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Review

INVESTIGATION OF THE POSSIBILITIES OF PHOSPHOGYPSUM APPLICATION FOR BUILDING PARTITIONING WALLS – ELEMENTS OF A PREFABRICATED HOUSE

Miloš B. Rajković and Dragan V. Tošković

Phosphogypsum is a waste product in the manufacture of phosphoric acid from phosphorite and sulphuric acid by so–called "wet process" and represents a refuse that is as such simply thrown away. Phosphogypsum which is produced by "dihydrating procedure" contains not only various impurties but also radionuclides, which limits its construction use.

Performed testings point to the complexity of phosphogypsum structure and composition while the electron microscope's pictures showed its different crystal composition compared to the natural gypsum. The calcined and refined phosphogypsum can be used for partition walls manufacture. To avoid the danger of the possible presence of radionuclides it is better to use the mixture of natural gypsum and phosphogypsum for this purpose. Substantial saving of materials and economic effect can be achieved in this way.

The analyse performed in this work have shown significant presence of radionuclides in phosphogypsum. Gammaspectrometric measurements of radioactivity have determined substantial radioactivity of phosphogypsum. Using the maximum tolerated level values that are legally accepted, as well as equations to calculate indexes of tolerated radionuclide presence, an index of 2.23 has been determined for interiors and 1.13 for exteriors, i.e. 0.64 in case of roads. On the basis of the maximum tolerated level of radioactive building materials contamination (< 1), ***forbiden is*** *the use of phosphogypsum in interiors,* ***allowed*** *(≈ 1) in exteriors and roads.*

The tests of heavy metal components in phosphogypsum have proved their presence in the amounts potentially producing consequences if present in closed spaces. That is conditioned by the phosphogypsum quantity, as well as by the area of space partitioned.

KEY WORDS: phosphogypsum, gypsum fiberboards, partitioning wall, radon, radioactivity, "building sickness", habitability

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INTRODUCTION

One of the most serious contemporary diseases in the world is **carcinoma**, the initiation of which modern medicine still cannot explain completely, although everything indicates that this disease has been present on our planet from ancient times. On the other hand, the science proved that **carcinoma** appears as a consequence of the presence of harmful substances related to the *syndrome of sick building (house)*, such as radioactivity, heavy metals, evaporation of lead compounds, as well as contamination (pollution) of the environment: air, water and food.

It is somewhat of an absurd that the contemporary man, in spite of achievement of high standards in the field of construction and maintainance of buildings, as well as in the field of sanitary habits, health and work culture and living culture, still finds himself in a position to suffer in his everyday life, from the agents of various nature and origin that exert harmful effects on his health.

For a long time it has been considered that building materials do not have any influence on human health, so raw materials from industrial wastes with high contents of natural radioactivity and toxic chemical substances were used for their production. The main criterion for choice of a raw material was the negligible price, on one hand, as well as the removing of that waste from the environment because it pollutes the environment (surroundings), on the other hand. Most advocates of environmental protection supported inclusion of industrial waste into building materials.

Industrial wastes, utilized for building and between-storey constructions are:

- alum slate;
- phosphogypsum, by-product obtained in phosphatic acid production by "wet procedure"
- "red mud" - by-product obtained in aluminium production;
- sludge from blast furnaces of steel mills and power plants;
- "flying ash" produced by coal combustion.

Apart from the industrial wastes, for construction (fabrication) of apartments use is also made of some materials that are harmful for people, or those that contain hazardous substances beyond maximal permitted values. These are primarily materials used for surface adaptation of rooms (artificial paints and polishes, etc.).

The most famous, long utilized materials for construction are cement asbestos products. **Asbestos** is a common name for fibrous silicate materials that have a high resistance to high temperatures, to agressive influence of chemical reagents, good hardness under pressure and bending, as well as good waterproof characteristics. Asbestos is used in combination with other materials (cement and other additives), and its content in individual products ranges between 20 and 70%. It is used in production of building materials, prefabricated elements, furniture, PVC panelings, masses for drought prevention, etc.

Each year there appear in the world market approximately 100.000 new chemical substances that may affect human health. As a consequence of their action, there occur health problems in human organisms, but also real, very serious illnesses. One of the most spread illness connected with the "syndrome of sick house (building)" is alergy. At the end of 60-ties of the XX century, the number of people affected with alergy amounted to 1%, while that percent at the end of 1992 increased to 25% of the total Earth population, with a tendency of still faster increase. Contemporary medicine proved that alergy is a basis for the development of a great number of disorders, including the most serious ones.

Sometimes these are less marked and more limited episodes, while sometimes the health of great number of people occupying the space in individual buildings is threatened. From such tragic experiences, during last decades of the XX century, there arises a disastrous conclusion that living in a building may lead to the occurrence of illnesses, which is very much in contradiction with basic functions of a building and rooms within it-living, resting, working, protection from unfavourable weather influences, etc. (1-8).

A previous paper (20) dealth with the possibility of utilization of purified phosphogypsum for prefabrication of partitioning wall that may be used in the construction industry. Also, there were explained its characteristics from the aspect of physico-chemical properties. In this paper, the aim was to show the application of partitioning wall made of phosphogypsum from the aspect of building biology (1), that is of the influence of the built-in phosphogypsum boards on the appearance (or aggravation) of the syndrome of "sick building".

DWELLING AS A HUMAN NEED (10)

During the history of human race, the notion of dwelling, as its physical realization, experienced great transformations, both in form and structure, and in size and equipment. There is a long way from a sheltered space under a tree or in a cave (during the time of ecological balance), through deepened holes in rocks or soil (the first greater human activities in space rearrangement), houses built of mud and wattled fence, wooden pile dwellings, then houses of stone, wood, bricks, concrete and iron, from houses spread over the ground level, dug into the ground, to skyscrapers with unlimited number of storeys!

For a wider and more general idea of dwelling - living in certain space, there is in use the expression *habitability.* This notion is of Latin origin and is derived from the Latin word *habitatio*, which means domicile (the same notion used in ecology to designate the most favourable place for a certain species of plant or animal origin). In the professional Anglosaxon literature, there is a common expression *habitat* (habitability), which corresponds to our notion - habitation, human dwelling, human settlement. The expression *habitability* was for the first time introduced by Benton McKay in 1928, searching for an appropriate term for the space favourable for human settlements for the purpose of regional (space) plans. Habitable is each natural or artificial space which provides to people appropriate external natural conditions for their continual existence in their search for food, shelter, favourable climate, various physical, natural conveniences of the space (vicinity of water, ground configuration, communications, etc.). Habitability included also some new - ecological factors of the environment: soil quality from the aspect of its fertility, climate and anthropogenic factors - changed space, formed as a product of planning (that is, purposeful human activity intended for the future).

In prehistoric period, habitability represented a condition determined by purely natural factors, because the man did not change or amend the surroundings he lived in. During further development of human civilization and as an important developmental process, construction technology of dwelling objects eliminates unfavourable environmental factors or amends them in an artificial way (heating of living quarters, light, water suppy, ventilation, cooling, draining pipes for feces and other wastes, etc) - that is, makes them habitable by construction technology.

In the meantime, ecological awareness has been gradually increasing, so, many measures and regulations were proscribed based on ecological way of thinking. Various kinds of considerations are present: quality of the environment within the quarters for shorter or longer stay during the day. Architecture and architectural objects are not viewed any more as the art and the objects of art, *but as human settlements of ecological kind - eco-systems, and the buildings as habitats.* In them, there is not only commercial quality - the standard of living the thing that is important, but also the ecological quality of the environment, the one that guarantees unpolluted life conditions.

Ecological quality of living

Ecological quality of living equally implies external and internal factors of the environment.

a) External ecological factors

- air and surface water quality;
- speed of supplying with drinking water of good quality;
- existence, quality and quantity of vegetation cover;
- quality, esthetics and climate of the area;
- direction and power of air currents and precipitations;
- insolation;
- the state of hygiene of settlements and towns;
- vicinity of sources of noise and other environmental pollutants (thermo-electrical and industrial plants; deposits of dispersed wastes, parking places and highways);
- level of underground waters;
- quality of the grounds and foundation - soil on which the settlement or a building will be positioned (from the aspect of geological composition, engineering-geodesical and hydrological conditions, seismical activities, etc.);
- configuration of the ground (relief);
- vicinity of natural maintained areas;
- elements relevant for the level of urban noise and the traffic as a source, type of communication lines (street, rail corridor, airport), type of vehicles, street width, type and heigth of the buildings in the street, base (asphalt, concrete, cobblestone), green plantations, size of the objects, type of openings, etc.
- organization of municipal services;
- neighbourhood (social aspect);
- number of storeys and vicinity of adjacent objects;
- number of overground and underground storeys, etc.

b) Internal ecological factors

Internal ecological factors include the microclimate in an object for living or in some other where a great deal of time is spent (hospital, school, hotel, office, factory, etc.), among which the most important is air quality, that is, all the things that influence formation of the so-called "air comfort". Undoubtedly, at present, except the choice of a location, microclimate and "air comfort" depend much on the choice of building material.

General internal factors that influence ecological quality of living, and eventually the health of inhabitants of the quarters, may be tentatively classified within five main groups:

- presence of radioactivity;
- utilization of some harmfull (for human health) building materials;
- presence of electrical and magnetic field in the quarters;
- influence of the heating system;
- influence of external factors in the quarters (noise, for instance).

SANITARY ASPECTS OF THE APPLICATION OF A BUILDING MATERIAL

In medicine, all the manifestations linked with appearance of an illness or some other disturbance of human health, are commonly divided into two categories:

1. **Specific illnesses connected with staying in buildings** (*Specific building related illnesses*, SBRI). By specific illnesses connected with staying in closed space (buildings) there are designated medically distinct, clinically or biochemically confirmed illnesses, where there is a clear link (on the basis of pathophysiological mechanism) with a known cause, such as: immunological mechanisms, infection (stimulation), intoxication and cancerogenesis.

2. **Syndrome of "sick (unhealthy) buildings"** (*Sick building Syndrome*, SBS). The notion of sick building was introduced in 1983, by experts of World Health Organization (WHO), as a substitute for up till then utilized terms, such as: *building sickness*, *insulated building illness*, *office building syndrome*, *tight building illness*, etc. Syndrome of "sick buildings" is defined as a health disorder which is not medically defined as an illness, where there exist only unspecific, sometimes only undefined subjective problems, without objective apparent signs.

Syndrome (a word of Greek origin, *med.* a set of symptoms that appear simultaneously in certain number of illnesses, association of the signs of an illness) **of "sick buildings"** according to the World Health Organization which investigated building quality from the aspect of their influence on human health for a long period, may be applied to the following types of buildings:

- the ones that are neglected, ruined, forgotten, left to a bad maintainance, with rotten and rusted construction elements, inclined to fall down, etc.
- the ones in bad sanitary conditions, with neglected rooms or neglected surroundings, without basic hygienic predispositions, without drainage system, without good

drinking water, with bad project (or without any project, or project made without consulting an expert), insufficiently shaded or insolated, with badly mounted isolation or without any isolation, with inadequately solved heating of the quarters, etc.

- the ones in apparently good (physical) condition, even esthetically acceptable, but containing within them elements harmful for human health, and because of that considered "sick" (10).

Syndrome of "sick buildings" is of benign nature in relation to the overall health of a person, but it significantly decreases the working ability of employees, or the well being of other inhabitants of a building. It is characterized by the existence of one or more symptoms, which are grouped by the World Health Organization in the following way, and which disappear after removing the person from the contaminated surroundings:

- irritation of mucous membranes of nose, eye and throat, like itching, pricking or sensation of dryness;
- neurotoxical, or general effects: headache, excessive lethargy or increased irritability, disturbed concentration;
- symptoms on the skin: itching, dryness, red colouring of skin;
- other symptoms are seen less frequently: sensation of heaviness in chest, audiable hissing during breathing, dry cough, sensation of sickness and unusual taste in the mouth.

However, in a number of studies on this problem, the most common are eight classical symptoms of "sick buildings":

1. Headache;
2. Blocked nose (difficulties in breathing through the nose);
3. Throatache;
4. Itching and/or watering of the eyes;
5. Lethargy (indifference, lack of working enthusiasm);
6. Dryness of throat;
7. Dryness of eyes;
8. Dry skin (with or without dandruff);

THE POSSIBILITY OF PHOSPHOGYPSUM APPLICATION FOR PREFIBRICATED BUILDING ELEMENTS

In comparision with conventional (classical) partitioning walls (built of bricks or blocks), prefabricated constructions of partitioning wall are more convenient and are given the priority. With equal soundproof, thermic and fire prevention qualities, as well as with substantially lower mass, their mounting is possible in various buildings, while a quick dry building in shortens the time of the work and cuts the expences, especially when there is a production of vast series (11).

Empty interior of a partitioning construction is ideal for quick and cheap installation of all kinds of ventilation, and dry and flat wall surface may be immediately painted, covered with wall paper or tiles. Depending on the type of a construction, partitioning walls have the mass (surface weight) between 25 and 50 kg/m^2 of the partition area (as a reference, mass of a partitioning wall of autoclaved gasconcrete is between 50 and 80 kg) (12,13), which significantly lowers the loading of constructive elements of a building. These walls are most often built with a subconstruction of metallic profiles, but also small wooden beams with cross-section area of 6 x 6 cm may be built in.

Partitioning walls may be single- of double-layered, and are paneled with one or two layers of gypsum boards. Construction is easily adjusted to the requirements of statical and constructive physics, so the most favourable properties are achieved by appropriate combination of subconstruction, gypsum board and isolator incorporated in the empty interspace. Besides, all the elements are easily adjusted to the plan of the building and the requirements of the space, and in the case of rearrangements, prefabricated partitionings are easily removed.

The basis of partitioning walls is a gypsum core covered on both sides by a special high quality cardboard. Gypsum is used:

- because it has neither smell nor any constituents harmful to human health, because the important properties are provided by preparation and production through an ecologically clean procedure. Due to the porous gypsum core, partitio-ning walls regulate air humidity in the room;
- gypsum boards are fireproof, also due to good properties of the gypsum core. Gypsum binds approximately 22 % of crystal water, which amounts to 3 dm^3/m^2 in boards with 15 mm thickness
- building materials with a low heat conductivity (e.g. wood, gypsum) produce a feeling of warmth and comfort.

For the production of partitioning boards there are used either natural gypsum, which is obtained by a "dirty technology " which represents a problem from the aspect of the environmental protection (ISO standards of 14.000 order), or chemical gypsum - phosphogypsum, which is obtained as byproduct during the production of *green* phosphatic acid by dihydrate, so-called "wet procedure", which is the only one applied in our country (14):

$$Ca_{10}(PO_4)_6F_2 + 10\,H_2SO_4 + 20\,H_2O \xrightarrow{75-80^\circ C} 6\,H_3PO_4 + 10\,\underbrace{CaSO_4 \times 2\,H_2O}_{phosphogypsum} + 2\,HF$$

In spite of a great similarity between the chemical and natural gypsum, the latter being $CaSO_4{\times}2H_2O$, this by-product (waste product, secondary product) is a burden and, considering the present impurities, moisture and different crystal form, is mostly left, after separation of the acid, on deposits near the factory for phosphatic acid production where it "ages", or is poured into natural water flows (river or sea). Because of enormous quantities of the phosphogypsum, which is produced in the ratio 5:1 against the main product - phosphatic acid, phosphogypsum deposits occupy a big area and may in time contaminate surroundings, soil and air (15).

Considering that phosphogypsum deposits represent a serious ecological problem, and, on the other hand, because of its similarity with natural gypsum, phosphogypsum represents a potential raw material for the application in construction industry, the aim of this paper was to investigate the possibilities of application of phosphogypsum in the production of partitioning walls, which is currently done in the whole world on a large scale. The most common procedures are: Babcock-BSH process, Kossatz-Bison semidry process, Schenk process, Siempelkamp/Fermacell process (16), Würtex process (17), Bison-Werke process (18), Rauma-Repola Onoda process (19), Rajković et al. (20), etc.

With phosphogypsum application (in large quantities) in construction industry, among other things, for fabrication of partitioning walls, two problems would be solved: *firstly*, the phosphogypsum deposits near factories for the production of phosphatic acid would be smaller or could even be removed, which would be a great contribution to environmental protection, and *secondly*, instead of digging out of natural gypsum, which is also a "dirty technology", there is offered a possibility of utilizating a cost-free substitute.

The aim of the present paper was to perform an analysis of partitioning wall built of phosphogypsum instead of natural gypsum and to compare the properties of such two walls. Also, as phosphogypsum contains radionuclides that originate from phosphates, the aim was to measure radioactivity of the partitioning wall made of phosphogypsum and to check its suitability for incorporation into apartments, because of radon that is liberated by radioactive decomposition of uranium.

MATERIALS AND METHODS

All the investigations were carried out with phosphogypsum which was obtained as a by-product in the production of *green* phosphatic acid by dihydrate, or so-called "wet procedure", which is applied in the chemical industry in Prahovo (IHP Prahovo) by a technological procedure in which raw phosphates are treated with sulphuric acid (14).

In spite of the great similarity with natural gypsum, $CaSO_4 \times 2H_2O$ (21-24), this byproduct is a burden and, considering impurities, moisture and different crystal shapes that are present in it, in general it is deposited, after separation of acid, in the vicinity of the factories for phosphatic acid production, where it "ages" or is poured into natural water recipients (river or sea) (25-31).

Phosphogypsum purification was carried out by the following procedure (32-34): solution of sulphuric acid in water was made, with concentration of 28%, which was heated up to the temperature of 90°C. Phosphogypsum was added to the solution and an emulsion was made with continual stirring. After phosphogypsum, $BaSO_4$ was added also with continual stirring, until the temperature reached 85°C. Then the mixture was cooled. After cooling, it was filtered through a special Buchner funnel, separating the solid from the liquid phase. Grinding of the purified phosphogypsum was carried out by laboratory mixer to the particle size mainly between 100 and 200 μm, with only 12% particles bigger than 200 μm.

Phosphogypsum and natural gypsum samples were analyzed using scanning electron microscopy (SEM), JSM–84OA, JEOL, Japan. X–ray diffraction analysis was carried out using the diffractometer for powder SIEMENS D–500 with Ni–filtered CuK_a radiation. Identification of crystalline phases in recorded samples has been carried out by position and intensity comparison of diffraction profiles with JC PDS data.

Gammaspectrometric analysis of phosphogypsum samples was performed as follows: phosphogypsum samples were homogenized, dried at 105°C (for 6 hours) and put into a container (marinelli) of appropriate geometric shape and kept closed airtight (30 days) in order to achieve radioactive equilibrium. Gammaspectrometric measure-ments were performed by three pure germanium detectors manufactured by EG&G "ORTEC", Germany, with the efficiency of 25-30% and energy resolution 1.75-1.95 keV (12). The detectors were connected to a multichannel analyser by the same manufacturer and to corresponding computer equipment. Energy calibration, as well as calibration of detector efficiency was performed by radioactive standard supplied by Amersham. The measurement time for one sample was 60.000 to 100.000 s, and the basic radiation was measured after 250.000 s.

Measurements of total activity were performed by α–β anticoincidental proportional gas counter ("COUNTERMASTER") with basic radiation of 1 imp/min. Planchet radius was 2.3 cm. Counter efficiency amounted to 24% and was determined by a standard of ^{90}Sr.

Determination of heavy metals content was performed by the next procedure (9): One gram of finely ground phosphogypsum was weighed into Erlenmeyer flask (100 or 150 cm^3), 20 cm^3 of concentrated nitric acid was then added and the contents of the flask were gently boiled for 2 to 3 hours. There was a small funnel on the top of the flask. After boiling with conc. HNO_3, the vessel was removed from the hot plate to cool the content. Hydrogen peroxide (3 cm^3 of 30% solution) was then added and boiling was continued for 15 minutes. The digested material was transferred into volumetric flask (50 cm^3) using distilled water. The volume of the liquid in the flask was adjusted to exactly 50.00 cm^3 with distilled water. At the end, the liquid in the flask was filtered through filter paper (*Whatman No.41*) and the filtrate was collected in a suitable reagent bottle. The solution obtained after the digestion was used for the determination of heavy metals in phosphogypsum. The following heavy metals were determined: iron, lead, zinc, cadmium, manganese, chromium, nickel and copper. The concentrations of the investigated elements in standard solutions were within the following limits (in μg/ml): Fe 0–20; Mn 0–10; Zn 0–2; Cu 0–5; Co 0–3; Pb 0–5; Cd 0–2; Ni 0–4.

All the determinations were performed using atomic absorption spectrophotometer Varian SpectrAA–10/20 Spectrophotometer Plus (Varian Australia Pty, Ltd, Springvale Rd, Mulgrave, Victoria, Australia).

The determination of other phosphogypsum components was carried out using dried samples. The following analytical methods were used for the determination of the analyzed components:

- P_2O_5 – spectrophotometric method with ammonium molybdate reagent;
- CaO – gravimetric method;
- Mg – gravimetric method;
- Al – spectrophotometric method with 8–hydroxyquinoline (oxine).

RESULTS AND DISCUSSION

Manufacturing of gypsum-cardboard boards

Gypsum-cardboard boards are utilized for mounting of partitioning walls, for wall and ceiling paneling and as dry plaster. For manufacturing of gypsum-cardboard boards, the standards presented in Table 1 were used.

The construction of partitioning wall made of gypsum-cardboard boards filled with phosphogypsum, is represented in Figure 1, and the board external appearance in Figure 2.

Table 1. Wall and ceiling gypsum cardboards filled with phosphogypsum

Standard thickness (mm)	Standard length (mm)	Standard width (mm)	Weight (kg/m^2)
Gypsum-cardboard boards			
12.5	2000–3000	1250	10.0
15.0	2000–3000	1250	12.0
Fireproof boards			
12.5	2000–3000	1250	10.5
15.0	2000–3000	1250	13.0
Impregnated gypsum-cardboard boards			
12.5	2000–3000	1250	10.0
15.0	2000–3000	1250	13.0
Impregnated fireproof boards			
12.5	2000–3000	1250	10.5
15.0	2000–3000	1250	13.0

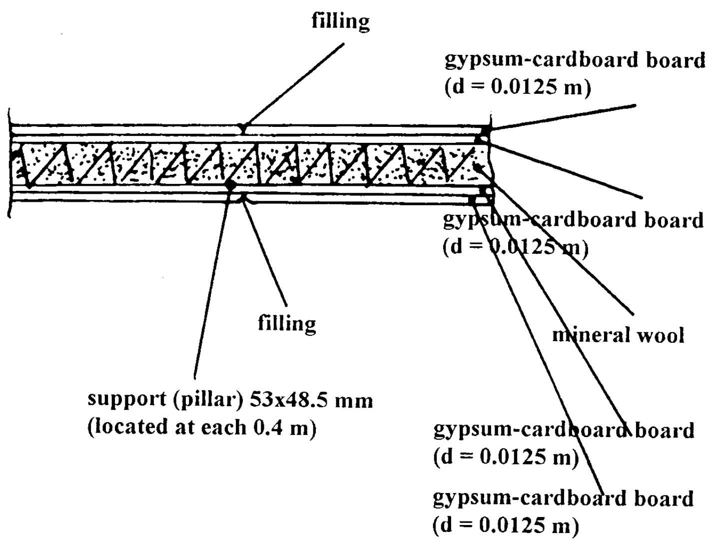

Fig. 1. The scheme of partion wall construction

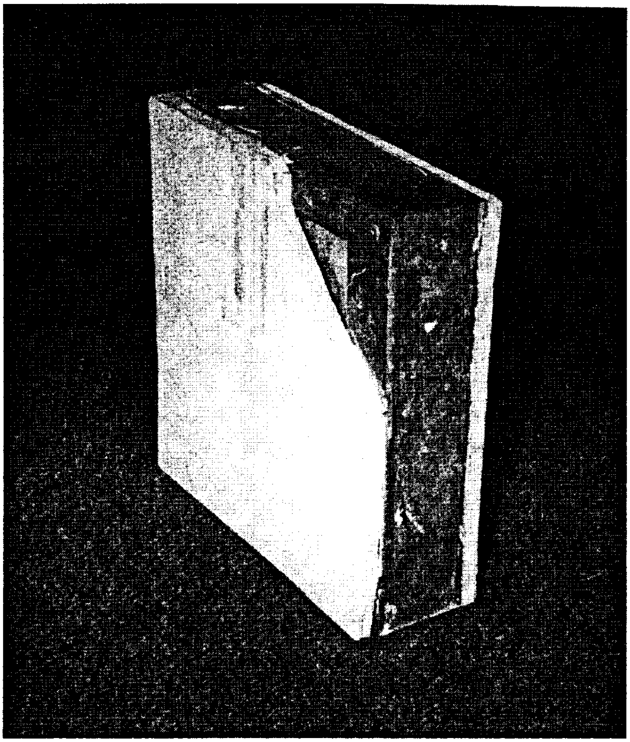

Fig. 2. The outlook out of partion wall made of gypsum cardboard filled with phosphogypsum

For partitioning walls filling, instead of natural gypsum, phosphogypsum was used, previously purified by the proposed procedure, described in our previous work (35,48), and the obtained results are presented in Table 2.

Chemical and mineralogical composition of phosphogypsum

The results given in Table 2, show that natural gypsum is basically a hemihydrate, $CaSO_4 \times 1/2\ H_2O$, and phosphogypsum is a dihydrate, $CaSO_4 \times 2H_2O$ (14,47). By the suggested procedure of purification and calcinization (32), phosphogypsum - dihydrate is transformed into the form most similar to the natural gypsum which contains 77.26 wt.% $CaSO_4 \times 1/2\ H_2O$ (natural gypsum has 81.34%). Because of this, it is thought better to apply the process of α-hemihydrate production, by "hemihydrate" instead of classical "dihydrate" procedure for obtaining of phosphogypsum of greater purity and better utilizability. So, a procedure was suggested which requires a longer burning (36). Other results show that the composition of purified phosphogypsum is very similar to natural gypsum and that in essence they are the same substance. The presence of some substances, e.g. P_2O_5 and Fe_2O_3 which are absent in natural gypsum, is not important for application in the construction industry, but may be very useful for the application of phosphogypsum in agriculture, e.g. for amelioration of solonetzes (37).

Table 2. The chemical and mineralogical composition of phosphogypsum

Chemical composition	Phosphogypsum (in wt. %)
$CaSO_4 \times 2H_2O$	11.54
$CaSO_4 \times 1/2H_2O$	77.26
$CaSO_4$, anyhidride	–
Bonded (fixed) water, H_2O	7.21
$CaSO_4$, total	81.59
CaO, free	0.29
SO_3	–
Sum:	89.09
Free water, at 45°C	1.03
Calcination loss	0.60
SiO_2 + insoluble residue	4.55
$MgCO_3$	–
Na_2O	–
K_2O	–
Al_2O_3+Fe_2O_3	0.17
MgO	–
$CaCO_3$	4.48
P_2O_5	0.09
Sum:	10.92
Total:	100.01

The photograph show the morphology of the crystal particles of phosphogypsum material. Phosphogypsum has a markedly crystalline structure, with mainly of rombic and hexagonal crystals, which indicates that it has a more complex composition than natural gypsum, which is in accordance with the literature data (38).

Fig. 3. The SEM picture of phosphogypsum crystal structure (×1000)

All the complexity of phosphogypsum originates from its crystal structure, which eventually influences its chemical behaviour. X–ray diffraction analysis was carried out by comparing the position and intensity of diffraction profiles with the JC PDS datas. The following crystalline phases were identified for phosphogypsum: $CaSO_4 \times 2H_2O$ (33–311); $CaSO_4 \times 1/2H_2O$ (33–310) spectra corresponded to natural gypsum (23).

The results of determination of heavy metal content in phosphogypsum are represented in Table 3.

Table 3. The content of heavy metals in phosphogypsum (ppm or mg/kg)

Heavy metal	Fe	Pb	Zn	Cd	Mn	Co	Ni	Cu
Content of heavy metal	785	45	45	7	8	10	20	17

The results of gammaspectrometric analysis of phosphogypsum, utilized for filling of gypsum-cardboard boards, are represented in Table 4.

Table 4. Gammaspectrometic analysis of phosphogypsum

Radionucludes	Radioactivity (Bq/kg)	Upper permitted limits for interiors	Upper permitted limits for exteriors	Upper permitted limits for roads
^{226}Ra	439	MAL = 200	MAL = 400	MAL = 700
^{232}Th	8.7	MAL = 300	MAL = 300	MAL = 500
^{40}K	8.7	MAL = 3000	MAL = 5000	MAL = 8000
Radionuclides synthetic origin	< 1.0	MAL = 4000	MAL = 4000	MAL = 2000
Index for interiors	2.23	MAL ≤ 1	–	–
Index for exteriors	1.13	–	MAL ≤ 1	–
Index for roads	0.64	–	–	MAL ≤ 1

MAL – Maximum admissible limit of radioactive building materials contamination (39)

On the basis of the general criteria that are to be obeyed when chosing a building material, the most marked feature is radioactivity (1). Radioactivity is investigated as the contents of three radioactive isotopes: radium-226, thorium-232 and potassium-40. Biologically harmful (life-threatening) evaluation of these three isotopes has the following relations (40):

$$^{40}K : ^{226}Ra : ^{232}Th = 1 : 12.5 : 16.50$$

Upper permitted limits of radioactivity are calculated according to the so-called Summary formula which has been formulated by National commission for protection from radiation of the former USSR, and according to our currently valid regulations (39) it has the following form:

$$\textbf{Index for interiors} = \frac{^{226}Ra\,(Bq/kg)}{200} + \frac{^{232}Th\,(Bq/kg)}{300} + \frac{^{40}K(Bq/kg)}{3000} + \frac{V_{interiors}(Bq/kg)}{4000} \leq 1$$

$$\textbf{Index for exteriors} = \frac{^{226}Ra\,(Bq/kg)}{400} + \frac{^{232}Th\,(Bq/kg)}{300} + \frac{^{40}K(Bq/kg)}{5000} + \frac{V_{exteriors}(Bq/kg)}{4000} \leq 1$$

$$\textbf{Index for roads} = \frac{^{226}Ra\,(Bq/kg)}{700} + \frac{^{232}Th\,(Bq/kg)}{500} + \frac{^{40}K(Bq/kg)}{8000} + \frac{V_{for\ roads}(Bq/kg)}{2000} \leq 1$$

In the opinion of the majority of experts, there is no negligible level of radioactive radiation; only one radioactive disintegration is enough to cause mutagenic change, that is, the appearance of cancer cells. Lowering of the dose only brings about the lowering of the probability of the illness appearance.

The obtained results of the investigation of radionuclide content indicate a significant presence of radium-226 isotope of 439 Bq/kg. This fact is in accordance with the literature data on the values of radium-226 isotope content in various forms of phosphogypsum (41,49), which range between 430 and 790 Bq/kg.

The presence of radium-226 isotope is dangerous because of the liberation of (the only) radioactive gas radon (isotope-222). Radon, which is produced by radium degradation and the half-life of which is 3.8 days, gets accumulated in the atmosphere of closed rooms (especially if these rooms are isolated to cut the heating expences). Radon is α-emitter and induces much more serious damage to living tissue when it is inhaled and the radioactive products of its decay are deposited in lungs._For that reason, the main cause of danger from radon is considered to be α-radiation. At radioactivity equilibrium, 1 g of Ra corresponds to $3.7 \cdot 10^{10}$ Bq Rn. It is a quantity of radon within which there occur $3.7 \cdot 10^{10}$ decays/s, that occupies the volume of 0.66 mm^3 and has the mass of 6.5 mg. In comparision with other radioactive gases, radon has a significantly longer half-life. During decay, radon yields at first a sequence of solid radioactive particles which emit α-, β-particles and γ-quant. Radiation danger at the contact of human organism with radon is connected with its radioactive descendents: $^{218}_{84}Po$, $^{214}_{82}Pb$, $^{214}_{83}Bi$, and $^{214}_{84}Po$, which mostly damage the lungs. These isotopes are called "short-lived radon descendents", which are responsible for the dose of radiation received by human lungs (42,43).

Radon is inhaled and expired by people, it does not remain in the breathing system, but the products of its radioactive decay adhere to all materials, and even to air particles. When they are inhaled, they remain in the lungs and constantly emit the radiation.

There is no a generally accepted scientific view on how small quantities of radon affect human health, but one thing is sure: radon in high quantities is certainly harmful, but for the low ones, there are no valid data. This is one of fundamental scientific problems that has not been solved yet. **It actually amounts to the question on the risk from "small doses". It is an old scientific controversy: according to some authors - each dose is harmful, even the smallest received dose may have disastrous conseqeunces. With the lowering of the dose, the probability of a harmful effect decreases, but does not disappear, untill the dose decreases down to zero.**

The International Commision for Protection from Radiation (ICPR) recommended the so-called "intervention level" (or "action level") for already existing contingent of buildings. Most countries accepted the level of **200 Bq/m^3** and incorporated that in its legislative regulations.

Influence of toxic chemical substances in building materials

Chemical toxins (toxic substances) are substances which can, in comparatively small quantities, destroy a tissue, organ or the whole organism. The conditions for expression of a harmful effect of some substance are nowhere so favourable as in closed dwelling space:

- with decreased ventilation in a closed space, a toxic substance (toxin) may be concentrated in the air, and it remains there for a longer period;
- great number of artificial building materials utilized in contemporary construction, contain more or less chemically-toxic additives. These toxic ingredients that are present in certain materials, have an ability of very prolonged evaporation, giving to the working and living space a toxic potential, which lasts as long as the material itself;
- there is practically no protection from the influence of these toxins, except to limit the stay in the closed space, which is not always feasible, or to substitute the critical materials with those that are biologically acceptable and healthy.

Current limit values of toxic substances for the air in a closed space are calculated by several methods, the most important of which are:

1. MAC - method of maximal concentration of toxic chemical substances in the air of a closed dwelling space.

   Materials that cause cancer, in the MAC list, are classified as follows:

   A - unequivocally designated as an instigator of cancer,
   A1 - materials that may cause malign tumours in man,
   A2 - materials which appeared dangerous only for animals,
   B - materials for which there exists a reasonable doubt of being cancerogenic.

2. IARC - method of International Institute for Scientific Investigation of Cancerogenic Diseases

According to the method elaborated by this institution, cancerogenic materials are divided in the following way:

1 Cause carcinoma in man,
2A Probably cause carcinoma in man,
2B Possibly cause carcinoma in man,
3 Do not cause carcinoma in man.

In current legislation, there are no specifications according to the type of a compound in the environment, but there has been such attempts for the working environment, in the Federal Bureau for Standardization in 1991 (44). In Table 5, there are presented the maximal concentrations (MAC) in the atmosphere of places of work and working sites for individual compounds of heavy metals through the standard value and the range of values.

Most developed countries have established by legislation the standards of air quality, which are based on reccomendations of the World Health and World Meteorological Organizations. According to OECD expert recommendations (45), there are introduced **unit risks of cancerogenity, UR.**

Unit risk of cancerogenity is individual risk for exposition of the concentration of individual pollutant in the air of 1 μg/m^3 during a life span (70 years) and represents the probability to obtain cancer, which does not always imply a **fatal outcome**. For arsenic, for instance, that value is 0.0043, that is, 1:230 for a life span.

Table 5. Allowed concentrations of heavy metals in the air in the working rooms (44)

Current numbers	Metals and specifications	The range of values
66.–70.	Sb and its compounds	0.3–2
77.–81.	As and its compounds: C, S	0.05–0.25
93.–98.	Cu and its compounds	0.1–4
121.	Be: C	0.002
211.–216.	Zn and its compounds	0.01–10
483.–485.	Fe and its compounds	0.8–10
581.–588.	Cr and its compounds: C, S	0.01–0.5
628.–630.	Cd and its compounds: C	0.05–0.1
633.–636.	Sn and its compounds: K	0.05–0.1
664.–667.	Co and its compounds: C, S	0.05–0.1
692.–704.	Mn and its compounds	0.2–5
812.–816.	Ni and its compounds: C, K	0.007–1
862.–869.	Pb and its compounds: C, K, S	0.5–0.5
965.–971.	Se and its compounds	0.2–10
11015.–1018.	Tl and its compounds	0.2–10
1161.–1165.	Hg and its compounds: K, S	0.005–0.1

Captions: C - cancerogene, **K** - resorbed through the skin; **S** - increases susceptibility and irritability

UR values for individual heavy metals are given in the Table 6, where the values are applied to the particles the radius of which is shorter than 2.5 μm and which are called respirable particles.

Table 6. Maximum allowed concentrations of heavy metals in the 24 hours samples of ambiental air (46)

Heavy metal	Concentration	Unit risk, UR
As	2.5 – day's value	4.3×10^{-3}
Be	–	2.4×10^{-3}
Cr(VI)	0.2 – day's value	1.2×10^{-2}
Cd	10 – middle month's value	1.8×10^{-3}
Mn	1000 – middle month's value	–
Ni	2.5 – middle month's value	2.8×10^{-4} (NiS)
Pb	1000 – middle month's value	–
Hg	1000 – middle month's value	–

On the basis of the results of investigation of heavy metal content in phosphogypsum, causality between heavy metal presence and the consequences on human health can not be established for sure. The thing that is certain is that a reliable control over toxic substances, such as are most heavy metals, must be provided. Consequences for the man in the environment are a function of their level in the air, of the time of exposition and of synergetic action of several pollutants.

As environmental protection includes prevention, not only reclamation, the presence of heavy metals should be prevented and thus their influence on man's working atmosphere completely eliminated (or at least decreased to a neutral level).

CONCLUSIONS

Phosphogypsum obtained by "dihydrate procedure" contains, in addition to impurities, also radionuclides which limit its application in construction. Investigations performed in the present paper pointed out the complexity of phosphogypsum structure and composition, while electrom microscopy photographs showed different crystal composition of phosphogypsum in comparision with natural gypsum. Due to the presence a large number of impurities which originate from the initial raw material and because of chemical mode of processing, the utilization of phosphogypsum requires its additional purification and calcination.

By the procedure of purification, phosphogypsum radionuclide content remains approximately the same, but it approaches natural gypsum by its physico-chemical properties. By a longer drying, at the temperature of 105°C, there is obtained a stabile phosphogypsum form, α-hemihydrate, which is by its chemical characteristics very similar to natural gypsum. As the presence of some inpurities is not limiting for its further application, thus purified phosphogypsum, with further finer grinding till reaching the particle size of 100 μm, represents a suitable substitute for natural gypsum.

Calcinized and purified phosphogypsum may be utilized for manufacturing of gypsum-cardboard boards, which may subsequently be utilized for mounting of partitioning walls. The quality of partitioning wall made of phosphogypsum did not differ from the quality of partitioning wall made of natural gypsum. In dependence of the function, gypsum mass for gypsum filling of gypsum-cardboard boards with various functions ranged from 10 to 13 kg, which means that by this application, vast (at present) deposits of phosphogypsum may be utilized. To remove completely the danger from the possible presence of radionuclides, it is better to utilize a mixture of natural gypsum and phosphogypsum. Thus, the waste of natural material would be greatly decreased and a full commercial effect would be achieved, both from the aspect of environment protection and recycling of waste materials.

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ИСПИТИВАЊЕ МОГУЋНОСТИ ПРИМЕНЕ ФОСФОГИПСА ЗА ИЗРАДУ ПРЕГРАДНОГ ЗИДА – ЕЛЕМЕНТА МОНТАЖНОГ ОБЈЕКТА

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Фосфогипс добијен "дихидратним поступком" садржи, осим нечистоћа и радио-нуклиде који ограничавају његову примену у грађевинарству.

Испитивања су указала на сложеност структуре и састава фосфогипса, док су снимци на електронском микроскопу показали морфолошки изглед кристала фосфогипса. Услед присуства великог броја нечистоћа које своје порекло воде из основне сировине - фосфата и због хемијског начина прераде, употреба отпадног фосфогипса захтева његово додатно пречишћавање и калцинисање.

Испитивања су указала на присуство радионуклида у фосфогипсу. На основу измерене радиоактивности гамаспектрометријском методом, утврђена је радиоактивност фосфогипса. Применом Закона дозвољених МДК вредности, као и једначина за израчунавање дозвољеног присуства радионуклида, утврђен је индекс за ентеријер 2,23, за екстеријер 1,13 и за путеве 0,64. На основу максимално дозвољене границе радиоактивне контаминације грађевинског материјала (< 1), **забрањује се** употреба чистог фосфогипса за ентеријер, а **дозвољава се** (≈ 1) за екстеријер и за путеве, што је **препоручено** да би се утрошиле огромне количине отпадног фосфогипса које леже на депонијама. Интенција у свету је да се количине фосфогипса на депонијама што више смањују да не би утицале на животну средину.

На основу испитивања садржаја тешких метала у фосфогипсу, утврђено је њихово присуство, али у количини која може довести до појаве последица услед присуства у затвореном простору. Та каузалност условљена је количином фосфогипса као и величином простора који се преграђује.

Калцинисани и пречишћени фосфогипс може се користити за израду гипскартонских плоча, које се затим могу употребити за израду преградних зидова. Квалитет саграђеног преградног зида у потпуности одговара квалитету зида изграђеног од природног гипса, међутим, да би се потпуно уклонила опасност од евентуално присутних радионуклида, за израду је боље да се користи мешавина природног гипса и фосфогипса која у значајној мери анулира штетно присуство по животну средину радионуклида у фосфогипсу и своди радиоактивност тако изграђеног преградног зида на нормални фон. На тај начин остварила би се велика уштеда у грађевинском материјалу а са аспекта заштите животне средине постигао пун економски ефекат.

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