



# Lithosphere Protection with Hardening Phosphate-Containing Soil-Cement Technology

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## Abstract

This research is relevant in the light of a need to protect lithosphere from pollution. A solution to this problem can be the exercise of a geocoprotective function by using building and construction technologies. Phosphate-containing bonding agents usually have low solubility in water. They are made of aluminosilicates that occur in common clay. These minerals react with phosphoric acid to release phosphates and silica gel with absorption properties. The purpose of this research is to make fake rocks from phosphate clay material, intended for building and construction purposes, and to investigate their longevity. The material used was treated to neutralize and detoxify the mobile ionic contaminants and heavy metal compounds, as well as petroleum pollutants, so that at the end we could get construction material like fake rocks or improved aggregates that would be good for the environment. These cement systems are called the geocoprotective systems. The leading methods that were applied were thermodynamic and experimental studies of service and geocoprotective properties of phosphate-containing cement systems, more specifically fake landscape rocks. Selected studies lasted for more than 10 years, and were carried out in accordance with the guidelines, adopted in the Russian Federation. This research shows that phosphate-containing cement systems made from contaminated clay/sandy artificial soils have long-lasting properties like strength, frost resistance, water resistance and contaminant binding. From this perspective, they are good for building and construction. For example, they can be used to improve the contaminated soil on road construction sites by detoxifying it until water-resistant products, and to make long-lasting water-resistant solutions from local raw materials that would be good for waste storage.

**Keywords:** geocoprotection, phosphate-containing cement systems, physico-chemical process, properties of phosphate clay material, detoxification

## 1. Introduction

The lithosphere's self-cleaning capacity is extremely low, so it removes less pollutants than gets from other sources. Therefore, the consequences of lithosphere pollution are progressive, and will reach the point soon, when they become non-reversible. The amount of fertile soils is gradually declining, which means that agriculture industry will fall into disarray. Forest and sea pollution will result in the extinction of animal and fish species [23].

The main consequences of lithosphere pollution threatening the environment today are [24]:

- terrain changes;
- activation of dangerous geological processes (karst erosion, landslides), subsidence and rock shifting;
- changes in physical fields, especially in permafrost areas;
- Chemical soil pollution and mechanical soil disturbance.

The problem of lithosphere preservation is linked to three facts of our time: natural resources are overused with a 2.5 coefficient instead of 1, lithosphere is being polluted (for example, the amount of lead ions discharged into the ocean is over 600 thousand tons) and there are no alternative technologies that would stop the practice of over consumption [1-3]. Therefore,

lithosphere preservation is to do, among other things, with restoration and decontamination of organic and non-organic impurities. The other thing linked to lithosphere preservation is the process of material decontamination until a product that would be good for the environment, such as a protective fake rock with long-lasting properties. The latter is no less important than soil detoxification because it serves a perfect solution to the problem of utilizing contaminant-free material by adding it into hardening cement systems intended for making fake rock covers and boulders. In this light, we not only utilizing the material, but also putting it into a meaningful context [4-6]. In general, the idea of a construction material going through natural hardening and turning into a fake geocoprotective rock is illustrated by the following guideline (Figure 1).





**Fig.1:** Lithosphere Protection by Detoxifying Contaminated Soil and Hardening Construction Product that is good for the Environment

We assume our method is environmentally protective because it allows removing detoxification products and mineral contaminants from the hardening cement system. Besides, fake rocks can serve as mineral vessels for storing organic contaminants like petroleum products. Thirdly, our geocoprotective system, which are contaminant-free fake rocks, can be used in construction as a cover, landscape item or even a building. Our research is guided by the Lithosphere Protection Guideline (Figure 1), and the research object are phosphate-containing cement systems, made from clay/sandy soils contaminated with petroleum products and heavy metal ions, that were put into reaction with phosphate acid to produce fake rocks. According to data available from [1, 4, 6], phosphate-containing cement systems have at least 3 properties that are good for lithosphere protection. Firstly, free ions are detoxified through the formation of sparingly soluble phosphates and heavy metal ions. Secondly, aluminosilicates react with phosphoric acid to release silica gel with absorption properties. Thirdly, fake rocks can be made by making the mineral powder to react with phosphoric acid.

Table 1 shows solubility product values for sparingly soluble phosphates and heavy metal ions, and related ion concentration in a saturated solution. Aside from that, the Table provides data on the maximum permitted concentrations (MPCs) of heavy metal ions in soil.

**Table 1.** Properties of Sparingly Soluble Phosphates

Free Ion	Phosphate	Solubility Product	Me(II) and Me(III) Concentration in Saturated Solution		MPC of Ion in Soil, g/kg
			mol/L	g/L	
1	2	3	4	5	6
Pb(II)	Pb <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub>	7.94·10 <sup>-43</sup>	1.49·10 <sup>-10</sup>	3.08·10 <sup>-8</sup>	20(32)·10 <sup>-3</sup>
Cu(II)	Cu <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub>	1.26·10 <sup>-37</sup>	1.63·10 <sup>-8</sup>	1.03·10 <sup>-6</sup>	3,0·10 <sup>-3</sup>
Ni(III)	Ni <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub>	5.01·10 <sup>-31</sup>	3.41·10 <sup>-7</sup>	2.01·10 <sup>-5</sup>	4·10 <sup>-3</sup>
Zn(II)	Zn <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub>	9.12·10 <sup>-33</sup>	1.53·10 <sup>-7</sup>	~1·10 <sup>-5</sup>	23·10 <sup>-3</sup>
Cd(II)	Cd <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub>	2.51·10 <sup>-33</sup>	1.18·10 <sup>-7</sup>	1.32·10 <sup>-5</sup>	0.5·10 <sup>-3</sup>
Fe(III)	Fe <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub>	1.29·10 <sup>-22</sup>	1.14·10 <sup>-11</sup>	0.63·10 <sup>-9</sup>	0.1·10 <sup>-3</sup>

From data in Table 1, one can see that phosphates have low solubility product values, under 10<sup>-8</sup>, and ion concentration in the saturated solution is significantly lower than the maximum permitted, so the sparingly soluble compounds are lithosphere friendly. In addition, a comparison of graphs 5 and 6 shows that, in the first approximation, the phosphate method of geocological protection can be used for impurities up to 100 (for Ni (II), Zn (II),

Cd (II) and up to 106 (for Fe (III) ) of MPC numbers (the multiplicity of the value).

Table 2 shows bonding properties of phosphate octahydrates and sparingly soluble hydrated phosphates that indicate the possibility of forming a good construction product through detoxification. Tables 3 and 4 show thermodynamic properties of clay systems that indicate the release of silica gel, which has absorption properties.

**Table 2.** Bonding Properties of Some Phosphate-Containing Cement Systems with Metal Oxide Additives

Powdered Metal Oxide	Compression Strength, MPa*	Bonding Agent
FeO	40-50	Fe <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> ·8H <sub>2</sub> O
ZnO	20-30	Zn <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> ·4H <sub>2</sub> O
CuO	50-60	Cu <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> ·3H <sub>2</sub> O
CdO	5-9	Cd <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> ·2CdHPO <sub>4</sub> ·8H <sub>2</sub> O
NiO	20-25	Ni <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> ·nH <sub>2</sub> O

Note: "\*" – value depending on the concentration of added H<sub>3</sub>PO<sub>4</sub> and on the water-to-powder ratio.

**Table 3.** Energy Reserves of Raw Materials, ΔH<sup>0</sup><sub>298</sub>, kJ/mol

Raw Material	-ΔH <sup>0</sup> <sub>298</sub> , kJ/mol
1. Cambrian Clay, Al <sub>2</sub> O <sub>3</sub> ·4SiO <sub>2</sub> ·4H <sub>2</sub> O	-5 764.6
2. Kaolin, Al <sub>2</sub> O <sub>3</sub> ·2SiO <sub>2</sub> ·2H <sub>2</sub> O	-4116.0
3. Coal, FeO	-272
4. H <sub>3</sub> PO <sub>4</sub>	-289.26
5. Sand, SiO <sub>2</sub>	-901.99
6. Fuel Oil Residual, C <sub>20</sub> H <sub>42</sub> ; C <sub>20</sub> H <sub>40</sub>	-456.4 -330.8

**Table 4.** Protective Reactions with Phosphoric Acid

Reactions of Kaolin and Montmorillonite Clay in the Phosphate System	Changes in Energy Content, kJ/mol, -ΔH <sup>0</sup> <sub>298</sub>
1. AlO <sub>3</sub> ·2SiO <sub>2</sub> ·2H <sub>2</sub> O+6H <sub>3</sub> PO <sub>4</sub> → 2(SiO <sub>2</sub> ·H <sub>2</sub> O)+2Al(H <sub>2</sub> PO <sub>4</sub> ) <sub>3</sub> +3H <sub>2</sub> O	-282.77
2. Al <sub>2</sub> O <sub>3</sub> ·2SiO <sub>2</sub> ·2H <sub>2</sub> O+2H <sub>3</sub> PO <sub>4</sub> +H <sub>2</sub> O→ 2(SiO <sub>2</sub> ·H <sub>2</sub> O)+2[Al(OH) <sub>2</sub> H <sub>2</sub> PO <sub>4</sub> ]	-292.49
3. Al <sub>2</sub> O <sub>3</sub> ·4SiO <sub>2</sub> ·2H <sub>2</sub> O+6H <sub>3</sub> PO <sub>4</sub> → 4(SiO <sub>2</sub> ·H <sub>2</sub> O)+2Al(H <sub>2</sub> PO <sub>4</sub> ) <sub>3</sub> +H <sub>2</sub> O	-315.82
4. AlO <sub>3</sub> ·4SiO <sub>2</sub> ·2H <sub>2</sub> O+2H <sub>3</sub> PO <sub>4</sub> +3H <sub>2</sub> O→ 4(SiO <sub>2</sub> ·H <sub>2</sub> O)+2[Al(OH) <sub>2</sub> H <sub>2</sub> PO <sub>4</sub> ]	-325.5

Research purpose is to use man-made raw materials, including those that were contaminated with petroleum products and heavy metal ions, to produce fake rocks in which these contaminants will be blocked from the interaction with the lithosphere, and thus, to protect it (geocoprotective function), as well as to evaluate the longevity of those rocks. Longevity factors the possibility of using geocoprotective systems (fake rocks) for construction purposes. Geocoprotection here is defined as a transformation of contaminated materials into environmentally friendly ones, and as elimination of the need for utilizing them by making a long-lasting construction product. The energy for detoxification and hardening of a system with good properties is taken from the system's energy reserve. Tables 3 and 4 show the energy reserves of some substances and how they reactively change. Reactions release energy, and if the amount of mineral powder dominates over the liquid (H<sub>3</sub>PO<sub>4</sub>), then construction products can be made with good properties, due to a reduction in the system's energy reserve. Following the research goal, properties that materials have, made from man-made contaminated hardened solutions of soils and phosphoric acid, have been investigated for more than 10 years in order to make a decision on whether they can be used, considering their behavior over time.

## 2. Methods

Longevity was investigated using standard physical/mechanical research methods for studying construction materials, and physical/chemical research methods for studying heavy metal ions. The ion-selective electrodes were used to study water extracts from the resulted geocoprotective construction products. Geocoprotective phosphate-containing cement systems were made from mixture paste consisting of some soil with specific salinity level and phosphoric acid. Pastes were hardened and investigated at different times. Water extracts from these hardened systems were investigated using the ion-selective electrodes.

## 3. Results and Discussion

Tables 5, 6 and 7 provide research results.

**Table 5.** Frost-Resisting Capacity of Protective Phosphate-Containing Cement System from Sandy Clay, FeO-H<sub>3</sub>PO<sub>4</sub> System

FeO in Sandy Clay, %	H <sub>3</sub> PO <sub>4</sub> to Powder Ratio	After 50 Cycles of Freezing and Thawing, MPa, timing	
		1 Year	10 Years
10	0.21	7.5	8.3
15	0.22	15.5	16.7
10+mineral additive (6.5 %)	0.23	8.5	9.5

**Table 6.** Compression Strength of Protective Phosphate-Containing Cement System from Soil Contaminated with Petroleum Products

Composition, weight, %				Density of H <sub>3</sub> PO <sub>4</sub> , g/cm <sup>3</sup>	Compression Strength, MPa, years	
Contaminated Soil	Clay Material, Al <sub>2</sub> O <sub>3</sub> ·4SiO <sub>2</sub> ·2H <sub>2</sub> O	Sand	FeO		5	10
58.0	14.0	-	7.0	1.42 (solids 21 %)	4.5	5.0
12.0	12.0	48.0	7.0	1.42	2.5	3.0
64.0	8.0	-	7.0	1.42	3.5	4.0
53.0	13.0	-	14.0	1.37	7.5	8.0

**Table 7.** Protective Phosphate-Containing Cement System from Clay Al<sub>2</sub>O<sub>3</sub>·4SiO<sub>2</sub>·2H<sub>2</sub>O and Sand Contaminated with Petroleum Products

Contaminated Sand + Al <sub>2</sub> O <sub>3</sub> ·4SiO <sub>2</sub> ·4H <sub>2</sub> O + FeO, %				Compression Strength, MPa, timing	
Sand	Clay	FeO	Petroleum Product	1 Year	10 Years
51.5	13.3	16.6	1.5	4.0	5.0
50.4	13.3	16.6	2.7	3.5	4.5
49.4	13.3	16.6	3.7	3.0	4.2

Tables show natural clay or artificial material in combination with other contaminant, like iron (II) oxide, enable the production of long-lasting construction system with geocoprotective properties, manifested when the mixture is hardening. These properties are the process of decontamination, which idea is to form sparingly soluble substances, and the process of blocking decontamination in case when petroleum contaminants are in the mixture. Water extracts from the system that were made a different times (1 month through to more than 10 years) revealed the removal of heavy metal ions.

The overall physico-chemical processes in such systems are spontaneous, and triggered by a decrease in Gibbs free energy, evident from the change in the system's energy reserve (Table 8) [7-12]. Based on data in Table 8, we can state that system's properties occur spontaneously, and are linked to a negative change in the Gibbs free energy. Thus, geocoprotective phosphate-containing cement systems run on their own energy reserves. Longevity and stability are also linked to changes in the system's energy reserves: the system is more stable when its energy reserve reduces. The mechanisms of chemical reactions

observed for different solids correspond to those described in [13-18].

**Table 8.** Heat of Hydration in Protective Phosphate-Containing Cement System from Clay

Protective Phosphate-Containing Cement System from Clay	Change in Gibbs Free Energy, ΔG <sup>0</sup> <sub>298</sub> , kJ/mol
ZnO+8/3H <sub>3</sub> PO <sub>4</sub> +AlO <sub>3</sub> ·2SiO <sub>2</sub> ·2H <sub>2</sub> O→ 1/3Zn <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> ·4H <sub>2</sub> O+2SiO <sub>2</sub> ·H <sub>2</sub> O	Negative
ZnO+8/3H <sub>3</sub> PO <sub>4</sub> +AlO <sub>3</sub> ·2SiO <sub>2</sub> ·2H <sub>2</sub> O→ 1/3Zn <sub>3</sub> (PO <sub>4</sub> ) <sub>2</sub> ·4H <sub>2</sub> O+4(SiO <sub>2</sub> ·H <sub>2</sub> O)+2AlPO <sub>4</sub> +8/3H <sub>2</sub> O	Negative

Our research shows that sandy clay reacts with phosphoric acid (density: 1370 kg/m<sup>3</sup>) releasing significant heat (70 kJ/kg after 24 hours). Iron oxide and copper oxide added to the mixture increase the amount of heat released by 20-70 kJ/kg. The greatest heat release was observed when mixing a composition containing 20% of iron (II) oxide (II), and it amounted to 141.85 kJ/kg in 24 hours. For copper (II) oxide, heat release was 125.15 kJ/kg, for copper (I) oxide – 93.22 kJ/kg. These additives apparently trigger the charge (electron or proton) transfer. With them added into the system, phase formation changes, resulting in more sparingly soluble phosphates. The mechanisms of their formation are described in [19-22].

Our research allowed making water- and frost-resistant pyrite cinder, containing iron (II, III) oxides, copper (I, II) oxides and phosphoric acid, which can be used as building bricks or tiles. The composition (sandy clay – 80%, pyrite cinder – 10%, copper (I/II) oxide – 10%) was mixed with a 5% solution of zinc oxide and phosphoric acid with a relative density of 1.25 at 0.3 w/c. Table 9 shows how strong and frost resistant the obtained materials were.

**Table 9.** Physical and Mechanical Properties of Phosphate-Containing Cement System from Clay

Composition, weight, %				Compression Strength, MPa		
Sandy Clay	Pyrite Cinder	Copper (II) Oxide	Copper (I) Oxide	After Saturation	After 30 Cycles of Freezing	After 10 Years of Storage
80	10	10	-	8.22	9.70	11.2
80	10	-	10	9.06	10.52	12.3

Our geocoprotective phosphate-containing cement systems of optimum composition have the following properties that silicate cement does not have: rapid-hardening, significant heat release, low temperature tolerance (hardening process lasts 1-3 hours), any clay and sandy clay additives can be used, protection against petroleum pollution, blocked detoxification of heavy metal ions into naturally soluble substances.

## 4. Conclusions

Based on research results, we can conclude the following:

1. Lithosphere protection method is introduced, which idea is to detoxify contaminated soil while hardening the system until a long-lasting construction product with good properties.
  2. Phosphate-containing cements systems are geocoprotective, and can be used in building and construction as cement articles, formations and buildings.
  3. Some physical-mechanical (strength, frost resistance) and geocoprotective (contaminant removal) properties of phosphate-containing cement systems were identified after they have been stored for more than 10 years.
- Thus, our method can significantly reduce the level of lithosphere contamination.

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