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Utilization of phosphogypsum from industrial dumps as an element of environmentally safe energy- and resource-conserving technologies

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Abstract. The paper discusses the problem of disposal and processing of phosphogypsum dumps as an element of environmentally safe energy- and resource- conserving technologies. The process of impact and possibility of phosphogypsum pre-treatment with weak electric fields to improve its physical and mechanical properties was studied. Two samples of different origin phosphogypsum, which are located in dumps on the territory of the Kamianske City (Ukraine), were used as research material. The research was conducted on samples of the following fractions: 1.0–2.0, 0.4–1.0, 0.1–0.4 mm. Further, balls were formed from the treated phosphogypsum and raw materials to study their compressive strength. The experiment was performed on a pellet strength meter, which operates in the range of 0–2.5 kgf/grain. Phosphogypsum balls, treated with electric current, did not collapse with the maximum values of the device. According to the research results, it was established that preliminary treatment with a low electric current leads to a decrease in the dehydration temperature of phosphogypsum, and the subsequent hydration allows to obtain a material with higher compressive strength properties. This substantiates the potential possibility of involving research results to obtain a cheaper product and will allow to liquidate multi-ton deposits of phosphogypsum.

1. Introduction

Utilization and processing of industrial phosphogypsum dumps, which are minor products of the mineral fertilizers production, is an important task of environmentally safe technologies of energy and resource conservation [1–3]. There are several long-range directions for its further use in various sectors of the economy. It stands to mention the effectiveness of using phosphogypsum as an ameliorant in agriculture [4–6] to improve the condition of the soil cover and as a bonding agent in the construction industry [7–9].

In current times, significant amounts of this material occupy large land areas and effect negatively on the natural environment [10]. Dumps and phosphogypsum sludge storages are considered as industrial deposits of construction and related raw materials [11], the use of which will reduce significantly energy costs for their extraction and save natural resources. The extraction of rare earth elements from phosphogypsum is a promising direction of utilization [12]. An analysis of the potential possibilities of phosphogypsum processing in view of environmental problems and modern technologies [13] indicates its wider use in the building industry. Phosphogypsum is used as a bonding agent and a kind of filler [14]. The possibility of complex use of phosphogypsum and building waste as infilling from hydrated cement is being actively studied. Various studies and tests on the strength and sedimentation of new materials and fillers are being conducted [15].

At the same time, in order to increase the intensification of the phosphogypsum processing from industrial dumps on the basis of energy and resource conservation, it is necessary to continue the study of its changes in physical and mechanical properties under the influence of various factors. Since in objects such as industrial dumps, processes, similar in form and substance to processes of sedimentagenous mineral and ore formation, must occur inevitably [16]. Thus, the regularities obtained during the study of natural deposits of mineral raw materials can be applied to industrial dumps of phosphogypsum with a high confidence.

Therefore, the relevance of this issue can be resolved with setting up a series of experiments to study changes in temperature, physical, mechanical and strength properties of phosphogypsum for further use in the building industry.

The scientific literature presents studies of the impact of electric current on the reductive-oxidative properties of phosphogypsum or the processing of phosphogypsum using electrolysis [17]. The given research results relate to the problems of extracting useful components, but the issues related to the change in the physical and mechanical properties of the substance for use in building purposes remained unresolved.

In the work [18], the authors use plasma technology for processing phosphogypsum. The main task is the removal of toxic elements contained in industrial raw materials. The experimental installation consists of such main components as: furnace, heat exchanger, filters and other equipment. At the same time, the current to ensure the efficient operation of the reaction chamber ranges from 100 to 160 A, and the voltage is 220 V. But the issue of changing the physical and mechanical properties has not been resolved fully, and the reason for this may be in objective difficulties associated with a significant part of the expenditure in terms of energy resources. Ensuring such operating parameters is quite expensive and energy-consuming. Therefore, for the conditions and purposes of research, there is a need to find cheaper and simpler laboratory equipment.

In order to obtain an extra strong material from phosphogypsum and improve its mechanical properties, the authors [19] offer another option for overcoming the relevant difficulties. The goal is achieved by adding spent zeolite to adsorb phosphate impurities and applying stamping to increase compressive strength. At the same time, additional elements also increase the cost of performing this type of work.

In work [20], the authors studied the influence of natural and electric and thermal fields on the processes of hydrothermal-metasomatic ore formation. The given research results indicate that under the combined influence of electric and thermal field factors, there is a migration of the substance in samples and test portion, destruction and recrystallization of existing minerals. Under the impact of this process, the formation of more advanced crystalline forms are made, as well as the formation of new mineral species and aggregates which were not characteristic of the original geologic material. Also, researchers [21] established the phenomenon of mobilization and migration of bound and crystalline water, which occurs at temperatures by 150–250 °C lower than during simple heating. In this regard, taking into account the time factor, it can be assumed that the presence of electric currents and elevated temperatures in the industrial phosphogypsum dump leads to certain changes; in particular, mineral, structural and, probably, chemical transformations of material at different depth levels.

The generalization of the analytical review of published scientific studies shows the following. Over the last decade, the number of publications has increased significantly (by 4 times) where various technical and organizational solutions for the environmentally safe disposal of phosphogypsum are investigated [22]. All this gives reason to confirm that it is reasonable to carry out a study devoted to the issue of its processing and further use, because a rather small number of them relates specifically to studies of the influence of weak electric currents on the physical and mechanical properties of phosphogypsum.

The aim of this work is an experimental study of the effect of electric and thermal fields on the physical-chemical and mechanical properties of phosphogypsum for its further safe reclamation as a substance that generates large-tonnage waste and is a source of environmental problems for the surrounding areas.

Achieving the set goal is ensured by solving the following problems:

– to establish initial parameters, granulometric composition and moisture content of the studied raw materials;

– to analyse the change in temperature and electric current during processing of the researched phosphogypsum samples;

– to evaluate changes in the physical and mechanical properties of phosphogypsum after pre-treatment with weak electric current.

2. Research materials and methods

Phosphogypsum obtained during the production of mineral fertilizers in the city of Kamianske (Ukraine) was chosen as the material for the research (figure 1). Samples weighing 5 kg each were taken from two sources. The first sample was taken from dumps that have been covering the tailings storage facility of the Prydniprovsk Chemical Plant for a long time [23]. The second sample is “fresh” phosphogypsum – a waste of current production.

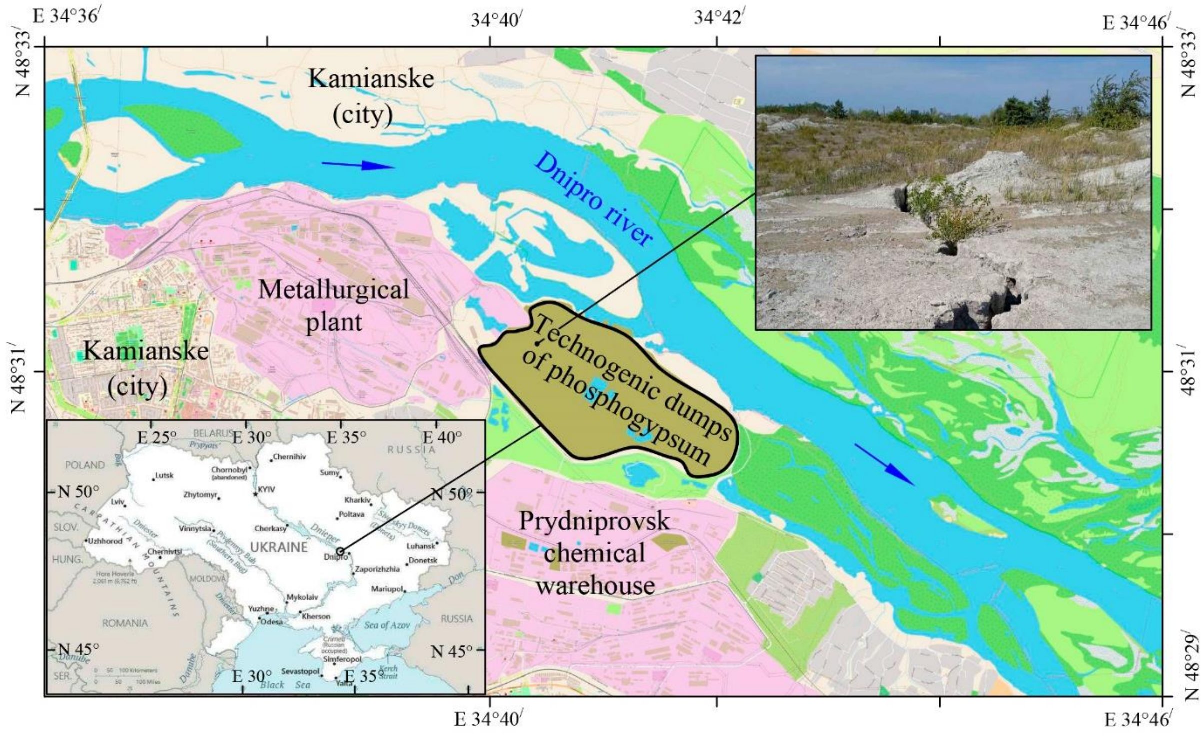

Figure 1. Overview map of the location of the research object.

The samples are quite different from each other. Thus, the test portion from the first sample has a grey-dirty-white colour, contains several plant residues, is quite lumpy, dry by touch (figure 2, a). Apatite concentrate, which was supplied during last century from the deposit of the Kola Peninsula or other objects, was the raw material for this phosphogypsum. Sedimentary phosphorite concretions imported from the African continent was the material for modern phosphogypsum (the second sample). Visually, it is a greyish-creamy disintegrated product that is moist by touch with a characteristic smell of raw materials (figure 2, b).

a

b

Figure 2. Appearance of phosphogypsum: a – phosphogypsum from the first sample (apatite concentrates from the Kola Peninsula); b – phosphogypsum from the second sample (phosphorite concretions from the African continent).

During the determination of the moisture content of the samples, 10 g test portions were taken. They were ground into powder and placed in pre-weighed porcelain crucibles. The crucibles were then placed in a chamber drier. The temperature in the chamber was kept at 120–130°C for 120 minutes. Every half hour of the experiment, the crucibles were weighed and the loss of mass, which was associated with the removal of excess moisture, was determined.

Experimental research was provided with the following installations and devices. At the first stage, an installation was designed (figure 3) for passing a small electric current through a sample of phosphogypsum, which consisted of:

1) direct current source VA-12 with a potential difference of 15.71 V;
2) working chamber made of porcelain container;
3) digital ammeter and voltmeter for measuring current and voltage.

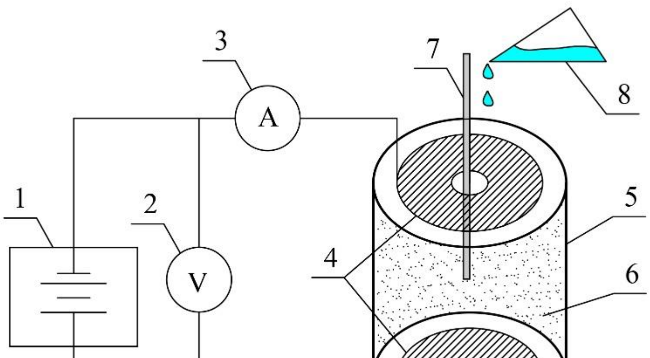

Figure 3. Installation scheme for conducting research on the treatment of phosphogypsum with electric current (author's development): 1 – current source; 2 – voltmeter; 3 – ammeter; 4 – electrodes; 5 – parcel cup; 6 – phosphogypsum; 7 – thermometer; 8 – water.

The experimental chamber is a laboratory chemical thin-walled porcelain beaker with an internal diameter of 35 mm and a total volume of 100 ml. A steel electrode in the form of a disk was placed at the bottom of the container to which an insulated copper wire was soldered. The location of the lower electrode excluded the possibility of its touching the walls of the beaker. A sample of phosphogypsum weighing 10 g was poured onto the electrode. A second electrode, which is similar in material and design to the first one, but contains a technological hole for adding water, was placed on top of the phosphogypsum. Distilled water in the amount of 6 ml was added to the sample through the hole in the centre of the electrode to create a conductive circuit between the electrodes. The potential difference was given taking into account the polarity, which changed in each of the conducted experiments. Additionally, a mercury thermometer with a graduated scale up to 100°C was fixed through the hole in the upper electrode in the phosphogypsum sample. Measuring devices (ammeter and voltmeter) were connected to the electrical circuit, and their readings were taken every 5 minutes simultaneously with the sample temperature values. The value of the electrical resistance of each experimental sample was calculated based on the obtained values of laboratory studies. The entire time of the experiment was about one hour. After that, the current supply was stopped and the fixation of values was ended. The research was conducted on samples which were divided into the following fractions: 1.0–2.0 mm, 0.4–1.0 mm, 0.1–0.4 mm.

Tests of the strength of the balls were carried out on the pellet strength meter (figure 4). This is a stationary laboratory device of cyclic action, designed to measure the breaking force of carbamide granules when determining their static strength in accordance with National State Standard 21560.2-82 in the laboratory "Dniproazot". The device is registered in the State Register of Measuring Instruments and allows measurements in the range of 0–2.5 kgf/grain.

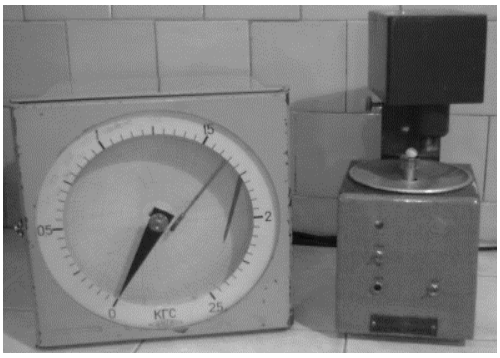

Figure 4. Appearance of the device for measuring the strength of pellets.

3. Results and discussion

3.1. Setting the initial parameters, moisture and granulometric composition of phosphogypsum

The phosphogypsum samples were dried for 10 days at a room temperature of 18 °C and an air humidity of 55–60%. Then they were classified according to different size fractions. The results of granulometric analysis showed that the phosphogypsum from the second sample is characterized by larger aggregates than the phosphogypsum from the first one. This is connected with the different duration of storage in dumps, differences in the chemical and structural composition of raw materials and the different ability of the material to aggregate. In experiments to determine the moisture content of various samples, such differences in the granulometric composition determined both the different natural moisture content of phosphogypsum and the nature of their loss of bound water. The results of the experiments are shown in figure 5.

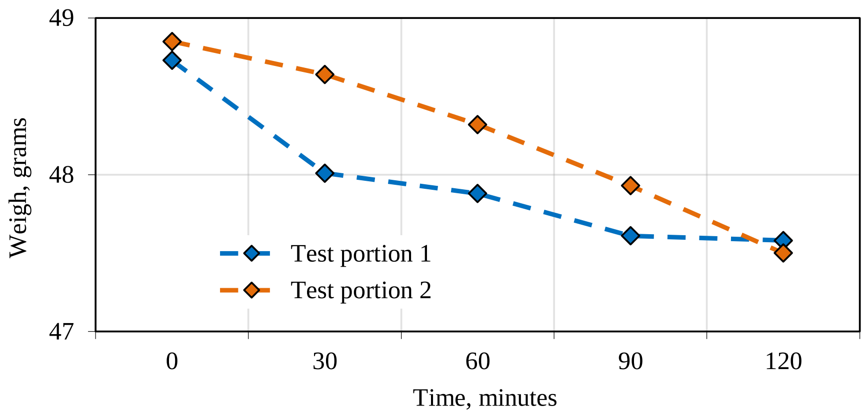

Figure 5. Dependence of change in weight in different samples of phosphogypsum the drying time.

From the obtained data it can be seen that the removal of moisture from different samples of phosphogypsum proceeds differently. This may indicate a different crystal structure of the material. Moisture calculations showed that the phosphogypsum from the first sample has a moisture content of 10.5%, and the phosphogypsum from the second sample has a moisture content of 12.3%.

3.2. Study of changes in temperature and electric current during processing of phosphogypsum

A peculiarity of the experiment when an electric current is passed through a phosphogypsum sample is a significant heating of the samples (sometimes up to 60°C). The form of the heating curve in all repetitions of experiments is approximately in the form of a parabola (figure 6). The nature of the change in the current during the experiment has a similar appearance and varies from 10 mA to 320 mA (figure 7).

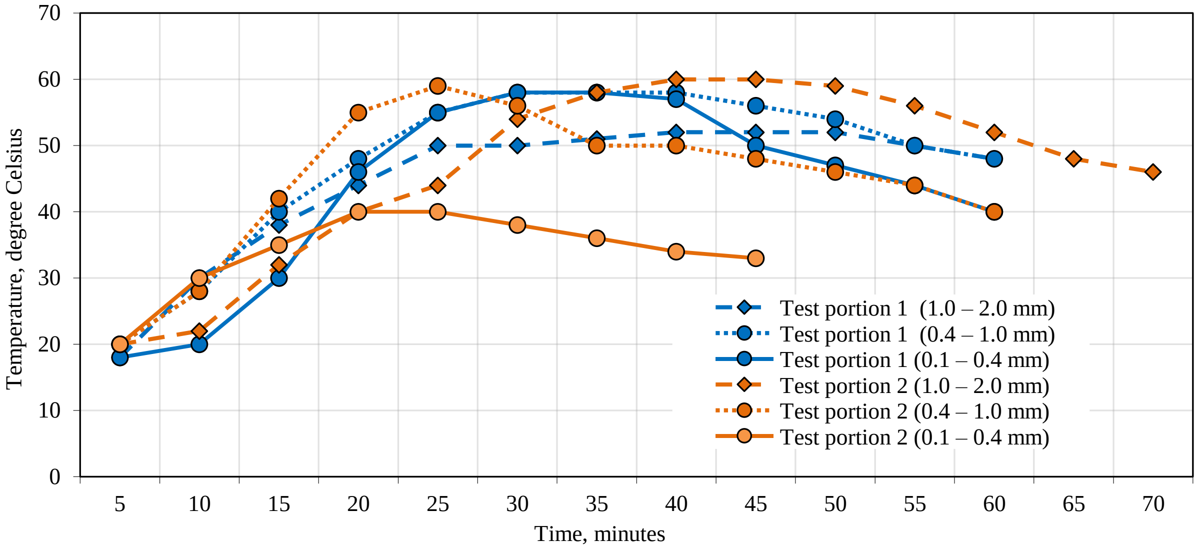

Figure 6. Dependence of temperature change on time of electric current supply for different fractions of phosphogypsum.

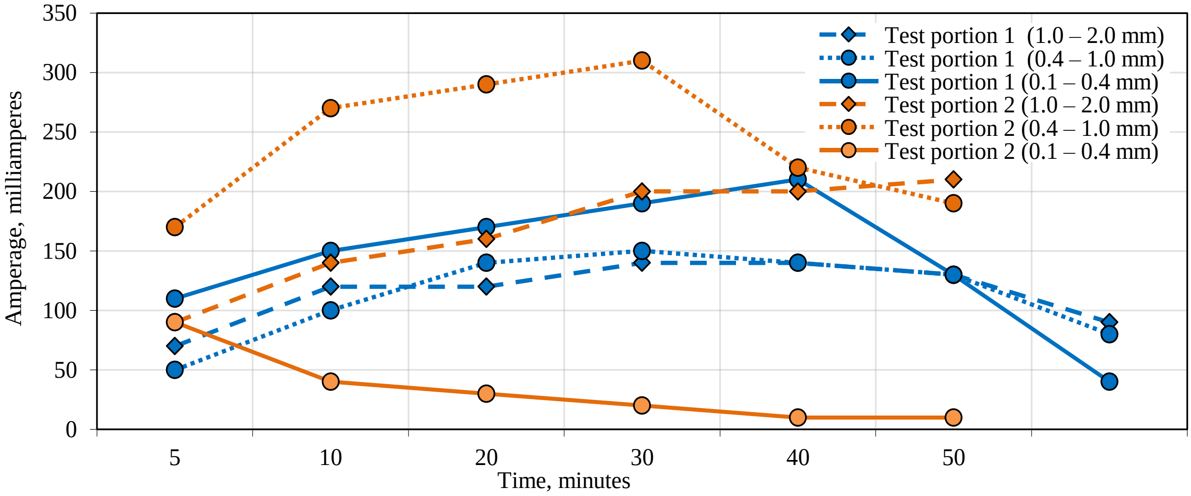

Figure 7. Dependence of change in current strength on time when passing through different fractions of phosphogypsum.

The analysis of changes in temperature and current strength in different samples of phosphogypsum shows that the material from the first and second samples differs significantly in terms of conductive properties. The reasons for this have not been established yet and require further research.

3.3. Study of changes in the physical and mechanical properties of phosphogypsum

In order to evaluate changes in the physical and mechanical properties of phosphogypsum, a study of its compressive strength properties was conducted. For this purpose, the material of all the samples that passed the experiments, as well as the material of the initial samples of phosphogypsum, underwent a two-hour firing in a muffle furnace at a temperature of 500°C. As a result, all samples of phosphogypsum turned into powder with a dust-like structure. Then, 1 ml of water was added to 2 g of powder of both samples with a fraction of 0.4–1.0 mm, and the same burnt material of the original samples, balls were rolled and left to harden at room temperature for two days.

According to the results of the ball strength test, the following was determined on the pellet strength meter. The ball, which was formed from burnt raw phosphogypsum from the first sample, collapsed at values of 1.9 kgf/grain. The ball formed from the burnt treated phosphogypsum from the second sample reached values of 2.2 kgf/grain before destruction. Balls made of calcined phosphogypsum treated with a weak electric current did not collapse at all when the device reached the maximum effort of 2.5 kgf/grain.

3.4. Discussion the research results of impact of weak electric fields on changes in the physical and mechanical properties of phosphogypsum

A number of examples of direct use of phosphogypsum in various branches of the national economy without important technologies for its preparation are known. In particular, phosphogypsum is very effective for amelioration of salt marshes and swampy soils, so it was widely used for these purposes in the South of Ukraine [24]. However, the weak point of the specified technical solution for the utilization of phosphogypsum was the fact that the issue of the cost-effectiveness of production and sale of finished products was not fully and objectively studied.

In addition, the theoretically waste-free utilization of phosphogypsum is achieved by implementing the technology of complete thermal decomposition of calcium sulphate to CaO and SO_2. Gases containing SO_2 are processed into sulphuric acid, and calcium oxide is released in the form of activated lime or as part of portland cement clinker. It is known that some technologies are implemented at very high temperatures up to 1450°C. In addition to high energy consumption, the need occurs for large-scale supplies of blast furnace slag and additional raw materials for obtaining cement clinker [25].

Research on the processing of phosphogypsum into gypsum bonding agent contributed to the development of important normative documents and state building standards of Ukraine. However, research and industrial tests of the technology could not be carried out due to the fact that phosphogypsum could not be add without special preparation as an additive to the mill for grinding a mixture of clinker and slag.

Thus, the problems of high energy consumption, economic practicability and difficulties with the preparation of raw materials, these results did not allow to solve fully the issue of phosphogypsum utilization. This becomes possible owing to the change in the physical and mechanical properties of phosphogypsum by a weak electric current.

The peculiarity of the proposed approach to the utilization of phosphogypsum from industrial dumps as an element of environmentally safe energy- and resource-saving technologies is as follows. The possibility of application and impact of weak electric currents on changing the physical and mechanical properties of phosphogypsum is shown. Carried out research concerns phosphogypsum of various origins (from apatite concentrates of the Kola Peninsula and phosphorite concretions from the African continent). The application of the proposed solutions is limited by the value of electric current passing through the phosphogypsum sample in the range from 10 mA to 320 mA. The disadvantage of this method is the rather significant heating of the samples (sometimes up to 60°C), which justifies the need for further research using specialized industrial equipment.

The analysis of the obtained laboratory research results shows that the dependence of the temperature change of phosphogypsum samples on the duration of the experiment can be divided conditionally into three zones (figure 6). Initially, during the first 20 minutes of the experiment, there is a rapid increase in temperature from 18 to 60°C. During the next 30 minutes of the experiment, the temperature of the samples remains almost unchanged, and after an hour it begins gradually to decrease, which indicates the termination of the process of physical and chemical interaction of phosphogypsum with electric current.

The dependence of the change in the current strength on the time of the experiment has a slightly different form of the graph (figure 7). For the most part, a constant increase in current strength from 50–

100 to 200–300 mA is noted for all fractions during the first half hour of the experiment, and then a significant decrease. For the fraction of 0.1–0.4 mm of the phosphogypsum second sample, an uncharacteristic shape of the curve was recorded, which is completely different from all other variants of the experiments.

The fact that the strength properties of phosphogypsum change after electric current treatment is important. The formed balls withstood the maximum device load of 2.5 kgf/grain, while the untreated phosphogypsum balls collapsed in the compression range of 1.9–2.2 kgf/grain. This indicates certain structural transformations and changes in the physical and mechanical properties of phosphogypsum.

Treatment of phosphogypsum with an electric current with a small potential difference causes an increase in its strength properties and can open wide ways for the use of this material in the production of building materials. This substantiates the further reasonability of researching the effect of electric fields on changing the properties of phosphogypsum, primarily for use in the construction industry as a bonding agent. Taking into account the huge volumes and speed of accumulation of industrial phosphogypsum reserves in Ukraine and all over the world and its ecological danger, this way of utilization it as a waste product may prove to be quite promising.

4. Conclusion

As a research result, the following conclusions were obtained:

1. For the two investigated samples, it was determined that apatite concentrate from the deposits of the Kola Peninsula (the first sample) and sedimentary phosphorite concretions from the African continent (the second sample) were the raw material for phosphogypsum. Differences in the granulometric composition determined the different crystalline structure of the material, different natural humidity of phosphogypsum and the nature of the binding water loss. Research has established that the phosphogypsum from the first sample has a moisture content of 10.5%, and the phosphogypsum from the second sample has a moisture content of 12.3%.

2. It was established that with passing a weak electric current of 12 V through phosphogypsum samples, heating of the samples to 60°C is observed, and the heating graph always has the shape of a parabola. At the same time, the nature of the change in current strength has a similar feature and varies from 10 mA to 320 mA.

3. According to the research results on changes in physical and mechanical properties and strength tests, the following was established. A ball made of burnt untreated phosphogypsum from the first sample collapsed at values of 1.9 kgf/grain. The ball of calcined processed phosphogypsum from the second sample collapsed at 2.2 kgf/grain. Balls made of treated with electric current and burnt phosphogypsum of both samples did not collapse at all when the device reached the maximum effort of 2.5 kgf/grain.

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