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Radioactivity of Building Materials Available in Northeastern Poland

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Abstract

This paper includes the results of measurements of natural radioactivity in building materials and raw building materials. The dose rate indoors was calculated on the basis of the contents of K-40, Ra-226 and Th-232 in building materials and the results were compared with literature data of measurements (in situ). The standard procedure for qualifying building materials for building houses designed for habitation was used.

Keywords: radioactivity in building materials, radioecology, natural radioactivity

Introduction

Ionising radiation is an element of the natural environment. It is bound up with cosmic radiation and radiation from natural radioactive elements in the ground and in building materials. Analysis of the effect of being indoors on total exposure has become extremely important since the time of introducing new building technologies based on by-products of energy, metallurgy and chemical industries: smoke-dust box, furnace slag, phosphogypsum. The above-mentioned sources of public exposure are responsible for small doses of radiation, but their large-scale character gives rise to the interest of biologists and medical doctors [1-4].

Natural radioactivity is mainly connected with the presence of potassium K-40 and radioisotopes of uranium U-238 series and thorium Th-232 series. Dose rates from gamma radiation depend mainly on the concentration of the above-mentioned radioisotopes in the soil and building materials. Radon Rn-222, a gaseous product of decay of radium 226, is extremely important in indoor exposure. Radon is a noble gas, whose α-radioactive derivatives (Po-218, Pb-214, Bi-214, Po-214) permeate through various levels of the breathing system in a non-bound with aerosols form. Radon easily diffuses from the ground and building materials to indoor air and is the source of exposing bronchus and lungs [5-7]. A specially unfavorable situation occurs in the case of using materials with high Ra-226 content with a simultaneous high factor of radon emanation from the walls.

In Poland, to limit the increase of natural background of radiation indoors, caused by the use of materials with high radioactivity in building, the Ministry of Building and Building Materials Industry together with The Central Laboratory for Radiological Protection (CLOR) and the Ministry of Health and Social Care introduced in 1980 norms for the contents of natural radioactive elements in raw and building materials [8]. Instruction No 234/80 of the Institute of Building Technology in Warsaw [9] defines two coefficients for qualifying whether building raw materials and final materials are acceptable for building houses designed for habitation.

Coefficient f_1 which must not be higher than 1, determines the limit of exposure of the body to gamma radiation and is defined as:

$$f_1 = 0.00027S_K + 0.0027S_R + 0.0043S_T < 1 \qquad (1)$$

where:
S_K, S_R, S_T are the contents of potassium K-40, radium Ra-226 and thorium Th-232 in a sample in Bq/kg.

Coefficient f_2, which determines the limit of the concentration of radium Ra-226 in a building material with reference to emanation of radon Rn-222 from the walls is defined as:

$$f_2 = S_R < 185 \text{ Bq/ kg} \qquad (2)$$

Only when both conditions are realized is the assessment positive and may the material be used in buildings designed for human habitation [10].

In this article there are presented the results of measurements of the natural radioactivity of raw and building materials, which were conducted in the laboratory of the Department of Biophysics Medical University in Bialystok, which does the research in northeastern Poland, a region with a well-documented radioecological analysis [11]. Such a great interest in the condition of the environment in this region is the result of the fact that the region is richly endowed with sights of natural beauty and as such has been subject to special eco-development policy [12].

Experimental Procedures

The standard measurements procedure designed for Polish laboratories was applied [9, 13]. The collected samples were 3 dm^3 in volume and weighed on average 2.5 kg. They were crushed in a ball grinder and put through a diameter 5 mm screen. Next they were dried to achieve a solid state at 110°C for about 48 hours. Marinelli-type containers of 1.5 dm^3 volume were used. The measurements were conducted with a 3 channel gamma analyser with a scintillation probe NaJ. The block scheme of the analyser is shown in Figure 1. The measuring method used by the laboratory is a comparative method. This consists of analysing the amount of counts of gamma radiation impulses registered in 3 measurement channels, separately for the researched sample and for 3 volumetric radioactivity standards. In the measurements were used standards Number 033 (in Marinelli DOP-50 containers) which were prepared in the Central Laboratory of Radiological Protection in Warsaw: potassium, radium and thorium with activities of: 10000, 2070, 688 Bq, respectively, and weighing 2.4 kg each. The

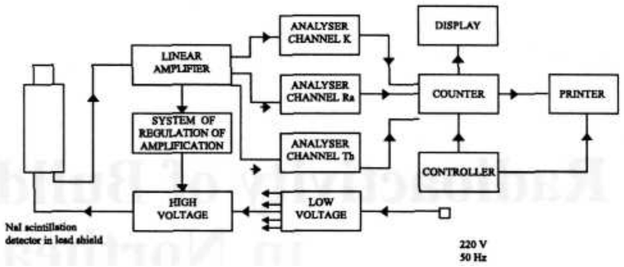

Fig. 1. The block scheme analyser.

analyser's channels were set for the following energy ranges:

- potassium channel (1.26- 1.65 MeV) - included photons with energy of 1.46 MeV coming from the decay ofK-40;
- radium channel (1.65- 2.30 MeV) - registered main ly photons with the energy of 1.76 MeV emitted by Bi-214 (uranium series)
- thorium channel (2.30- 2.85 MeV) - where were counted mainly the impulses coming from gamma radiation with the energy of 2.62 MeV, originating in the decay of Tl-208 (thorium series).

The method of measurements assumed identical time of measurements for the standards, samples and backgrounds, which was 2000 seconds. For each sample at least five measurements were taken. Calculation of the concentration of the radiation of potassium, radium and thorium and qualifying coefficients f_1 and f_2 was done with the use of a specially created computer program.

Results and Discussion

170 samples were measured, including 81 raw material samples and 89 final building material samples. They were delivered to the laboratory by the local power stations and boiler rooms, by the producers of building materials as well as by individuals interested in getting radiological certificates of the materials that had been used in the construction of their private houses. Mineral resources came mostly from local deposits (clay, sand) exploited by the pottery / ceramics factories. Partly, they were a by-product of the power engineering industry

Table 1. The mean concentrations of K-40, Ra-226, Th-232 and values of coefficient f_1 for building and raw materials.

Kind of material	Number of samples	Mean concentration (range) $Bqkg^{-1}$			f_1 mean (range)
		K-40	Ra-226	Th-232	
Brick	39	963 (411 – 1205)	50 (22 – 71)	50 (26 – 68)	0.6 (0.29 – 0.74)
Prefabricates	50	429 (20 – 1179)	27 (4 –96)	24 (2 – 60)	0.29 (0.02 – 0.78)
Raw materials used in building	81	495 (126 – 1032)	74 (4 – 154)	72 (22 – 91)	0.64 (0.2 – 1.1)

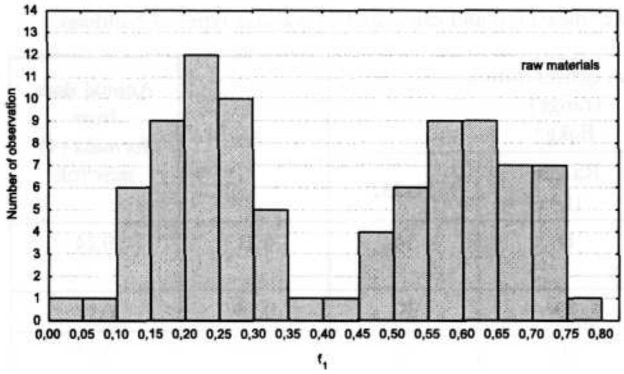

Fig. 2a. Variety of qualifying coefficients f_1 for raw materials.

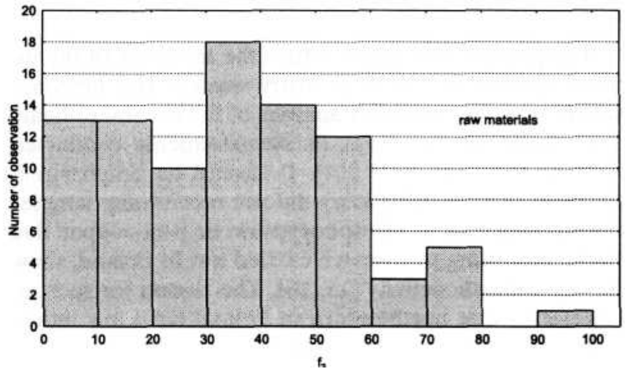

Fig. 2b. Variety of qualifying coefficients f_2 for raw materials.

(slag, ashes, dust) originating in local power stations and heating plants. The results of measurements are presented in Table 1.

The most favourable situation was found in the group of prefabricated elements, where coefficients fi and f_2 were the lowest and were rated at 29% and 15% of the acceptable value. Coefficients fi and f_2 (as shown in Table 1) are given without the statistical error. No sample in the group of prefabricates was higher than the norm stated in Instruction ITB 234/80 [9]. The mean values of the qualifying coefficients and the range of their variability for 50 measured prefabricates are:

$$f_1 = 0.29\ (0.02 - 0.78)$$
$$f_2 = 27\ (4 - 96)$$

If we accept the more stringent criteria of the year 1995 [13], which demand that the values of fi and f_2 are taken with the statistical measurement error (level 2a), then two samples of hollow clay blocks with slag addition should be discarded.

In the discussed group of prefabricates we can distinguish a number of basic types of building materials, i.e. lightweight concrete, silica bricks, solid concrete (OWP) and hollow clay bricks with slag addition. The values of the concentration of K-40, Ra-226, Th-232 and the qualifying coefficients f_1 and f_2 for these materials are shown in Table 2. The lowest radioactivity was found in the silica materials with the mean value of coefficient $f_1 = 0.15$, in which potassium K-40 inclusion is 54% and the mean concentration of Ra-226 and Th-232 at 10 Bq/kg. Slightly higher values of the qualifying coefficients are found in lightweight concrete and solid concrete. Coefficients f_1 are 21% of the norm with the inclusion of Ra-226 for lightweight concrete and OWT (solid concrete) at 11 and 26 Bq/kg respectively. Markedly higher concentrations of natural radioactive elements were found in slag-added hollow clay bricks, in which the mean values of f_1 and f_2 are 0.75 and 77.

Ceramic bricks showed the mean values of qualifying coefficients twice as high as those of prefabricates. The mean values of 39 measured samples were:

$$f_1 = 0.6\ (0.29 - 0.74)$$
$$f_2 = 50\ (22 - 71)$$

Only one sample of the ceramic brick did not meet the stringent criteria of the year 1995 with reference to f_1. The distribution of fi and f_2 for all ready made building materials is presented in the form of histograms (Figs. 2a, 2b). The distribution is of two-modal character, which

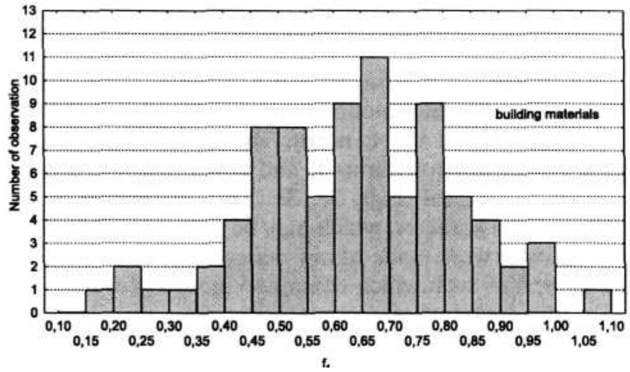

Fig. 3a. Variety of qualifying coefficients f_1 for building materials.

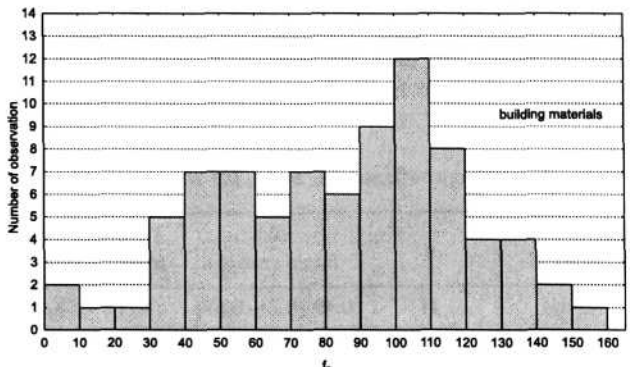

Fig. 3b. Variety of qualifying coefficients f_2 for building materials.

Table 2. The mean concentrations of K-40, Ra-226, Th-232 and dose rates measured and calculated for various types of buildings.

Kind of building material	Dose rate measured [17] $nGyh^{-1}$	Dose rate from formula (3) $nGyh^{-1}$	Mean concentration (range) $Bqkg^{-1}$ K-40	Ra-226 (f_2)	Th-232	f_1	Annual dose from formula (4) mSv/rok
Lightweight convrete	99 (82 – 115)	50	418	11	16	0.21	0.24
Silica brick		36	308	10	10	0.15	0.18
Solid concrete (OWT)	87 (73 – 90)	52	282	26	17	0.22	0.25
Slag concrete brick		177	1048	77	60	0.75	0.86
Ceramic brick	102 (71 – 131)	144	963	50	50	0.60	0.71

confirms the variability of the qualifying coefficients for prefabricated materials and ceramic bricks.

The largest diversity of results was found in the group of raw building materials. The mean values of coefficients f_1 and f_2 were 64% and 40% of the norm, respectively, and were:

$$f_1 = 0.64\ (0.2 - 1.1)$$
$$f_2 = 74\ (4 - 154)$$

According to the stringent norms accepting the coefficient with the inclusion of measurement error, 11 samples exceeded the f_1 norm. The distribution of the mean values of coefficients f_1 and f_2 is presented in the form of histograms, Figs. 3a, 3b. The distribution is of multi-modal character, which may be due to the fact that prefabricates were made of raw materials of different origin, as well as such which contained industrial waste.

In this category of raw materials, four main groups were differentiated. Their coefficients f_1 and f_2 are shown in Table 3. The lowest mean values of coefficients f_1 and f_2 are found in clay (f_1 = 0.49; f_2 = 40), which is the main raw material of the region used for the production of ceramic bricks. Industrial waste materials of the power engineering industry, which are added in the production of some building materials, show higher natural radioactivity and may be the reason of high qualifying coefficients in the ready made materials. The mean values of f_1 for slag, ashes and dust are: 0.62, 0.71 and 0.8, and the values of coefficient f_2: 91, 81, 101.

Table 3. Qualifying coefficients f_1 and f_2 for raw materials.

Kind of raw material	n	f_1 mean (range)	f_2 mean (range)
clay	11	0.49 (0.2 – 0.67)	40 (4 – 58)
slag	46	0.62 (0.2 – 1.1)	91 (15 – 154)
ash	5	0.71 (0.6 – 0.87)	81 (42 – 124)
dust	16	0.8 (0.61 – 0.98)	101 (49 – 150)

The presented results confirm the tendency of the increase quality factors in industry wastes. The tendency was observed in an earlier analysis of the natural radioactivity made on the basis of measurements conducted prior to the year 1993 [14]. It should be emphasized, however, that the laboratory did not receive any samples of such materials as phosphogypsum or post-copper slag which, according to research carried out in Poland, show the highest radioactivity [15, 16]. The reason for such an omission is that northeastern of Poland lacks any industry which would produce them.

The conditions existing in northeastern Poland enable us to verify the model assumptions used for Instruction ITB 234/80 [9] that served as a basis for the calculation of norms of the coefficients fi and f_2 with reference to the direct measurements of the exposition and the levels of Rn-222 in buildings [17, 18]. In the Instruction the exposition rate dose absorbed in the air, expressed in nGy/h was calculated according to a semi-empirical equation (formula):

$$D = (0.043S_k + 0.43S_{Ra} + 0.66S_{Th}) < 1.5 \qquad (3)$$

where:

S_k, S_{Ra} and S_{Th} - concentration of K-40, Ra-226 and Th-232 [Bq/kg]; coefficient 1.5 is the result of changing geometry from 2Π to 4 Π indoor building.

Using formula (3) and taking the mean values of radioactive radium, thorium and potassium of the typical building materials from Table 2 the rate doses and annual equivalents of doses for the buildings built of these materials were estimated. These model assumptions give a great diversity of results, from 36 $nGyh^{-1}$ for silica bricks, 50 $nGyh^{-1}$ for lightweight concrete, 52 $nGyh^{-1}$ for solid concrete (OWT), 144 $nGyh^{-1}$ for bricks and up to 177 $nGyh^{-1}$ for houses made of hollow bricks made of slag. Direct measurements of the exposition dose in the buildings in northeastern Poland show the values (including the cosmic radiation) for the houses made of lightweight concrete, solid concrete and bricks 99, 87, 102 $nGyh^{-1}$ respectively [17]. Thus, the model adopted in Instruction ITB 234/80, in the case of brick buildings, seems to be too stringent. In the case of solid concrete and lightweight concrete buildings the Instruction underestimates the dose inside the building.

Values of dose rates absorbed in the air can be used in estimating [5] the annual dose equivalent (H) expressed in mSv/year according to the formula.

$$H = 0.69 \cdot 7008 \cdot D \tag{4}$$

where:

0.69 - coefficient of transfer from dose absorbed in the air to dose equivalent

7008 - number of hours spent indoors in the year (80% of the year).

The limit of $f_1 < 1$ assumes that an additional annual dose equivalent indoors is acceptable at the level of 0.8 mSv above the value of 0.32 mSv, which is the result of average content of radioactive radium, thorium and potassium for the earth's crust. Direct measurements showed that the overall value of 1.12 mSv/year had not been exceeded in any of 342 apartments on the examined area, with a maximum found level at 57% of the above-mentioned overall value [17]. Model predictions based on formula (4) give the highest average for the buildings made of slag concrete bricks with 0.86 mSv/year, whereas the average for the most typical buildings made of brick is at the level of 0.71mSv/year and buildings made of solid concrete (OWT) - 0.25 mSv/year.

Establishing the norm for limiting the inner exposure of lungs and bronchus to alpha radiation emitted by the products of radon decay was made assuming the maximum concentration of Rn-222 at 46 Bqm^{-3}. Model considerations (presented at Instruction ITB 234/80 [9]) show that such concentration of radon may be the result of concentrations of Ra-226 in the building material at the level of 185 $Bqkg^{-1}$, which is the limit value of coefficient f_2. In none of the tested materials or raw materials was this norm exceeded.

In the light of radon measurements, the norm for coefficient f_2 proved inadequate. Numerous tests proved that only 20-25% of radon in the air indoors is the result of emissions from building materials. Most radon comes from the ground and in especially unfavourable conditions may greatly overstate the doses [4,5]. Thus, the current Polish regulations of acceptable concentrations of radon in habitable buildings introduces the value 400 Bq/m^3 for old buildings and 200 Bq/m^3 for buildings built after 1 January 1998 [19].

Extensive radon measurements conducted in north-eastern Poland showed that in 18% of the buildings the level 46 Bq/m^3 is exceeded, and in 3.2% buildings the exceeded level is 200 Bq/m^3 while in 1% - that of 400 Bq/m^3 [18].

These comparisons show that the norm for f_2 introduced in 1980 is not always sufficient. Thus, it was understood that new regulations for the acceptable concentration of radon in the air indoors had to be introduced [19], whereas the norm for fi limiting the dose rate from gamma radiation fulfils its role in eliminating building materials with excessive contents of natural gamma emitters.

The control activities that had been carried out with reference to the natural radiation of raw materials and building materials in northeastern Poland showed that the problem of increased exposure to humans, which might result from the use of building materials, does not concern the above-mentioned area. It may be the consequence of favourable natural conditions and the lack of any industry whose end-products (possessing a high level of natural radioactivity) might be used in the production of building materials and prefabricates. This is also confirmed by a positive radiological assessment of the discussed area in the earlier study [11]. Radiological control of building materials conducted in the laboratory located in the area is a good way of maintaining the existing state in future years.

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