Radiological constraints of using industrial by-products in construction

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ABSTRACT: The advantages of utilization of coal fly ash, phosphogypsum and some other industrial by-products in construction are demonstrated, as well as the technological and environmental problems caused by an elevated content of chemical/radioactive contaminants. Radiological aspects and legislation issues are analyzed. The tendency to develop stricter environmental norms observed in the last years in both national and international scale is discussed. The reasons and possible methods of solving the difficulties with the application of legislation rules and radiation controls in construction industry are discussed. As an example, an experience with the new Israeli Standard 5098 regulating radioactivity in building products is reported. The principles of this standard are analyzed and compared to the Radiological Protection Principles Concerning the Natural Radioactivity of Building Materials Principles of Radiation (Radiation Protection 112, European Commission) and other existing national standards and guidelines.

1 BUILDING MATERIALS AS A SOURCE OF INDOOR RADIATION EXPOSURE

Building materials contain various amounts of natural radioactive nuclides. For example, materials derived from rock and soil contain mainly natural radionuclides of the uranium (^{238}U) and thorium (^{232}Th) series, and the radioactive isotope of potassium (^{40}K). In the uranium series, the decay chain segment starting from radium (^{226}Ra) is radiologically the most important and, therefore, reference is often made to radium instead of uranium. The world-wide average concentrations of radium, thorium and potassium in the earth's crust are about 40 Bq/kg, 40 Bq/kg and 400 Bq/kg, respectively [RP-112 1999].

Increased interest in measuring radionuclides and radon concentration in building materials is due to the health hazards and environmental pollution.

Timber-based building materials have low concentrations of natural radioactive substances. In stone-based building materials the concentrations depend on the constituents. Materials used by the building industry that maybe of radiological significance include marl, blast furnace slag, fly ash, phosphogypsum, Portland cement clinker, anhydrite, clay, radium-rich and thorium-rich granites (used as aggregates in concrete or in dimension stone products) [RP-112 1999, UNSCEAR 2000, NOR 2000].

Most individuals spend 80% of their time indoors and natural radioactivity in building materials is a source of indoor radiation exposure [Zikovsky & Kennedy 1992, Othman & Mahrouka 1994]. Indoors-elevated dose rates may arise from high activities of radionuclides in building materials. Chronic exposure of human beings to low doses of ionizing radiation can cause health damages which may appear 5-30 years after the exposure [ICRP 1991]. The most critical damage which can result from such exposure is an increase in the probability of contracting malignant diseases by the person who was exposed and by his offspring. The risk increases with the dose, and the probability of the appearance of the damage is greater when the exposure starts at a younger age. It appears that the large scale use of by-products with enhanced levels of radioactivity as a raw material in building products can increase considerably the exposure of the population and therefore constitutes a real potential risk.

Radiation exposure due to building materials can be divided into external and internal exposure. The external exposure is caused by direct gamma radiation.

The internal exposure is caused mainly by the inhalation of radon (^{222}Rn) and its short lived decay products. Radon is part of the radioactive decay series of uranium, which is present in building materials. Because radon is an inert gas, it can move rather freely through porous media such as building materials, although usually only a fraction of that produced in the material reaches the surface and enters the indoor air. This fraction is determined by so called emanation ratio (or emanation coefficient) of the building product.

As the presence of radon gas in the environment (indoor and outdoor), soil, ground water, oil and gas deposits contributes the largest fraction of the natural radiation dose to populations, tracking radon concentration is thus of paramount importance for radiological protection.

The most important source of indoor radon is the underlying soil. In most cases the main part of indoor radon on the upper floors of a building originates from building materials. Typical excess indoor radon concentration due to building materials is low: about 10–20 Bq/m^3, which is only 5%-10% of the design value introduced in the European Commission Recommendation (200 Bq/m^3) [RP-112 1999]. However, in some cases the building materials may be an important source also. For example, in Sweden, the radon emanating from building materials is a major problem. There are about 300,000 dwellings with walls made of lightweight concrete based on alum shale (so called “blue concrete”) [NOR 2000].

2 EUROPEAN AND NATIONAL REGULATIONS OF NATURAL RADIOACTIVITY OF BUILDING MATERIALS

The European Basic Safety Standards Directive (BSS) sets down a framework for controlling exposures to natural radiation sources arising from work activities. Title VII of the directive applies to work activities within which the presence of natural radiation sources leads to a significant increase in the exposure of workers or of members of the public. Amongst the activities identified in the BSS as potentially of concern are those “which lead to the production of residues … which contain naturally occurring radionuclides causing significant increase in the exposure of members of the public…”. Such materials may include coal ash from power stations, by-product gypsum and certain slags which are produced in large volumes and which may potentially be used as building materials. The purpose of setting controls on the radioactivity of building materials is to limit the radiation exposure due to materials with enhanced or elevated levels of natural radionuclides. The recently published EC document [RP-112 1999] provides guidance for setting controls on the radioactivity of building materials in European countries. This guidance is relevant for newly produced building materials and not intended to be applied to existing buildings.

The guidelines of the European Commission [RP-112 1999] are the first comprehensive document issued by the EC, which sets the principles of radiological protection principles concerning the natural radioactivity (both external and internal) of building materials. RP-112 states that restricting the use of certain building materials might have significant economical, environmental or social consequences locally and nationally. Such consequences, together with the national levels of radioactivity in building materials, should be assessed and considered when establishing binding regulations.

Gamma doses due to building materials exceeding 1 mSv/year are very exceptional and can hardly be disregarded from the radiation protection point of view. Therefore, RP-112 recommends that national controls should be based on a dose in the range 0.3 – 1 mSv/year. This is the excess gamma dose to that received outdoors. This criterion is aimed to restrict exceptionally high individual doses.

When gamma doses are limited to levels below 1 mSv/year, the ^{226}Ra concentrations in the materials are limited, in practice, to levels which are unlikely to cause indoor radon concentrations exceeding the design level of 200 Bq/m^3. At the same time, some countries apply separate regulation for ^{226}Ra content, which requires that the amount of radium in building materials should be restricted to a level where it is unlikely that it could be a major cause for exceeding the design level for indoor radon introduced in the Commission Recommendation (200 Bq/m^3). For example, Nordic countries recommend 100 and 200 Bq/kg, respectively, as exemption and upper levels for the activity concentration of ^{226}Ra in building materials for new constructions as a source of indoor radon [NOR 2000].

The activity index in the EC document RP-112 and in the other national standards regulating radioactivity of building materials is calculated on the basis of the activity concentrations of radium

(^{226}Ra) in the uranium (^{238}U) decay series, thorium (^{232}Th) in the thorium (^{232}Th) decay series, and potassium (^{40}K). Other nuclides are sometimes taken into consideration as well; for example, the activity concentration of caesium (^{137}Cs) from fallout is regulated in the Finnish guidelines [Guide ST 12.2 2005].

If the activity index exceeds 1, the responsible party is required to show specifically that the relevant action level is not exceeded. If the activity index does not exceed 1, the material can be used, so far as the radioactivity is concerned, without restriction.

The criterion of meeting the standard is the non-dimensional value of so called activity concentration index taking into account the total effect of three main natural radionuclides, which can present in building materials ($I = {}^{226}Ra/300 + {}^{232}Th/200 + {}^{40}K/3000$, concentrations are given in Bq/kg). According to RP-112, the activity concentration index I shall not exceed the following values depending on the dose criterion and the way and the amount the material is used in a building (Table 1).

Table 1. Dose criterion recommended by EC [RP-112 1999].

Dose criterion	0.3 mSv/year	1.0 mSv/year
Materials used in bulk amounts, e.g. concrete	$I \leq 0.5$	$I \leq 1$
Superficial and other materials with restricted use: tiles, boards, etc.	$I \leq 2$	$I \leq 6$

The EC guidelines allow for controls to be based on a lower dose criterion, if it is judged that this is desirable and will not lead to impractical controls. It is recommended to exempt building materials from all restrictions concerning their radioactivity, if the excess gamma radiation originating from them increases the annual effective dose of a member of the public by 0.3 mSv at the most.

Most of the European countries apply their controls based on the upper end of the dose scale (1.0 mSv/year), however the recent Danish regulations [NIRH 2002] apply the strictest criterion based on the lower end of the dose scale (0.3 mSv/year). Among non-EU countries only Israel applies the strict regulations based on the maximum allowable dose excess of 0.3 mSv/year [SI 5098 2007]; the rest of the countries, which have similar regulations, apply more liberal dose criteria. The decision to apply a strict criterion of 0.3 mSv/year in these two countries can be explained by relatively low radioactive background resulting from the local geological conditions, because the majority of mineral resources in both Denmark and Israel are of sedimentary origin [NOR 2000, Kovler et al. 2002].

As we can see from Table 1, the EC regulations are different for building products having different thickness, products used in “bulk amounts” and relatively thin products such as superficial materials. The separation between these two groups is not defined precisely, however this approach, even in such a simplified form, is encouraging, because it reflects an attempt of the legislator to take into account the overall mass of radionuclides in dwellings, which is indeed a very important forming the radiation dose. The consideration of the product geometry in the norms is a big step forward in comparison with the most of existing national standards, which still do not address the effect of the product thickness.

At the same time, the EC guidelines and most of the existing national standards still do not consider the density of building products, which is another important component primarily influencing the overall radiation dose of the inhabitants. The first Israeli standard SI 5098 published recently tries to overcome this shortage and provides the information on the maximum allowable activity concentrations of all the three main radionuclides (^{226}Ra, ^{232}Th and ^{40}K) depending on the mass per unit of surface (kg/m^2) of the building products in walls, ceilings, floors, coatings etc. [SI 5098 2007]. In addition to testing the activity concentrations of these radionuclides, this standard requires testing radon emanation of building products isolated from the sides (such a test arrangement simulates conditions of the each unit in the wall of the given thickness with two other dimensions infinite, where the number of radon atoms entering each building unit is equal to that exhaling toward its “neighbor”), and thus the contribution of radon gas into the internal radiation exposure (which is also dependent on the specific surface mass of the product) is taken into account.

According to SI 5098 [2007], the activity concentration index $I = {}^{226}Ra/A({}^{226}Ra) + {}^{232}Th/A({}^{232}Th) + {}^{40}K/A({}^{40}K) + e^{226}Ra/A({}^{222}Rn)$ should not exceed 1 for building products used in bulk amounts and 0.8 for superficial (thin) products, respectively. Coefficients $A({}^{226}Ra)$, $A({}^{232}Th)$, $A({}^{40}K)$

and $A(^{222}Rn)$ are determined from Table 2, e is radon emanation ratio. The radon emanation test is executed on the sample of building product in a special hermetically closed chamber during at least 4 days after preconditioning during at least 1 week under temperature of (20-25)ºC and relative air humidity of 50%±20%.

Table 2. Requirements of the Israeli Standard SI 5098 (2007).

Specific Surface Mass kg/m^2	$A(^{226}Ra)$ Bq/kg	$A(^{232}Th)$ Bq/kg	$A(^{40}K)$ Bq/kg	$A(^{222}Rn)$ Bq/kg
50	952	709	10139	38.2
100	520	379	5541	20.0
150	391	288	4130	14.1
200	319	238	3352	11.0
250	292	217	3028	9.17
300	271	201	2786	7.90
350	255	189	2597	6.96
400	242	179	2444	6.24
450	231	170	2316	5.67
500	221	163	2208	5.20
550	218	160	2166	4.78
600	215	157	2129	4.42
700	210	153	2064	3.85
800	205	149	2010	3.42
900	201	146	1963	3.08
1000	198	144	1922	2.80

Table 2 reflects the fact that for the lightweight or thin building products the dependence of gamma dose on the density or thickness is close to linear, but for the normal-weight products used in buildings in bulk amounts this dependence becomes significantly non-linear, and the effect of density/thickness almost disappears.

3 INDUSTRIAL BY-PRODUCTS INCORPORATED IN BUILDING MATERIALS

The building industry uses large amounts of by-products from other industries. In recent years there is a growing tendency in European and other countries to use new recycled materials with technologically enhanced levels of radioactivity. The most known examples are coal fly ash and phosphogypsum (which is a by-product from phosphorous fertilizers production [O'Brien 1997, Dinelli et al. 1996, Beretka et al. 1996, Smith et al. 2001].

As can be seen from Table 2 adopted from [RP-112 1999], radioactivity concentrations found in fly ash, phosphogypsum and in some other industrial by-products can be often significantly higher in comparison with most common building materials.

Table 3. Typical and maximum activity concentrations in common building materials and industrial by-products used for building materials in Europe [RP-112 1999].

Material	Typical activity concentration (Bq/kg)			Maximum activity concentration (Bq/kg)		
	^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K
Most common building materials (may include by-products)						
Concrete	40	30	400	240	190	1600
Aerated and light-weight concrete	60	40	430	2600	190	1600
Clay (red) bricks	50	50	670	200	200	2000
Sand-lime bricks	10	10	330	25	30	700
Natural building stones	60	60	640	500	310	4000
Natural gypsum	10	10	80	70	100	200
Most common industrial by-products used in building materials						
Phosphogypsum	390	20	60	1100	160	300
Blast furnace slag	270	70	240	2100	340	1000
Coal fly ash	180	100	650	1100	300	1500

Large quantities of coal fly ash are expelled from coal-fired thermal power plants and these may contain enhanced levels of radionuclides along with other toxic elements. More than 280 Mt of coal ash (fly ash and bottom ash combined) are produced annually. About 40 Mt of these are used in the production of bricks and cement [IAEA 2003]. Since most of the process residues further processed into building materials do not meet the required technical specifications, they are typically mixed with pristine raw materials. The net effect is a dilution of the NORM (Naturally Occurring Radioactive Material) content relative to the process residues. In 1996 it has been estimated that up to 15% of phosphogypsum was recycled and that within the European Union some 2 Mt were recycled annually [IAEA 2003]. Its activity concentration depends on the origin and the chemical treatment of the raw material: for example, phosphogypsum from phosphate rocks generally contains considerably higher concentrations of ^{226}Ra than gypsum from carbonate rocks. In any case, not only the ^{226}Ra concentration, but also the radon exhalation from it can be higher than normal.

Blast furnace slag is used mainly as crushed aggregate in concrete as well as a finely ground mineral additive in cement. The activity concentration in slag depends on the ore type, the origin of the raw material and the metallurgic processes. The use of coal fly ash and slag in concrete is a well-recognized source of gamma exposure that is due to the presence of activity concentrations of ^{226}Ra, ^{232}Th and, to a lesser extent, ^{40}K, while its effect via radon exhalation is controversial, due to the low emanation coefficient from the ash [Kovler et al. 2005]. Phosphogypsum used, for example, in the production of plasterboard may give rise to a concern about extremely high concentrations of ^{226}Ra, and high radon exhalation.

Decommissioning or rebuilding the structures made of fly ash, slag, phosphogypsum and the ensuing dust generation or landfilling of secondary wastes may lead to exposure.

These waste materials are used to make a variety of mainly lightweight construction materials. Lightweight building blocks and plasterboard are typical examples with a potential to result in external exposures.

Some regulations address the radioactivity of the waste materials and industrial by-products specifically, but others do not distinguish between building products containing the waste materials and regular building products.

The Finnish Guide [ST 12.2 2005] can serve as an example of the first group of regulations. According to this document, when there are plans to incorporate industrial by-products or wastes in building materials and it is discovered or there is reason to suspect that these contain radioactive nuclides in greater amounts than normal, the activity concentrations of these radioactive nuclides in the final product shall be measured. Where necessary, also other nuclides than ^{226}Ra, ^{232}Th and ^{40}K shall be taken into consideration. If a by-product or wastes containing radioactive nuclides are incorporated in building materials, it must be confirmed that the action level of 1 mSv/year is not exceeded.

The Guide ST 12.2 regulates radioactivity of both building materials and fly ash (including its handling and uses in construction) in one document, which seems to be a useful approach. For example, when ash is added into a material that will be used in building, ST 12.2 sets that the gamma radiation from the contained caesium (^{137}Cs) shall not contribute more than 0.1 mSv per year to the total effective dose of the population due to the material. The action level of 0.1 mSv/year is not exceeded, if the activity concentration of ^{137}Cs in the ash is less than 1000 Bq/kg and the maximum amount of ash incorporated in the concrete is 120 kg/m^3. If the amount of ash incorporated in the concrete is less than 120 kg/m^3, the activity concentration of the ash may be correspondingly higher.

It has to be noticed that the level of 120 kg/m^3 is usually not exceeded in concrete mixes applied in dwelling construction. The high-volume fly ash (HVFA) concrete mixes containing 125-225 kg/m^3 of fly ash [Malhotra 2002] can be cast successfully in pavements and road construction applications, and thus these mixes are not of concern from radiological point of view.

According to the EC guidelines [RP-112 1999], the use of industrial by-products containing natural radionuclides in building materials, which could result in activity concentration indices exceeding the values specified in Table 3, should be justified on a case by case basis by Member States. It is expected that such justification would include non-radiological criteria. For example, some traditionally used natural building materials contain natural radionuclides at levels such that the annual dose of 1 mSv might be exceeded. Some of such materials may have been used already for decades or centuries. In these cases, the detriments and costs of giving up the use of such materials should be analyzed and should include financial and social costs.

4 ADDRESSING RADON EMANATION FROM BUILDING MATERIALS IN THE REGULATIONS

Most of the existing regulations do not address radon emanation from building materials. At the same time, the inclusion of radon emanation test in the standards regulating radioactivity of building materials has both pros and contras, and sometimes is even desirable. In particular, RP-112 recommends considering separate limitations for radon isotopes (radon ^{222}Rn and thoron ^{220}Rn) exhaling from building materials, where previous evaluations show that building materials may be a significant source of indoor radon or thoron and restrictions put on this source are found to be an efficient and a cost-effective way to limit internal radiation exposures.

It is known that the macrostructure, specific surface area of the solid phase and porosity play an important role in radon emanation behavior. Let us consider two extreme cases of radon emanation from building products made of coal fly ash (for example, concrete or masonry blocks) and from phosphogypsum (for example, gypsum wallboard).

For example, the emanation ratio for gypsum from fly ash was found for gypsum as 30%-50%, and for fly ash as less than 1% [Bossew 2003, Stoulos et al. 2004, Kovler et al. 2005]. In other words, the emanation abilities of gypsum and fly ash differ by two orders of magnitude, representing opposite ends of the emanation scale. The emanation power of radon is thus strongly dependent on the microstructure and morphology of the solid particles. It is well known, for instance, that fly ash particle has a dense glassy structure with most of the mass concentrated in the particle shell, preventing radon atoms from escaping the material. In addition, fly ash particles are known for their ideally spherical shape having the minimum surface to volume ratio among all possible particles geometries.

In contrast, gypsum crystals usually are of longitudinal (fibroid) shape with well-developed surface area and have lower density. The typical “layered” structure of gypsum crystal is relatively weak (for example, it easily disintegrates under heating, resulting in the formation of calcium sulfate hemihydrate crystals of high specific surface area, up to 10 m^2/g). All these features make the process of radon release from gypsum relatively easy.

As was mentioned before, radon emanation test is required by the Israeli Standard SI 5098, which includes the dose from radon inhalation in the total dose excess for the inhabitants (0.3 mSv/year). Exposure to radon gas is also addressed in the Austrian Standard ÖENORM S 5200 [Steger 1992]. However, the radon emanation test in this standard is not mandatory, in contrast to the Israeli Standard SI 5098. For the calculation of the activity concentration index I ÖENORM S 5200 allows to use the precondition value e = 10%, if the emanation factor is not known. The real emanation factor can be also determined in the direct experiment, but its value should not be higher than the precondition value. The coefficients A(^{226}Ra), A(^{232}Th) and A(^{40}K), which consider external radiation exposure, and the coefficient A(^{222}Rn), which is responsible for radon inhalation in the final dose criterion, are 1000, 600, 10000 and (0.15 + e ρ d)/1000, respectively, where ρ is the density and d is the wall thickness. The precondition values for the wall thickness are d = 0.3 m, for the density ρ = 2000 kg/m^3 and for the emanation factor e = 10%. It can be seen that the part of this combined dose criterion responsible for gamma exposure does not depend on the density and geometry of the building element, however the part responsible for radon inhalation does.

The comparison between the two standards addressing radon emanation properties, SI 5098 and ÖENORM S 5200, shows that the Austrian Standard, which is guarantees that inhabitants of dwellings do not receive a higher dose from natural radioactivity as 2.5 mSv/year, is several times more liberal, than RP-112 and other national standards.

On one hand, knowing the real radon emanation properties of the product tested in the laboratory makes the decision by legislators more accurate. On the other hand, there are still difficulties with recommending an optimum standard test procedure for getting the results reliable and reproducible. For example, it is well-known that moisture of the porous building products significantly influences the radon exhalation rate. That is why the climatic conditions at the sample preparation/curing and during the radon test itself should be chosen as stable as possible. There are also other factors, which can influence the emanation test result, for example, the dimensions of the sample and radon chamber, temperature, test duration, method of the approximation of “exhalation rate – time” dependence needed to calculate the emanation ratio, age of the materials, changing there properties in time (cementitious materials, for example). As a consequence, the radon-dependent part of the dose criterion is more uncertain, which should be compensated by more liberal criterion (similar to “safety factors” accepted in the structural design). This correction is a part of a non-radiological justification of the regulations, which should be based on technological, social, environmental and economical considerations. At the same time, such non-radiological criteria are seldom applied in the legislation practice, because of the difficulties related to the methodology of cost-benefit analysis.

5 HOW STRICT SHOULD REGULATIONS BE?

It has to be emphasized that RP-112 requires that the activity concentration index should be used only as a screening tool for identifying materials which might be of concern. Any actual decision on restricting the use of a material should be based on a separate dose assessment. Such assessment should be based on scenarios where the material is used in a typical way for the type of material in question. Scenarios resulting in theoretical, most unlikely maximum doses should be avoided. The purpose of controls is to restrict the highest individual doses. Therefore, the dose criterion used for national controls should be chosen in a way that the majority of normal building materials on the market fulfill the requirements. Therefore, measurements of activity concentrations are needed only in case where there is a specific reason to suspect that the dose criterion for controls might be exceeded. At the same time, Member States should require, as a minimum, the measurement of types of materials which are generically suspected.

As an example of the complicated situation with applying a standard, which does not take into account these financial and social costs, would be a publication of the first version of the Israeli standard SI 5098 in the year 2002. The author of the present paper served as a member of the Standardization Committee and was involved in the preparation of this standard. As simple calculations showed, even regular building products, such as ordinary Portland cement concrete, some types of ceramic tiles, different types of light-weight masonry blocks, granite and some other products, hardly met the norms. The first version of this standard was widely discussed by public and governmental authorities, before the decision was made to start the revision, shortly after the first version of the standard has been published. The standard SI 5098 has been revised in December 2006, and its revised version is implementing nowadays in the local construction industry, while the concrete and prefabricated concrete products are exempted from the requirement to meet the controls criterion till the end of the year 2007. It has to be noticed that even after the revision, the standard SI 5098 seems to be still the strictest one among the standards regulating radioactivity of building materials. The standard does not exempt any group of building products of mineral origin from execution of controls, even those resulting in relatively low doses. In addition, its criterion is not only based on the lowest end of the dose scale (0.3 mSv/year), but also includes a dose from radon inhalation into this value. In the end of the year 2007 the results of the standard implementation in the local construction industry will be discussed by the Standardization Committee, and the decision regarding a possibility to keep the strict criterion further will be made by the authorities.

6 CONCLUSIONS

In recent years there is a growing tendency to use new recycled materials with technologically enhanced levels of radioactivity (e.g. phosphogypsum, coal fly ash, slag, etc.). Many of them are valuable industrial by-products having a potential to be re-used in construction, however the problem of their contaminants has to be addressed. In view of this, there is a need to develop and introduce in both international and national levels environmentally safe and economically reasonable standard regulations, which should be based on justified radiological, social and economical legislation concepts.

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