# Radon exhalation from concrete containing coal fly ash

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## Abstract

Building materials and products contain traces of natural radioactive elements, which originate from soil and rock. The radioactive elements in building products are mainly natural radionuclides from the disintegration chains of uranium-238 ($^{238}U$) and in particular the part of this chain starting from radium-226 ($^{226}Ra$) and its decay products, thorium-232 ($^{232}Th$) and its decay products, and natural radionuclide potassium-40 ($^{40}K$).

An addition of coal fly ash (FA) as replacement of sand in the concrete industry, resulting from environmental and economic considerations, can enhance the content of radionuclides in building products.

The research question, accordingly, was whether the addition of FA to concrete might affect its radon exhalation rate, and if it may harm the radiation exposure of buildings' occupants. In Israel, residential buildings must include shelter rooms, which are made of massive concrete, and also are equipped with extremely airtight window. Consequently, these rooms, which are used regularly as children's bedrooms or studies, are more prone to radon exposure than the other rooms of the dwelling. For the sake of conservative conclusions, this kind of rooms is in the focus of the current study.

Due to the interdisciplinary character of the project, it was executed jointly by researchers from the Soreq Nuclear Research Center and the Faculty of Civil and Environmental Engineering at the Technion. The study was coordinated by two professional steering committees, from the Ministry of Construction and Housing, and from the National Coal Ash Board.

The study dealt with a number of issues: laboratory tests of the content of radionuclides and radon exhalation of concrete mixes, with and without FA,

manufactured both in the lab and in the concrete plant; tests of radon concentration, in six shelter rooms of a building located in the center of the country, under different ventilation conditions provided by varying opening/closure of the window and door; testing air change rates in the studied rooms using the $SF_6$ decay method; development of analytical models for the assessment of radon exhalation in testing conditions and estimation of radon concentration in living conditions, and the relation between them; studying the correspondence between the model and the experimental measurements; conservative evaluation of the daily and annual exposure to radon under typical occupancy conditions.

The main conclusions are:

1. The literature survey showed that the Israeli standard for radioactive materials in building products, SI 5098, which is based on a maximal excess dose of 0.3 mSv including the effect of Radon on the Radiation Index, is the strictest. In comparison the Austrian standard, which also addresses radon, is based on an excess dose of 1 mSv, and the Danish standard, which is based on the 0.3 mSv excess does, does not address radon. SI 5098, as the Austrian standard, addresses the areal mass density of the product, thus handling the material density in the appropriate manner. Whereas foreign regulations, such as RP-112 and the Danish standard, which assume a single density (e.g., 2300 kg/m3) are too strict with lightweight materials and too lenient with heavyweight ones.
2. A composite analytical model was developed for the simultaneous direct calculation of radon exhalation rate from the building elements and expected radon concentration. It is based on basic properties of the material, which are usually not available from the measurements. Consequently, for practical applications calculations can be based on disengaged analysis, where the surface free exhalation rate, $E_{0w}$, is the coupling term. This factor can be derived from the free radon exhalation rate, $E_0$, which is obtained when testing concrete samples according to SI 5098, multiplied by 3.76.
3. The assumption that zero or negligible air change rate prevails is allowed only when results show that the effective radon decay rate does not differ significantly from its natural decay rate, i.e. $\lambda_{eff} \approx \lambda_{Rn}$. In this caes simplified

formulations apply. If this condition is not met, the calculations have to be carried out taking into account the more accurate model, which includes all the expressions which include the air change rate in the space or testing chamber. The results of the current study are based on this conclusion.

4. The comparison between the direct measurements of air change rate (using $SF_6$ decay) and the values obtained from the difference between effective and natural radon decay rates demonstrated that they are of the same order of magnitude, but not sufficiently similar. The differences are explained partially by radon "back diffusion", which was found not negligible. The value obtained for the radon surface transfer coefficient, $\beta_{Rn}$, was not constant, and could not be determined accurately due to the large experimental scatter. The literature survey, as well, did not reveal accepted values.

5. Verification of the model was obtained by comparing values of the surface free exhalation rate, $E_{0w}$, which were derived in two independent manners: 1) by the laboratory tests of the cast concrete samples, 2) by the model, using the measured time evolution of radon in the rooms. The fit of results was excellent (6.5-9.2 $Bq/m^2h$ for the lab tests, and 5.9-11.7 from the model).

6. Laboratory tests of concrete mixes manufactured in the lab showed that, as expected, radionuclides concentration, and in particular that of $^{226}Ra$ and $^{232}Th$ in FA concrete was higher than in no FA concrete by 22-36% and 55-110%, respectively, where the high repeatability indicates good homogeneity of the mixes. In contrast, the radon emanation coefficient tends to decrease with increase in the FA content. In addition, it was found that deficient curing (1 day only) decreased significantly emanation coefficients.

7. Laboratory tests demonstrated that radon exhalation rate and emanation coefficients of concrete without FA were higher than those in concrete with FA (by 18-37% and 13-66%, respectively). At the same time, from the analysis of the measurements in situ, the influence of the concrete type on calculated free radon exhalation rate was not found as significant. The dependence of the emanation coefficient on concrete age was not unequivocal, despite the fact that most concrete mixes (except for the concrete mix

containing 150 kg/m$^3$ of FA) showed, under standard curing conditions, a slight tendency to decrease.

8. The general experimental scatter, due to the big number of uncontrolled influencing factors under the conditions of the real construction site, including the influence of the different calculation methods, was larger than the range of the expected influence of FA addition.

9. Direct measurement of air change rate by means of the $SF_6$ gas in two shelter rooms showed that under the hermetically closed conditions, when both the window and steel door were closed tightly, ventilation rate was rather low and close to the physical radon decay rate. In the tightest scenario under typical occupancy conditions, when both the window and regular wooden door are closed, the air change rate was 0.13 to 0.26 $h^{-1}$. For the rest of the scenarios, simulating different service conditions from the viewpoint of the wood door and window position, the air change rate was always higher than 1.0 $h^{-1}$ reaching even several tens $h^{-1}$.

10. A conservative estimation of the expected radon concentration in typical shelter rooms under regular occupancy conditions, i.e. when the air change rate is 0.1 $h^{-1}$ and radon exhalation rate from all the room envelope elements is 12 Bq $m^{-2}$ $h^{-1}$, showed that the maximum radon concentration will not exceed 150 Bq $m^{-3}$, and that the annual average (even if these conditions return every night) will be lower than 60 Bq $m^{-3}$. Taking into consideration that free radon exhalation rate in the lab tests was lower than this value, and that such a low air change rate will usually force occupants to open the window or door, due to increased relative humidity and water condensation on the window surface, and because of the general discomfort of staying in the room, it is reasonable to assume that such conditions will not persist under regular service conditions, and may happen only rarely, in very rare extreme cases. In the more realistic situation, when the air change rate is not less than 0.25 $h^{-1}$, the estimated radon concentration did not exceed 90 Bq $m^{-3}$, and the annual average radon concentration is expected to be less than 35 Bq $m^{-3}$. Moreover, even under self-defense conditions, when the air tightness degree becomes especially high and the ventilation rate decreases virtually to zero, the maximum radon concentration after 5 hours of staying in the hermetically

closed shelter room will not exceed 130 Bq $m^{-3}$, which has also been confirmed by the direct measurements in the building.

Hence, we conclude that radon exhalation from concrete of all the tested types (both with and without coal fly ash) is not expected to exceed the 200 Bq/$m^3$ action level, neither under regular everyday occupancy conditions (air change rate does not fall below 0.25 ACH), nor under special self-defense conditions, even if the latter persist for several hours. Addition of FA up to 150 kg/$m^3$ did not increase the radon exposure. These findings apply in general whenever the concrete samples are tested according to SI 5098 (on 10X10X20 cm prisms) show $E_0$<3.2 Bq/$m^2$h and the wall surface translated value is: $E_{0w}$<12 Bq/$m^2$h.