the extent of 25 wt%, this being the maximum amount allowed locally for building purposes. PFA is a by-product of the burning of fossil-fuels and therefore by definition contains uranium and thorium and their progeny, depending to a greater or lesser degree upon the source material and the fraction remaining after burning.

Radon Emanation from Pore Systems

Radon-222 is an inert gas with a half life of 3.8 days, and is a radioactive decay product of ^{226}Ra which is present in concrete at natural levels. When a radon atom is formed from radium through α decay, by virtue of its suffering recoil, several possible fates await it within the concrete depending on the position of its formation and the particular characteristic medium through which it traverses. It may, for instance, stop within the originating grain material, traverse to adjacent grains or otherwise stop in the pore system and capillary network of the concrete. Only those atoms which stop in the pore system and capillary network can efficiently leave the concrete by diffusion through the pore system or by transportation within the pore-fluids, generally being air or water. As such, only a fraction of the radon generated will ever exit the building material medium, the fraction of exhalation being characterized by an emanation coefficient. Depending upon the amount of water inside the pore spaces of the material, the radon atom will have a greater or lesser chance of being trapped inside the pore system and ultimately leaving the concrete (Stranden *et al.*, 1984; Fleischer, 1987). However, the greater the water content, the slower will be the diffusion rate of radon in the pore system, and exhalation of radon atoms from the concrete will be inhibited.

Due to the limited half life of the radon atoms, those trapped in the pores near the surface of the concrete are more likely to be exhaled; we refer these pores as the superficial pores. These pores have good contact with the ambient air, so that water loss rates in these pores are expected to be higher. On the other hand, those radon atoms which are trapped more deeply within the pores have a greater likelihood of suffering decay than exhalation from the concrete; we refer to these pores as the inner pores. These pores have poor contact with the ambient air and as a consequence the water loss rates in these pores are expected to be smaller.

Methodology

A total of 48 concrete blocks have been used in present investigations. All of the blocks were formed into a uniform dimension of cube of $15 \times 15 \times 15$ cm. Details of the casting of the concrete blocks are listed in Table 1. These blocks are categorized by mix, namely, concrete mix A with 25% substitution by weight of type A PFA, concrete mix B with 25% substitution by weight of type B PFA and concrete mix C without substitution by PFA. Each mix is represented by four sub-category of concrete blocks, corresponding to curing periods of 1, 3, 7 and 28 days. The concrete blocks were cured at 27°C in a temperature controlled water tank. Each sub-category comprises of four concrete blocks, each with the same composition and each cast under the same conditions. Mixing, casting and commencement of curing for all the concrete blocks was performed on a given day. The zeroth day for each concrete block has been defined to be the day on which the curing period ended. The mix contents of each 1 m^3 batch volume of the three mixes are listed in Table 2.

Table 1. Description of concrete blocks used in present investigations. The date of mixing, casting and commencement of curing for all the concrete blocks was 10 January 1995

Concrete mix	Reference No.	Curing period (days)	Zeroth day
A	A25C1–4	1	11.01.95
	A25C5–8	3	13.01.95
	A25C9–12	7	17.01.95
	A25C13–16	28	07.02.95
B	A25K1–4	1	11.01.95
	A25K5–8	3	13.01.95
	A25K9–12	7	17.01.95
	A25K13–16	28	07.02.95
C	A1–4	1	11.01.95
	A5–8	3	13.01.95
	A9–12	7	17.01.95
	A13–16	28	07.02.95

Table 2. The mix portions for each batch volume of 1 m^3 of the mixes A, B and C

Mix	Cement (kg)	PFA (kg)	Coarse 20 mm (kg)	Coarse 10 mm (kg)	Fines (kg)	Water (kg)
A	265	85	760	390	615	208
B	265	85	760	390	615	208
C	350	0	740	370	670	210

Subsequent to curing, the concrete blocks were removed from the water tank. Since in the present method radon-absorbing activated charcoal canisters (Cohen and Cohen, 1983) were to be affixed to the surfaces, the concrete surfaces were immediately dried using dry cloths to prevent the canisters from adsorbing too much water. The method for measurement of the radon emanation rates from concrete surfaces has been described in Yu *et al.* (1995). Of particular note is the sensitivity of the standardized charcoal canisters, this requires that exhaled radon be collected for about 3 days for accurate measurements. The canisters were sealed against the concrete surfaces using silicone sealer to prevent leakage of air around the edge of the detectors. After collection, the charcoal canisters were removed from the surface, sealed, and stored for in-growth of radon progeny to reach equilibrium. The radon progeny levels retained within the activated charcoal canisters were then determined by counting the gamma ray photons emitted by the radon decay products at energies 295, 352 and 609 keV using a NaI gamma spectrometer and a counting period of 10 min.

Since during a 3 day exposure the number of radon atoms exhaled from a concrete surface within the area enclosed by an activated charcoal canister is far smaller than the number that can be adsorbed onto the canister, an efficiency of 100% can be assumed for the adsorption of the exhaled radon. Under this assumption, the radon emanation rate (in units of Bq m^{-2} s^{-1}) can be calculated using the relation

$$\varepsilon = \frac{\lambda(NET)e^{\lambda t}}{SE(1 - e^{-\lambda T})3600} \qquad (1)$$

where $\lambda(= 0.00756\ h^{-1})$ is the physical decay constant of radon, NET is the net photon count rate (cpm) due to the three gamma photon emissions at energies mentioned above, S is the area of the concrete surface covered by the charcoal canister, E is the detection efficiency of the detector system, T is the radon collection time, t is the time elapsed from the end of collection to the start of

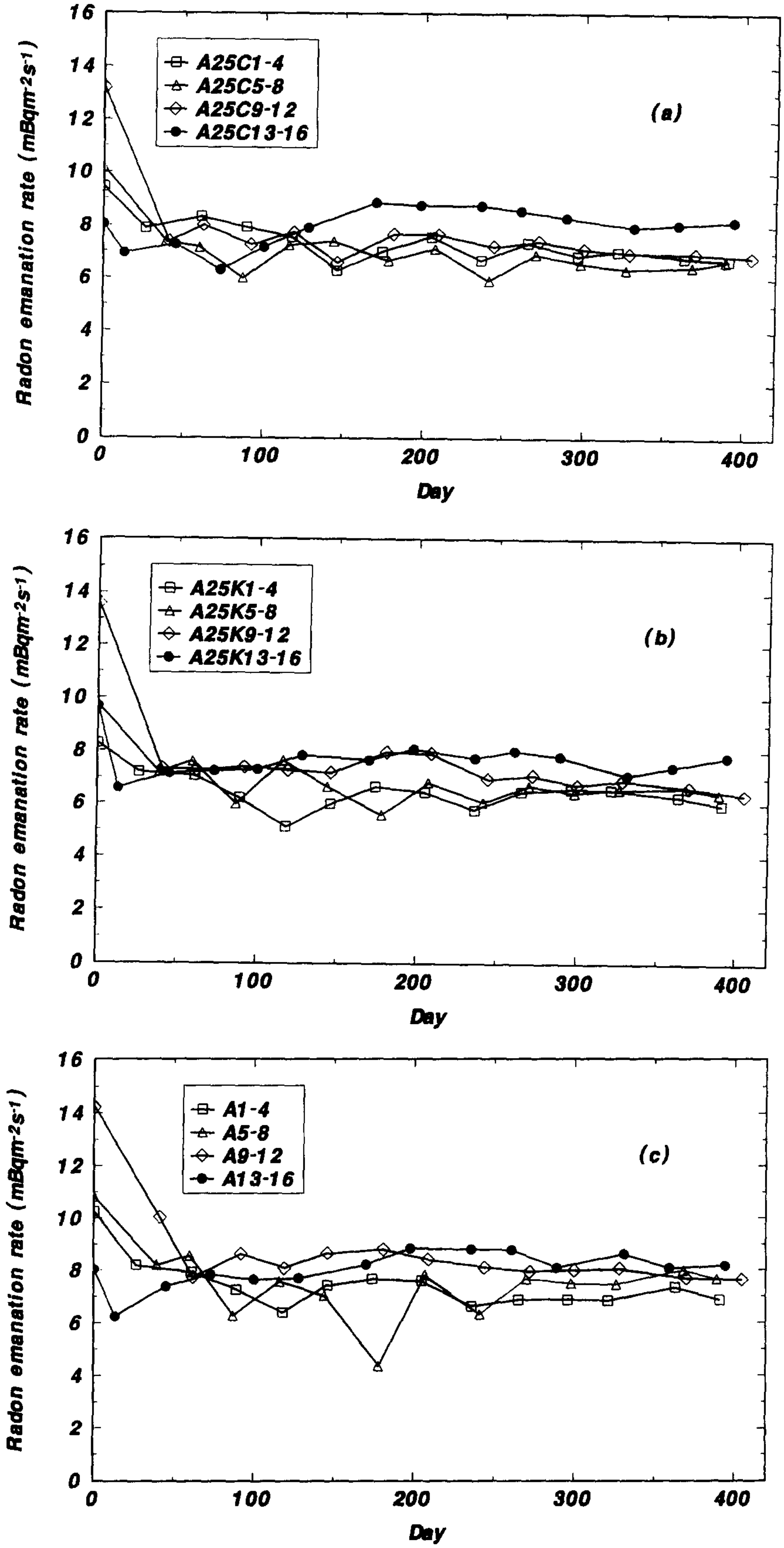

Fig. 1. Comparison of the temporal variation of radon emanation rate (mBq m^{-2} s^{-1}) from concrete blocks with different curing periods. (a) Mix A; (b) mix B; and (c) mix C.

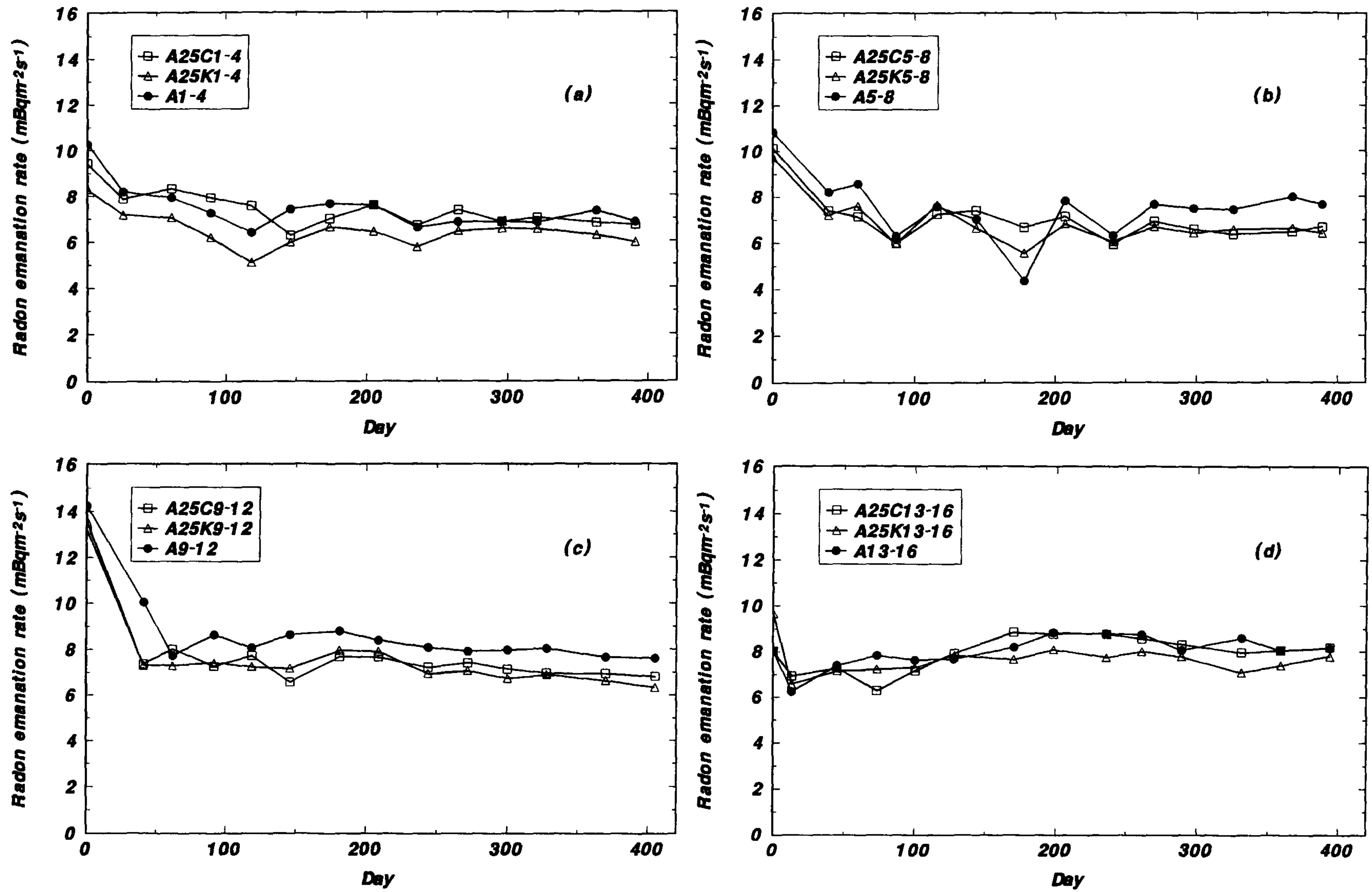

Fig. 2. Comparison of the temporal variation of radon emanation rate (mBq m^{-2} s^{-1}) from concrete blocks with different mixes. Curing periods: (a) 1 day; (b) 3 days; (c) 7 days; and (d) 28 days.

measurements and 3600 is the conversion factor from hours to seconds (Yu *et al.*, 1995).

The radon emanation rates from the surfaces of all of the concrete blocks have been monitored over a period in excess of 1 yr in a laboratory maintained at an even temperature of 23°C and a relative humidity of about 60%. Each measurement of a block represents an average of the monitoring of three adjacent surfaces in order to minimize the effects of possible uneven distribution of granite inside the concrete blocks.

Results and Discussions

The results of measurement of radon emanation rates from the concrete blocks are shown in Fig. 1. Each data point in Fig. 1 represents the average of 12 data (measurements on three adjacent faces on each of the four identical concrete blocks). From Fig. 1, the following may be observed for all mixes. The first major feature is that rates of change of radon emanation can be identified for two distinct periods, i.e. the first month following curing and for the period following the first month. During the first month, it is evident that the radon emanation rate is mainly controlled by emanation from the superficial pores. The rapid fall in radon emanation during this period can be directly linked with the rapid drying out of the superficial pores. After the first month, the superficial pores will have effectively dried out and with radon atoms being less likely to be trapped inside superficial pores by moisture, exhalation will slow to a steady rate controlled by the inner pores which are drying out at a much slower rate.

In terms of the first period, on the zeroth day, the concrete blocks cured for 1 day have the smallest radon emanation rates, those for 3 days are somewhat larger, while those for 7 days have the largest. This can be rationalized to be due to the larger water content of concrete blocks cured over longer periods. Conversely, however, for the concrete blocks cured for 28 days, these have either the lowest or close to the lowest radon emanation rates on the zeroth day (the results for mix B, which record the second lowest emanation rates, might be due to fluctuations as discussed by Ulbak *et al.*, 1984; and Roelofs and Scholten, 1994). The small radon emanation rates for long curing periods is suggested to be due to the nearly saturated water content of the superficial pores and the consequent inhibition of diffusion of radon atoms from the pores.

In terms of the second period, for concrete blocks cured for between 1–7 days, the radon emanation rates quickly merge together and follow a slow trend decreasing towards an equilibrium value of exhalation. This behaviour is suggested to be due to the gradual dehydration of concrete with ageing, with gradual reduction of the water content of inner pores and a commensurate slow release of radon due to a lessened probability of retaining radon within the pores. Again, conversely, for concrete blocks cured for a period of 28 days, the radon exhalation rate tends to increase with the age of the block. This may be rationalized to be due to the gradual dehydration of the initially relatively high water content of the inner pores of these particular blocks, with the net effect that the probability of radon emanation from these pores is now enhanced. Over the long-term, the enhanced radon emanation rates from concrete blocks cured for 28 days are maintained.

Figure 2 shows the comparison of the mean temporal variation of radon emanation rates from concrete blocks having different mixes. It can be observed that the general pattern is similar for the different mixes. Although the present results show that the radon emanation rates are reduced by the addition of PFA, there is no general conclusions for results from PFA substituted concrete, previous research having sometimes shown there to be an enhancement (Siotis and Wrixon, 1984), sometimes a reduction (Stranden, 1983; Van der Lugt and Scholten, 1985) and sometimes no effects at all upon radon exhalation rates (Ulbak *et al.*, 1984). The present group are unable to provide further resolution of this variability. More study is required in this important area as PFA is a potentially useful building material.

Acknowledgement—This research is supported in part by the research grant 7000426 from the City University of Hong Kong.

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