RADON EXHALATION RATES FROM BUILDING MATERIALS AND FLY ASH-CONTAINING CONCRETES

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ABSTRACT

Natural radioactivity and radon exhalation rates on a large number of Italian building materials are presented and compared with identical measurements on coal ashes. The influence of fly ashes on concrete exhalation rates was investigated by performing accurate radon emanation measurements on slabs with different ash-to-cement ratios.

INTRODUCTION

High indoor radon concentrations are usually considered important sources of radiological hazard for the general public. For this reason an experimental program has been implemented to investigate the following parameters:

a) the naturally occurring radionuclide content in different building materials selected according to the manufacturer's location and capacity, and in a large number of ash samples taken from Italian power plants;
b) the radon emanation characteristics of above materials;
c) the effect, on radon emanation, of ashes used as a partial substitute for cement in concrete.

MATERIALS AND METHODS

Naturally occurring radionuclide analysis

Germanium-detector gamma-ray spectrometry was used to determine ^{226}Ra, ^{228}Ra and ^{40}K concentrations (see Table 1).

Exhalation rate and emanating power

Exhalation rate and emanating power were measured in:

a) powder samples (cements, hydrated limes, sands) analyzed without grinding;
b) bricks and concretes analyzed both in their original form and ground (particle size between 4 and 10 mm);
c) 20x20x10 cm concrete slabs made by mixing products individually characterized as described above.

Powder and ground samples were placed in containers which are small (11 cm dia. and 8 cm high) compared with radon diffusion length for this kind of material; it may therefore be assumed that radon will not decay significantly in the sample[1].

The radioactive gas emanation was assayed by evaluating the initial radon growth slope[2] through the continuous monitoring of the gas concentration in a box sized to ensure easily measurable radon activity levels by keeping gas back-diffusion within acceptable limits. CISE-designed flask monitors[3] ensuring an efficiency of 0.27 cpm/(Bq/m^3) and a background of 1.2 cpm were used.

Analytical results (mean and standard deviation), are summarized in Table 2 for different kind of material.

Radon emanation from concrete and moisture content

The influence of free water content on radon emanation from 20x20x10 cm concrete samples was studied by measuring radon exhalation rates from concrete slabs saturated with water and then sequentially dried in oven.

Radon emanation from concrete with fly ashes

The effect of cement hydration on emanation properties was investigated measuring radon exhalation rates from 20x20x10 cm concrete slabs made with low-radium inert materials (white limestone) and different types of cement. In Table 3 are listed the radiological characteristics of the used materials.

The effect of fly ashes on concrete radon emanation was investigated on 20x20x10 cm slabs in which cement was replaced with South African and American fly ash in a proportion ranging from 5% to 40%. Results are reported in Table 4 (columns 2 and 3). In columns 8 and 9 are presented fly ash emanating power values computed on the assumption that a) cement has an average emanating power of 56.8%, calculated from the results of previous tests (see Table 3), and b) the inert material exhalation rate remains unchanged.

RESULTS AND DISCUSSION

Naturally occurring radionuclides and exhalation rates

Tables 1 and 2 show a marked variability in natural radionuclides concentrations and in exhalation rates for all groups of materials. Insofar as average values are concerned, coal ashes have ^{226}Ra levels comparable with those found in volcanic materials, while exhalation rates are half-way between those of portland-pozzolanic cements and the other tested materials. Coal ashes have very low emanating power values, comparable with those of bricks.

Radon emanation from concrete and moisture content

Results show that the radon exhalation rate varies, with a good linear correlation, from 0.2 to 4 $\mu Bq\ kg^{-1}\ s^{-1}$ when moisture content rises from 0% to 5%. This phenomenon may be due to the radon atoms adsorption on the pore surface within the concrete[(4)(5)]. The presence of water favours radon atoms migration outside the concrete. When there is no water inside the pores, radon atoms can adsorb on the pore surface; when water fills the pores, an increasing amount of radon can move out.

Radon emanation from concrete with fly ashes

Results summarized in Table 4 (columns 2 and 3) show that the addition of fly ashes causes a slight increase in radon emanation. Increase is smaller for American fly ashes than for South African ones.

Table 3 (column 4) and Table 4 (columns 6 and 7) show that concrete emanating power values are in the range 15÷30%. These values are higher than those of cements, inert materials and fly ashes (see Table 3). Assuming that inert materials do not change their exhalation rate when imbedded in concrete, cement emanating powers after hydration can be calculated. Cements after combination show emanating power values in a narrow range from 55 to 59%

(average value 56.8%) for concrete slabs made with different kind of cement and inert materials. Now, assuming that a) inert materials do not change their emanation properties when they are in concrete and b) cements have an emanation power of 56.8%, fly ashes radon emanation power values inside the concrete (see Table 4 columns 8 and 9) range from 11.2 to 21.9% (average value 17%). Fly ashes show an increase from 1 to 17% which is similar to that showed by cements (from about 4 to 57%).

CONCLUSIONS

The investigation carried out on Italian building materials has shown a substantial homogeneity in natural radioactive levels, except for volcanic materials (tuff, pozzolana, etc.). As far as exhalation rate values are concerned coal ashes are at an intermediate level between portland cements and pozzolanic cements.

Tests on concrete with fly ashes show that there is a slight increase in radon emanation but the radiological hazard connected with the use of fly ashes in the building industry seems to be negligible taking into account that the incidence of radon exhalated by building materials on indoor radon levels can be estimated in the 10-20% range[6][7] of the total indoor concentration.

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Table 1 - Natural radionuclides concentrations in building materials and coal ashes (average ± st.dev.).

Material	^{226}Ra [Bq/kg]	^{228}Ra [Bq/kg]	^{40}K [Bq/kg]
Portland cements (31 samples)	26 ± 15	18 ± 12	210 ± 40
Pozzolanic cements (21 samples)	49 ± 26	45 ± 38	390 ± 200
Sands and gravels (61 samples)	15 ± 6	17 ± 11	390 ± 230
Bricks (124 samples)	40 ± 17	40 ± 16	710 ± 210
Hydrated limes (13 samples)	10 ± 5	2.2 ± 2.7	77 ± 130
Peperinos (6 samples)	160 ± 50	190 ± 70	1420 ± 130
Pozzolanas (15 samples)	210 ± 130	250 ± 150	1660 ± 430
Tuffs (26 samples)	160 ± 120	200 ± 140	1640 ± 510
Ashes (151 samples)	170 ± 40	140 ± 40	410 ± 190
Fly-ashes USA (71 samples)	170 ± 40	130 ± 30	470 ± 100
Fly-ashes South Afr. (70 samples)	170 ± 40	150 ± 40	330 ± 210
Bottom ashes (10 samples)	130 ± 30	100 ± 20	470 ± 290

Table 2 - Exhalation rates and emanating powers of building materials and coal ashes (average ± st.dev.).

Material	Exhalation rate [μBq kg^{-1} s^{-1}]	Emanating power [%]
Portland cements (21 samples)	2.1 ± 1.2	5.2 ± 4.0
Pozzolanic cements (15 samples)	9.3 ± 6.7	8.8 ± 4.2
Sands and gravels (31 samples)	2.1 ± 1.4	8.4 ± 3.7
Bricks (30 samples)	1.3 ± 1.0	1.7 ± 1.5
Hydrated limes (6 samples)	1.2 ± 0.4	8.6 ± 5.3
Coal ashes (34 samples)	5.4 ± 3.6	1.6 ± 1.4
Fly-ashes South Afr. (19 samples)	4.5 ± 2.2	1.3 ± 0.7
Fly-ashes USA (10 samples)	6.4 ± 4.0	1.6 ± 0.8
Bottom ashes (5 samples)	6.8 ± 5.4	2.9 ± 3.0

Table 3 - Radiological characteristics of materials used to make 20x20x10 cm concrete slabs. Emanating power values in column 4 marked with * are referred to calculated values for cement embedded in concrete assuming that inert materials do not change their exhalation rate.

Material		^{226}Ra [Bq kg^{-1}]	Exhalation rate [μBq kg^{-1} s^{-1}]	Emanating power [%]
Portland cement (4 samples)		13.9÷19.9	0.8÷2.7	2.1÷6.9
Inert	White limestone	3.1	0.51	7.8
	Sand and gravel	17.4	2.5	6.8
Concrete with both kinds of above inert material		4.1÷5.5	2.6÷5.5	16÷33 (55÷59)*
Fly ash	USA	175	3.7	1.0
	South Africa	150	3.5	1.1

Table 4 - Radiological characteristics of 20x20x10 cm concrete slabs made using sand and gravel as inert materials and different percentage of fly ash with respect to cement. Emanating power values in the last column are calculated assuming that inert materials exhalation rate does not change and cement exhalation rate is 56.8%.

Fly ash percentage [%]	Measured exhalation rate [μBq kg^{-1} s^{-1}]		^{226}Ra [Bq kg^{-1}]		Concrete emanating power [%]		Fly ash emanating power [%]	
0	5.49		16.16		16.2		---	---
	South Africa	USA	South Africa	USA	South Africa	USA	South Africa	USA
5	5.63	5.77	16.97	13.14	15.8	20.9	14.5	17.3
15	6.01	5.74	18.10	13.22	15.8	20.7	17.3	11.2
26	6.63	6.65	18.87	13.27	16.7	23.9	20.6	17.8
40	7.20	6.81	19.35	13.33	17.7	24.3	21.9	16.3