EXHALATION OF RADON-222 FROM BUILDING MATERIALS

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Abstract.

Many building materials contain small amounts of the radioactive element radium. By decay of radium is formed the radioactive gas radon, which might diffuse out of the material and thus be found in the ambient air. When radon and especially its short lived daughter products are inhaled radiational damage of the lung tissue might result.

Some results of field measurements of radon levels in apartments and houses are reported and it is shown that summer measurements with high natural ventilation rates are generally lower than winter measurements.

The exhalation of radon from building materials can be studied by placing samples in closed vessels and following the growth of activity in the vessels. It is shown that a ventilation rate of one air exchange per hour will lower the theoretical maximum level to 0,8 % of the unventilated maximum value.

Several of the indoor climate parameters have effects upon the human organism which can be directly observed through one or more of the human senses. This is for example the case with impressions of light, sound, odors etc.

In contrast to these we find a series of influences where the effect is uncertain (for instance atmospheric electrical influences) or statistical by nature and often with long time lags between the exposure and the appearance of the effect. The latter is for example the case with radioactive influences.

Radioactivity

The nuclei of certain elements are by nature unstable and can through a radioactive process transite to a more stable state.

The excess energy of the nucleus is given off during the transition either in connection with the emission of a particle (α- and β-activity) or directly as electromagnetic radiation of very short wavelength (γ-activity).

In the natural environment of man, sources of radioactivity are found as part of the soil, the atmospheric air and of many building materials. In the present context we will limit ourselves to the role of building materials as sources of potential radiation hazards. Several radioactive elements like radium, thorium, sodium and strontium are found as trace elements in building materials like concrete, bricks etc. The α- and β-radiation from these elements will be almost completely stopped in the material itself and is thus without hygienic interest. The γ-radiation will give a contribution to the total radiational load of human beings in the surroundings in question, but this contribution is for most materials too low to significantly increase any radiational health hazard.

One of the commonly occurring radioactive elements, radium, however, can give rise to health problems of a special kind. Radium, $^{226}_{88}Ra$, is found in low concentrations ($\sim 10^{-12}$ g of radium per g of other materials) in most building materials. The radium atoms are embedded in the base material matrix and the α-radiation accompanying the decay of the radium atoms is of no health interest.

Radon

By the decay of radium, however, a new element, radon, $^{222}_{86}Rn$, is formed. Radon is an inert gas and has a certain probability for diffusing out of the material into the surrounding air where the radon atoms will be mixed with the molecules of the air and thus form a part of our breathing air. Radon itself is also α-radioactive, and if a radon atom decays in the lungs the emitted α-particle will very likely destroy or change cells of the epithel of the lungs. Although a part of the inhaled radon atoms will be dissolved in the water in the respiratoral tract the major part will be exhaled again before they decay. By the decay of radon a series of daughter products are found of which especially the four so-called short lived radon daughters are of interest.

Radon daughters

The short lived radon daughters are $^{218}_{84}Po$ (radium-A), $^{214}_{82}Pb$ (radium-B), $^{214}_{83}Bi$ (radium-C) and $^{214}_{84}Po$ (radium-C'). All four daughters are chemically very active and will easily attach to airborne particles and condensation nuclei. Consequently they are very likely to be deposited in the respiratory tract when inhaled, and since the two polonium isotopes, radium-A and radium-C', are α-emitters the radiation from the radon daughters represents a considerably greater health hazard than does the radiation from radon itself.

Maximum permissible levels

The potential effect of an exposure to the radiation from radon and its daughter products is damage of the epithel cells in the respiratory tract, in the worst case with cancer of for instance the lungs as a result.

There are no well defined threshold concentrations of airborne radioactivity below which the exposure is harmless. There has, however, over the years been suggested certain maximum permissible levels (MPL) corresponding to various situations.

The units used are picocurie per liter (pCi/l) or bequerel per m^3 (Bq/m^3) where 1 pCi/l = 37 Bq/m^3. These units can be used for the individual concentrations of radon and the daughter products. Alternatively a unit called a working level (WL) can be used to give the total radiational load of the respiratory tract due to α-radiation from the daughter products of radon. The WL-unit is defined so that a concentration of 1 pCi/l of radon in equilibrium with its daughters will give 0.01 WL. Most international advisory committees on radiation protection seem to agree in recommending a MPL-value of 0.03 WL for work rooms (40 hour per week exposure) and 0.01 WL for living quarters. These values are suggested for the general public and the corresponding values for professional exposures are 10 times higher. Since radon rarely is in equilibrium with its daughter products the WL-values of 0.01 and 0.03 may correspond to radon concentrations of 2 and 6 pCi/l.

Field measurements

The possibility of indoor elevated levels of airborne radioactivity was pointed out and demonstrated in the middle fifties, and over the last decade a series of investigations have shown that people in many areas of the world are in fact exposed to indoor levels of radon and radon daughters which should be considered unacceptable. The main result of danish measurements performed in 1976-1977 (1, 2, 3) showed, that in ordinary brick houses radon levels above 1 pCi/l are only found in unventilated basement rooms but never in ordinary living rooms. For houses made of concrete, concentrations above 1 pCi/l were rarely found in rooms which were mechanically ventilated or just ventilated through windows, while levels considerably above 1 pCi/l were often found in rooms with no mechanical ventilation. It was furthermore clearly demonstrated that naturally occurring levels were often much higher during the winter months, where people tend to keep doors and windows closed, than during the summer months.

A comprehensive swedish investigation deals with this same problem and will be discussed by the next speaker.

Exhalation

The main sources of radon in the indoor environment are the building materials in the walls, floor and ceiling of the rooms. The radon atoms are fed into the room air through diffusion from the walls by the so-called exhalation process. The removal processes of radon are radioactive decay and ventilation.

The exhalation from a given material can at constant pressure be considered as a material constant. The exhalation rate can be determined by enclosing a sample of the material in a closed vessel and follow the growth of radioactivity in the container over a period of 10-14 days (4). The exhalation rate E is for example measured in atoms per m^2 per second. If it is assumed that one can scale up the results obtained for small samples to a real room, the maximum concentration of radon $R_{max,o}$ in a room with the volume V and surface area A is

$$R_{max,o} = \frac{A}{V} \cdot E \qquad (1)$$

if the exhalation rate is E for all the surfaces of the room and if the room is completely sealed off.

If $\frac{A}{V}$ is assumed to be 2 m^{-1} we find that a maximum concentration of 1 pCi/l or 37 Bq/m^3 corresponds to an exhalation rate of

$$E = 19 \frac{\text{atoms}}{m^2\text{-}s} .$$

As an example ordinary concrete may show exhalation rates in the order of 150-200 $\frac{\text{atoms}}{m^2\text{-}s}$ which thus leads to

$$R_{max,o} \simeq 10 \text{ pCi/l}$$

for a completely sealed off room with walls, floor and ceiling made of concrete.

Influence of ventilation

If the room is ventilated with a rate of n radon free air exchanges per unit time equation (1) becomes

$$R_{max,n} = \frac{\lambda}{\lambda+n} \cdot \frac{A}{V} \cdot E = \frac{\lambda}{\lambda+n} \cdot R_{max,o} \quad (2)$$

where λ is the decay constant for radon = $7{,}554 \cdot 10^{-3} h^{-1}$.

It thus appears from (2) that a ventilation rate of 1 h^{-1} (one exchange per hour) will lower the radon concentration to 0,8 % of the unventilated value. Considering again the concrete lined room we find that in order not to exceed a radon concentration of 1 pCi/l a ventilation rate of 0,07 exchanges per hour is necessary. Certain types of alum shale based light weight concrete however have exhalation rates of 10 times that of ordinary concrete. The corresponding necessary ventilation rate is then approximately 0,6 times per hour, still under the assumption that all the faces of the room is made of the material in question.

It should be mentioned that since the half life of radium is approximately 1600 years the ventilation rate does not slow down significantly with time. It is possible to a certain degree to decrease the exhalation rate by sealing the pores of the materials. This effect, however, has not yet been sufficiently examined.

Concludingly it can be summarized that several commonly used building materials are able to give rise to unacceptably high levels of indoor radioactivity. It is, however, possible beforehand by a simple measurement to predict the maximum expected level at a given ventilation rate.

Litterature

1 Niels Jonassen, Radon og dets datterprodukter i indendørs luft, Indledende målinger, Rapport fra Laboratoriet for teknisk Fysik I, DTH, Lyngby, 1975.

2 Niels Jonassen and J.P. McLaughlin, Radon in Indoor Air I, Research report 6, Laboratory of Applied Physics I, Techn. University of Denmark, Lyngby, 1976.

3 Niels Jonassen and J.P. McLaughlin, Radon in Indoor Air II, Research report 7, Laboratory of Applied Physics I, Techn. University of Denmark, Lyngby, 1977.

4 Niels Jonassen and J.P. McLaughlin, Exhalation of radon-222 from building materials and walls, Natural Radiation Environment III, Houston, Tx, 1978, (available at Laboratory of Applied Physics I, Techn. University of Denmark, Lyngby).

DISCUSSION

S.-P. Nygaard
Danish Employers' Confederation

Why do you find higher radon concentration in homes during the winter than during the summer?

Does the combustion of fossile products contribute to the radon concentration?

Has any yearly variation in the atmosphere's radon concentration ever been measured?

N. Jonassen

The elevated radon concentrations during the winter season is undoubtedly caused by a lower ventilation rate.

The combustion of fossile products (coal and gas) has been shown to produce extra radon. We have not made any measurements to determine this effect.

The concentration of radon in outdoor air has been shown to depend upon several meteorological paramters, for example pressure variations, precipitation, and snow cover. A yearly variation will differ strongly from place to place, and I do not recall ever having seen results of such measurements.

D.J.Nevrala
British Gas
Corporation

Have you measured the ventilation rate in the monitored dwellings, or in similar dwellings?

N. Jonassen

The ventilation rate was not measured.

J.S.R.Nielsen
Birch & Krogboe K/S,
DK

You mentioned a max. concentration with ventilation = 0, given by

$$R_{max,o} = \frac{A}{V} \cdot E \quad \frac{\text{atoms}}{\text{sek} \cdot m^3}$$

As far as I can see the concentration of atoms will raise with $R_{max,o}$ per sek per m^3.

Did I misunderstand the word concentration in relation to this?

N. Jonassen

With an exhalation rate of E the input of radon atoms per m^3 per sec is

$$\frac{A}{V} \cdot E$$

The maximum concentration is reached when the activity (number of decays per sec (and per m^3)) equals the input. The maximum activity $R_{max,o}$ is therefore

$$R_{max,o} = \frac{A}{V} \cdot E \, \frac{\text{decays}}{\text{sec} \cdot m^3}$$

The words concentration and activity are both used for this quantity. If one wants the maximum number of radon atoms per m^3 this figure is given by

$$N_{max} = \frac{R_{max,o}}{\lambda} = \frac{A \cdot E}{\lambda \cdot V} \quad \frac{\text{atoms}}{m^3}$$

where $\lambda = 2 \cdot 10^{-6} s^{-1}$ is the decay constant of radon.

I.Andersen
University of
Århus, DK

As far as I remember you have in a study shown that the radon concentration in brick-houses was much lower than in concrete-houses. Could you comment on that, and give an estimation of the relative health risk of living in houses made of these two building materials?

N. Jonassen

Danish clay based bricks have a lower radium content and lower (diffusion) porosity than does ordinary concrete. The exhalation of radon is therefore lower and so is, as a consequence the radon concentration in the air at comparable ventilation rates. A rough estimate suggests radon concentrations in concrete houses twice those in brick houses. I do not feel qualified to extrapolate the health risk estimates to the levels in question.