Lithosynthesis in transport construction for geosphere protection

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Abstract. Geoecological problems of lithosphere protection in transport construction are of great importance nowadays. The article deals with the research of the new lithosynthesis technology which can be used for geosphere protection in transport construction. Lithosynthesis is a new method of geosphere purification by means of engineering structures and building materials. The main aim of the paper is to show a possibility to restore lithosphere using mineral artificial stones of different nature, e.g. foam concrete or phosphates. Both theoretical, namely thermodynamic, method and experimental one are used in the study. Two techniques of lithosynthesis are demonstrated in the paper. The first technique is to apply so-called the “stone sponge” to absorb pollutions and detoxicate them. The second technique is formation of the artificial phosphate stone to detoxicate pollutions during hardening. The level of concentration of heavy metal ions to be detoxicated by means of foam concrete lithoreactions is determined. The study suggests creation of a new lock surface, e.g. through sols’ use, after detoxication of pollutions.

1 Introduction

Lithosynthesis is a new method of geosphere purification and geoecological quality improvement of transport construction systems. There are a few problems of geosphere protection during transport construction and one of them is connected with strengthening and detoxication of soils. The main geoecoproblem of soils in construction is inorganic and organic pollutions. The most dangerous substances are heavy metal ions and oil products. One of the best solutions of their detoxication can be pollution absorption from a soil by means of a mineral building material such as an artificial stone. The “stone sponge”, e.g. foam concrete, has these properties and reactions in it are named lithosynthesis [1-6]. There are 4 lithosynthesis requirements to be met: an artificial stone of mineral nature; over 5% porosity of the stone; capillary absorption of the solution; the reaction between heavy metal ions and the stone substances with formation of new substances having a very low solubility product (SP). But it is possible to detoxicate pollutions without reactions, e.g. oil products, and then to block them in an artificial stone. After absorption of geosphere

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pollution and its detoxication inside the “stone sponge” an artificial stone is the same one as it was before absorption but there is the detoxicated pollution in it. This means that geosphere will not have wastes to be disposed. So, in view of the above an artificial stone which works like the “stone sponge” absorbs a solution with pollutions, detoxicates them and blocks them in the solid phase of the stone. Besides pollutions can be detoxicated or blocked during hardening of the mix and it is the second technique of lithothyntesis application. To make sure of pollution binding after absorption an artificial stone can be treated by a sol of different nature, e.g. alumina or silica sol. An artificial stone with sol surface is like a safe that is why this technology has been named safe one. If reactions between minerals of an artificial stone occur in pores and capillaries substances having a very low solubility product (SP), less than 10^{-8} , are formed and provided negative value of Gibbs energy, $\Delta G^{\circ}_{298} < 0$. Therefore, detoxication of pollutions will takes place. The Table 1 shows the examples of reactions in pores and capillaries. These reactions have been named lithoreactions. It should be noticed that apart from hydroxides hydrates of heavy metal silicates having a lower solubility than that of hydroxides can be formed in an artificial stone. Therefore it is possible for hydrates of silicates to be generated.

Table 1. Reactions in pores and capillaries inside the “stone sponge”.

Nature of an artificial stone (base phases)	Examples of ions in the absorbed solutions	ΔG°_{298} of detoxication reaction	Examples of the new phases lythosynthesis
$x\text{CaO} \cdot y\text{SiO}_2 \cdot n\text{H}_2\text{O}$; $\text{Ca}(\text{OH})_2$, $\text{pH} > 7$ (silicate cements)	solutions with ions of Cu(II) and other heavy metals	$\Delta G^{\circ}_{298} < 0$	$\text{Cu}(\text{OH})_2$ and hydrates of silicates
$\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$, $\text{pH} \approx 7$ (sulphate cement)	solutions with ions of Pb(II); Ba(II)	$\Delta G^{\circ}_{298} < 0$	PbSO_4 ; BaSO_4
$x\text{CaO} \cdot y\text{Al}_2\text{O}_3 \cdot n\text{H}_2\text{O}$, $\text{pH} > 7$ (aluminate cement)	heavy metal ions	$\Delta G^{\circ}_{298} < 0$	$\text{Pb}(\text{OH})_2$ $\text{Cd}(\text{OH})_2$
$x\text{Na}_2\text{O} \cdot m\text{Al}_2\text{O}_3 \cdot y\text{SiO}_2 \cdot n\text{H}_2\text{O}$, $\text{pH} > 7$ (geopolimers, alkali cement)	solutions with ions of Ni(II); Fe(III); Co(II) and others	$\Delta G^{\circ}_{298} < 0$	$\text{Ni}(\text{OH})_2$ $\text{Fe}(\text{OH})_3$ $\text{Co}(\text{OH})_2$

The Table 1 introduces the first technique of lithothyntesis for pollution detoxication during binding system hardening. The fact of the matter is that apart from silicates and hydroxides phosphates are suitable for heavy metal ion pollution detoxication. In the Table 2 parameters of heavy metal ion phosphates are presented.

Table 2. Parameters of phosphates.

Ion	Phosphate	Solubility product (SP)	Concentrations of ions		Tolerable concentration (TC) of ions in the soil, g/kg
			mol/l	g/l	
Pb(II)	$\text{Pb}_3(\text{PO}_4)_2$	$7,49 \cdot 10^{-43}$	$1,49 \cdot 10^{-10}$	$3,08 \cdot 10^{-8}$	$20(30) \cdot 10^{-3}$
Cu(II)	$\text{Cu}_3(\text{PO}_4)_2$	$1,26 \cdot 10^{-37}$	$1,63 \cdot 10^{-8}$	$1,03 \cdot 10^{-6}$	$3 \cdot 10^{-3}$
Ni(III)	$\text{Ni}_3(\text{PO}_4)_2$	$5,01 \cdot 10^{-31}$	$3,41 \cdot 10^{-7}$	$2,01 \cdot 10^{-5}$	$4 \cdot 10^{-3}$
Zn(II)	$\text{Zn}_3(\text{PO}_4)_2$	$9,12 \cdot 10^{-7}$	$1,53 \cdot 10^{-7}$	$\approx 1 \cdot 10^{-5}$	$23 \cdot 10^{-3}$
Cd(II)	$\text{Cd}_3(\text{PO}_4)_2$	$2,51 \cdot 10^{-33}$	$1,18 \cdot 10^{-7}$	$1,32 \cdot 10^{-5}$	$0,5 \cdot 10^{-3}$
Fe(III)	$\text{Fe}_3(\text{PO}_4)_2$	$1,29 \cdot 10^{-22}$	$1,14 \cdot 10^{-11}$	$0,63 \cdot 10^{-9}$	$0,1 \cdot 10^{-3}$

According to the Table 2 phosphates have a very low solubility product which results in binding of free heavy metal ions and formation of the phosphate artificial stone under the certain conditions. The phosphate stone has a constructional function because detoxicated pollutions are inside the artificial “stone sponge”. So, an artificial stone is useful in construction as during absorption and hardening it detoxicates both heavy metal ions and oil products. Different clays such as $\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O}$ or $\text{Al}_2\text{O}_3 \cdot 4\text{SiO}_2 \cdot 2\text{H}_2\text{O}$ were chosen as phosphates of construction systems with heavy metal ions and oil products’ pollutions as the solid component in the reaction with H_3PO_4 . The Table 3 demonstrates that all the reactions have negative change of ΔH°_{298} which is important at negative temperatures.

Table 3. Reactions of clay with H_3PO_4 .

Reactions	ΔH°_{298} , kJ/mol
$\text{AlO}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O} + 6\text{H}_3\text{PO}_4 = 2(\text{SiO}_2 \cdot \text{H}_2\text{O}) + 2\text{Al}(\text{H}_2\text{PO}_4)_3 + 3\text{H}_2\text{O}$	-282,77
$\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 2\text{H}_2\text{O} + 2\text{H}_3\text{PO}_4 + \text{H}_2\text{O} = 2(\text{SiO}_2 \cdot \text{H}_2\text{O}) + 2[\text{Al}(\text{OH})_2\text{H}_2\text{PO}_4]$	-292,49
$\text{Al}_2\text{O}_3 \cdot 4\text{SiO}_2 \cdot 2\text{H}_2\text{O} + 6\text{H}_3\text{PO}_4 = 4(\text{SiO}_2 \cdot \text{H}_2\text{O}) + 2\text{Al}(\text{H}_2\text{PO}_4)_3 + \text{H}_2\text{O}$	-315,82
$\text{AlO}_3 \cdot 4\text{SiO}_2 \cdot 2\text{H}_2\text{O} + 2\text{H}_3\text{PO}_4 + 3\text{H}_2\text{O} = 4(\text{SiO}_2 \cdot \text{H}_2\text{O}) + 2[\text{Al}(\text{OH})_2\text{H}_2\text{PO}_4]$	-325,5

The main aim of the study is to get thermodynamic calculations and practical estimation of detoxication reactions in pores and capillaries (lithoreactions) of building materials and structures. Moreover, taking into account an artificial stone’s ability to absorb heavy metal ions, it is possible to estimate the concentration level of lithosynthesis detoxication in order to create a lock surface which prevents any extraction from the stone. Besides, one can suggest examples of lithosynthesis application. As preliminary information about a possibility to use a mineral stone (as the “stone sponge”) for lithosynthesis it is necessary to know an artificial mineral stone’s physical properties, given in the Table 4. Based on the physical properties mentioned below there were experiments with foam concrete articles’ examples.

Table 4. Physical properties of some mineral building materials.

Mineral material articles	Density, kg/m ³	Porosity, %	λ , wt/m·K (average value)
silicate concrete	2600-2700	8-12	≈ 2
construction gypsum	1250	≈ 75	$\approx 0,35$
foam concrete	400-800	≈ 80	0,1-0,3
granite	2700-2800	0,5-1	$\approx 2,4$

2 Methods

The thermodynamic and experimental methods were applied. The samples of foam concrete articles were saturated with water solution contained heavy metal ions up to 10-200 TC (tolerable concentration) for 1 day. Cu(II), Pb(II), Cd(II), Ni(II) were used as heavy metal ions. Later, in 28 days, the foam concrete samples with absorbed heavy metal ions were put into the water for ion extraction and then, in 1 week, the water was examined using ion selective electrodes.

To create a lock surface of foam concrete and to block phosphate materials’ illuviation after 28 days of hardening the samples were put into 3% silica sol solution or sprinkled with sol solution.

Phosphates and calcium silicates were tested for heavy metal ions and oil products’ binding as well as durability of the material.

In the Table 5 the base for thermodynamic calculations is elaborated.

Table 5. Thermodynamic base for calculations, where s - solid substance, l – liquid substance.

Substance	Condition	ΔG°_{298} , kJ/mol
H ₂ O	l	-237,4
Ca(II)	l	-552,5
Ca(OH) ₂	s	-895,9
(OH) ⁻	l	-157,1
5CaO·6SiO ₂ ·10,5H ₂ O	s	-11065,7
Cd(II)	l	-77,5
Cd(OH) ₂	s	-473,1
Pb(II)	l	-24,3
Pb(OH) ₂	s	-420,5
Ba(II)	l	-560,12
5CaO·6SiO ₂ ·5,5H ₂ O	s	-9870,86
BaSO ₄	l	-1363,2
CaSO ₄ ·2H ₂ O	s	-1798,65
CaSO ₄ ·0,5H ₂ O	s	-1432,82
PbSO ₄	s	-814,38
CaO·AlO ₃ ·10H ₂ O	s	-4581,2
Al ₂ O ₃ ·3H ₂ O	s	-2301,5
Al ₂ O ₃ ·2SiO ₂	s	-3097,8
(NaAlSi ₂ O ₆ ·H ₂ O)	s	-3074,79
CaO·2Al ₂ O ₃	s	-3779,97
Al(OH) ₃	s	-1136,54
NaOH	l	-418,77
Na(I)	l	-261,6

3 Results and discussion

The Table 6 illustrates thermodynamic calculations performed according to the classical method.

Table 6. Thermodynamic calculations of the reactions in an artificial stone.

Reactions in pores of an artificial stone of different nature (lithoreactions)	ΔG°_{298} , kJ
$5\text{CaO} \cdot 6\text{SiO}_2 \cdot 5,5\text{H}_2\text{O} + \text{Pb}^{2+} + 2\text{OH}^- + 5\text{H}_2\text{O} = \text{Pb(OH)}_2 + 5\text{CaO} \cdot 6\text{SiO}_2 \cdot 10,5\text{H}_2\text{O}$	-89,84
$\text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{Pb}_{\text{aq}}^{2+} = \text{PbSO}_4 \downarrow + 2\text{H}_2\text{O} + \text{Ca}^{2+}$	-18,73
$2(\text{CaSO}_4 \cdot 0,5\text{H}_2\text{O}) + \text{Ba}^{2+} = \text{BaSO}_4 + \text{CaSO}_4 \cdot 2\text{H}_2\text{O} + \text{Ca}^{2+}$	-288,59
$2(\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2) + 4\text{NaOH} + 4\text{H}_2\text{O} + \text{Cd}^{2+} = 2(\text{NaAlSi}_2\text{O}_6 \cdot \text{H}_2\text{O}) + \text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O} + \text{Cd(OH)}_2 + 2\text{Na}^+$	-549,6
$2(\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2) + 4\text{NaOH} + 4\text{H}_2\text{O} + \text{Pb}^{2+} = 2(\text{NaAlSi}_2\text{O}_6 \cdot \text{H}_2\text{O}) + \text{Pb(OH)}_2 + \text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O} + 2\text{Na}^+$	-550,2
$\text{CaO} \cdot 2\text{Al}_2\text{O}_3 + 13\text{H}_2\text{O} + \text{Cd}_{\text{ag}}^{2+} + 2\text{OH}^- = \text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 10\text{H}_2\text{O} + 2\text{Al(OH)}_3 + \text{Cd(OH)}_2$	-69,51

The result of the calculations was the negative value of lithoreactions in every case, e.g. foam concrete as calcium silicate system (the first system in the Table 6) had free Gibbs energy -89,84kJ during lithoreaction of detoxicftion. Therefore, thermodynamics of lithoreactions is sure to be allowed.

Foam concrete articles of different density (400-800 kg/m³) were saturated with the solution having 10-200 TC (tolerable concentration) of heavy metal ions, their holding time

being 1 day. The Table 7 has the data about aqueous extract from the foam concrete samples after storage them in solutions with heavy metal ions both in 1 day and in 1 week.

Table 7. Aqueous extract from the foam concrete samples.

Ions in the water before the foam concrete samples saturation	The water after the foam concrete samples storage in it for 1 day and 1 week
Cu(II)	Cu(II) is not found
Pb(II)	Pb(II) is not found
Cd(II)	Cd(II) is not found
Ni(II)	Ni(II) is not found

Needless to say that it is necessary for detoxication to determine a minimum concentration of heavy metal ions through the reactions with substances of a stone.

Solid products, e.g. Pb(OH)_2 , Cd(OH)_2 , Cu(OH)_2 and other hydroxides, are generated in a stone's pores and capillaries with $\text{pH} > 7$ (more than 10). The Table 8 contains calculations which are the base for estimation of a minimum concentration of heavy metal ions through lithosynthesis in order to detoxicate them in a stone, solubility product (SP) being taken into consideration.

Table 8. The level of heavy metal ion detoxication through lithoreactions.

Substance	Solubility product (SP)	Concentration of heavy metal ions in the saturated solution, mol/l	Solution with $\text{pH}=10$ mol/l
Cu(OH)_2	$5,6 \cdot 10^{-20}$	$\approx 10^{-7}$	$5,6 \cdot 10^{-12}$
Pb(OH)_2	$0,2 \cdot 10^{-15}$	$\approx 10^{-5}$	$0,2 \cdot 10^{-7}$
Cd(OH)_2	$4 \cdot 10^{-15}$	$\approx 10^{-5}$	$4 \cdot 10^{-7}$
Ni(OH)_2	$6 \cdot 10^{-18}$	$\approx 10^{-6}$	$6 \cdot 10^{-10}$

According to the principles of general chemistry and with reference to the data from the Table 8 even a minimum concentration of heavy metal ions in the saturated solution with $\text{pH}=10$ and $[\text{OH}^-]=10^{-4}$ mol/l results in formation of sediments which bind pollutions.

$$\text{Ksp Cu(OH)}_2 = [\text{Cu}^{2+}][\text{OH}^-]^2; \text{Cu(II)} = 5,6 \cdot 10^{-20} / 10^{-8} = 5,6 \cdot 10^{-12} \text{ mol/l}$$

$$\text{Ksp Pb(OH)}_2 = [\text{Pb}^{2+}][\text{OH}^-]^2; \text{Pd(II)} = 0,2 \cdot 10^{-15} / 10^{-8} = 0,2 \cdot 10^{-7} \text{ mol/l}$$

$$\text{Ksp Cd(OH)}_2 = [\text{Cd}^{2+}][\text{OH}^-]^2; \text{Cd(II)} = 4 \cdot 10^{-15} / 10^{-8} = 4 \cdot 10^{-7} \text{ mol/l}$$

$$\text{Ksp Ni(OH)}_2 = [\text{Ni}^{2+}][\text{OH}^-]^2; \text{Ni(II)} = 6 \cdot 10^{-18} / 10^{-8} = 6 \cdot 10^{-10} \text{ mol/l}$$

So, it is obvious that foam concrete articles can detoxicate systems with heavy metal ion pollutions. It goes without saying that the concentration of heavy metal ions in the saturated solution according to SP (solubility product) is the same or less than TC (tolerable concentration). Solubility product of heavy metal ions' silicates and hydrates of silicates is lower than that of hydroxides. Therefore, reliability of detoxication is higher during lithosynthesis.

Lithosynthesis is a method which is reliable for durability of engineering transport structures and building components. The increase of some technical properties in 1-10 years is the result of the study which examines heavy metal ions. As mentioned above heavy metal ion silicate hydrates have a lower solubility product than that of hydroxides and the results of properties increase can be explained by silicate hydrates' formation inside the cement stone [7-12]. But when combining with silica sol absorption or sol solution

sprinkling, lithosynthesis method is certain to lock substances in an artificial stone. It means that if an article's surface is saturated with a sol, e.g. silica sol, after lithosynthesis of heavy metal ions a lock membrane is generated on this surface. Such an effect, no doubt, is good for environmental protection.

The research proves that hardness of foam concrete increases up to 50% that is why reliability of the "stone sponge" is raised after detoxication. Papers [13-15] investigate pores and capillaries of a surface and consider properties of an inorganic surface as the object of the impact. According to proceedings [16-20] the reactions start in active places of surfaces and then the development of the process takes place in pores and capillaries. The artificial phosphate building stone was obtained when using clay polluted by heavy metal ions and oil products in the reactions with H_3PO_4 . In the Tables 9 and 10 there are results of clay and phosphoric acid composition application [20].

Table 9. Clay phosphate composition with FeO.

FeO, %, with clay, %, to 100% of all mix	Ratio of H_3PO_4 and clay	Compressive strength, MPa	
		1 year	10 years
10	0,21	7,5	8,3
15	0,22	15,5	16,7
17	0,23	8,5	9,5

Table 10. Clay phosphate composition with oil pollutions.

Composition, %, with H_3PO_4 (p-1,42 g/cm ³), %, to 100% of all mix				Durability according to compressive strength, MPa	
soil with oil pollutions to 10%	Clay, $Al_2O_3 \cdot 4SiO_2 \cdot 2H_2O$	Sand	FeO	5 years	10 years
58,0	14,0	-	7,0	4,5	5,0
12,0	12,0	48,0	7,0	2,5	3,0
64,0	8,0	-	7,0	3,5	4,0
53,0	13,0	-	14,0	7,5	8,0

One can see that compressive strength does not lower over time. If phosphate composition with heavy metal ions is used strength will be a little more over time. Solubility product (SP) of phosphates with heavy metal ions having a very low value (see the Table 2), aqueous extract of phosphates has no free ions. So, foam concrete articles and structures can absorb inorganic pollutions and detoxicate them. Phosphates material can detoxicate an inorganic pollution or block oil pollution during hardening. That is why the properties of lithosphere are preserved during transport construction. At the same time geocoprotection takes place.

Apart from free heavy metal ions oil pollutions can be detoxicated by phosphates because of oxides binding (see the Table 11).

Table 11. Binding properties of phosphates.

Formula	Compressive strength, MPa	The main product of hardening
FeO	40-50	$Fe_3(PO_4)_2 \cdot 8H_2O$
ZnO	20-30	$Zn_3(PO_4)_2 \cdot 4H_2O$
CuO	50-60	$Cu_3(PO_4)_2 \cdot 3H_2O$
CdO	5-9	$Cd_3(PO_4)_2 \cdot 2CdHPO_4 \cdot 8H_2O$
NiO	20-25	$Ni_3(PO_4)_2 \cdot nH_2O$

There are two aspects of phosphates system use. The first one is silica sol obtaining (see the Table 3) and these substances are of great importance for geosystem preservation. The second aspect refers to clay's high standard values of ΔH°_{298} , kJ/mol, listed in the Table

12. Comparing energies one can make a conclusion that clay and H₃PO₄ system is very useful for purification due to construction systems' inner energy use.

Table 12. Standard values of ΔH°_{298} .

Clay	ΔH°_{298} , kJ/mol
Al ₂ O ₃ ·4SO ₂ ·2H ₂ O	-5764,6
Al ₂ O ₃ ·2SO ₂ ·2H ₂ O	-4116,0
H ₃ PO ₄	-1289,26

4 Conclusions

- Theoretical calculations and practical estimation have shown that foam concrete can absorb and detoxicate heavy metal ions. Some lithoreactions in foam concrete was calculated in terms of thermodynamics for heavy metal ion detoxication. It was established that negative change of Gibbs energy allows the processes.
- A minimum concentration of some heavy metal ions to be precipitated inside pores and capillaries of a stone, particularly foam concrete, with pH=10 was determined.
- Durability of phosphate building materials contained polluted clay was proved because oil products and other pollutions are locked in an artificial stone.

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