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Bio resistant Building Composites Based on Waste Glass

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Abstract: *In this paper investigation results of properties of autoclave-free building materials based on waste glass are given. Quantitative ratios of change of strength and density are obtained. It has been theoretically and experimentally proved, that composites based on binding agent from broken glass possess the raised stability in biologically excited environments. Potential biodestructors able to stocking on the surface of considered materials at their operation in air-dry conditions are established.*
Key words: *broken glass, strength, cement, epoxy resin, liquid glass, microorganisms, biological resistance.*

I. Introduction:

Progressive industrial development leads to such changes in biosphere, which are mainly contraindictive to all living beings. Therefore the problem of decreasing of human adverse effect on his natural habitat is obtaining a considerable actuality.

At the present day it is not given enough attention to the problem of utilization of production wastes in our country. Annually industrial enterprises dump hundreds of waste tonnes, polluting the environment and negatively influencing the environmental situation in general. Taking into account the fact that the attitude to the process for their utilization has not a tendency to change for better, it is possible that in the course of time a great actuality will gain this problem. That is why it is already necessary to pay a careful attention to it and try to draw a maximum number of economy branches to the solving of such important task.

One of the main obstacles to the solving of the previously mentioned problem is the absence of sufficient number of actual projects, consisting in the development of process solutions, allowing providing the repeated usage of industrial wastes at obtaining of products of different purpose.

Investigations with the purpose of the development of utilization technology of broken glass by means of industry of building materials have been carried out by the specialists of Mordovian State University over a period of more than 15 years [3, 7, 15, 17].

Works of Y.P. Gorlov, A.P. Merkin and their followers have been used as a theoretical background for the preparation of building materials based on industrial broken glass [1, 4]. It has been established by them that curing of systems, consisting of natural or imitate glasses is based on the interaction reaction between silica and aqueous solutions of alkali as a result of which compounds are synthesized close to their chemical composition to sedimentary and metamorphic rocks of natrolite, mordenite type, etc. However this process is carried out at increased temperatures and pressures. It has been found by us that the formation of the above mentioned compounds can be performed without use of autoclave treatment. This can be achieved if the corrective ingredients are additionally introduced to the system. It has been found out that native clays and also production wastes of plants of building industry, specialized in the output of ceramic materials and articles can be used as such ingredients.

Experimental Procedure

The advantage of the technology for the preparation of building composites based on autoclave-free binding agent from broken glass consists in that at preparation of raw materials, used as components of binding agent and mainly being production wastes, there is no a necessity to carry out energy-consuming operations except for the separation of metallic inclusions and mixed grinding of broken glass and active mineral admixture to the specific surface area of 3000-3500 cm^2/g. The process for the preparation of solutions and concretes based on binding agent from broken glass can be realized on technological lines of enterprises of building materials industry, specialized in the output of portland cement articles.

The analysis of characteristic features of hydration and structure formation of alkali glass systems, investigated with the help of modern physical methods, shows a considerable intensification of physico-chemical curing processes at thermally wet treatment in comparison with curing in standard conditions.

The formation process of cementing agents of double-water silicate glasses differs fundamentally from curing of ground quartz with liquid glass at production of acid-resistant binders and concretes, from curing of alkali-slag binders on liquid glass, alkali and soda.

Firstly, in contrast to quartz sand alkali glasses are subjected to desalination with water of silicate alkali metals. Silicates of heavy metals even interact with water (Pb, Cd, Zn). They are also hydrolized, but nonsoluble hydrolysis products are deposited in places of their formation. At temperatures higher than 60 ºC the water begins to transform silica into solution. At limited water volume interaction products remain in places of their formation, their concentration grows with the passage of time, and this causes secondary reactions between components of glass and products of its disintegration. By their mechanism and results the reactions taken place come close to the actions of alkali to glass.

Glass fracture by solutions of alkali is the result of change of glass silica at their interaction to stable anions $Si_2O_5^{2-}$, SiO_4^{4-} and SiO_3^{2-}. Placed in glass cations Na^+, K^+, Ca^{2+}, Ba^{2+} and others form hydroxides or compounds of aluminate type in the presence of AlO_3^{3-} or $Al(OH)_2^+$ anion, zincates with ZnO^{2+}. In addition to the mentioned processes in solutions of hydroxides of alkali-earth metals of Ca, Mg, Ba stable in alkali hydrosilicates are formed. The formation of the last is peculiar to the glasses of incandescent lamp with increased content of CaO (5–6 %), MgO (3.2–3.8 %) and BaO (2.2–5.5). Hydrosilicates are also formed at the influence of alkali to the ground container glass, in which CaO – 5.2 %, MgO –3.2 %. Calcium hydrosilicates are practically absent in hydrolyzed crystal glass for the lack of MgO, BaO and a very small content of CaO (1 %).

It is important that by the level of influence different hydroxides at equal normality of a solution are placed in the following order NaOH > KOH > LiOH, NH4OH. The increasing of alkali concentration from 0,5 N to 10 N (20–400 g/l) affects the damage of the surface of glass as follows:

– the thickness of etched layer in glasses, rich in silica (container, lamp) is grown proportionally to concentration logarithm;

– the damage of glasses, containing Ca, Ba, Cr is not depended on concentration;

– the damage of glasses, containing Pb, Mg at first is increased, passed through maximum at concentration of 6–7 N, and then is decreased.

The concentration of 6–7 N corresponds to 160–200 g of NaOH in 1 l of gauged water.

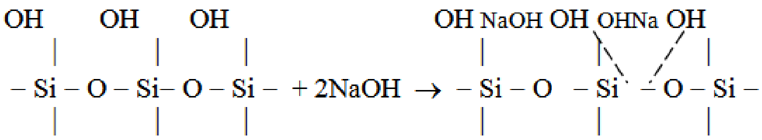
Therefore the most optimal concentration of NaOH will be its quantity of 6 % from weight of ground glass at W/S ratio equal to 0.4.

The considered influence of NaOH relates to the damage of glasses from the surface. Possibly, the influence rate of NaOH to ultimate particles of the ground glass will be incomparably high.

The change of finely dispersed powder of binding agent based on broken glass to stony body is taken place because of interaction of components, consisting of it, with aqueous solutions of alkali. For the clarification of curing mechanism of such binding agent the interaction process of aqueous solutions, having the increased level of hydrogen index with silica (the main component of binder) will be considered.

This process represents heterogenous reaction and consists of series of stages, the main of which are diffusion of components of liquid medium to the surface of phase separation, process for the adsorption of hydrated cations of alkali metals on active sites of silica surface (the sorption in general is followed by the change of the state of silicon oxygen tetrahedrons, which results in depolymerization silicic acid by means of breaking of silica acid bonds), further act of chemical interaction and, finally, the outlet of reaction products to intergranular volume.

The process for depolymerization of silica is connected with adsorption phenomenon and is explained by the idea about coordination nonsaturation of atoms of silica. Hydroxyl ion OH hydroxylates the surface of silica in such a way that OH groups form a coordination bond with atom of silica. The mechanism according to which the interaction of NaOH with SiO_2 is taken place at the stage of depolymerization of silica, may be represented as follows:



The increasing of coordination number of silicon to six corresponds to the adsorption of both hydroxide ions on silane site of the surface of silica atom, which is in tetrahedral arrangement. With the achievement of it the redistribution of energy bonds is taken place. The formation of such transition complex leads to the weakening of≡ Si − O − Si ≡ bonds. The particle, corresponding to monomer in such transition state is retained firmly by its neighbors, and in such a way conditions are formed for its transition to the solution. With the increasing of temperature a mobility of elements of silicon oxygen skeleton increases, С увеличением температуры возрастает подвижность элементов кремнекислородного скелета, rigidity of bonds decreases and therefore less energy for the removal of the tetrahedron is required.

Taking into account the above mentioned processes, carried out at interaction of aqueous solutions of alkali with silicon, the mechanism for curing the binding agent based on broken glass is represented in the following manner. At first under the influence of alkali and increased temperature a finely dispersed amorphous silica is solved from the surface of particles of glass, as a result of which increases its concentration in the solution, the vapor is condensed, leading to the decreasing of pH medium and causing a reaction of polycondensation with formation of gel of polysilicic acid, bonding incompletely solved particles of glass and grains of filling material. The further influence of temperature in the course of thermally wet treatment leads to the crystallization of acid gel with formation of hardly soluble hydroaluminosilicate compounds.

The X-ray structure method has been used for the revelation of structural changes, taken place in binding agent in the course of hydration and curing processes. The research consisted in comparison of phase content of separate components of binding agent with phase content of the composition, obtained by their tempering with alkali solution and curing. The purpose of the experiment was the revelation of the developed new formations. The results of the X-ray structure analysis of raw materials has shown that except glass phase there is little quantity of crystalline phases in the sample, evidenced by incomplete set of diffraction reflections of weak intensity (d = 0.424; 0.334; 0.228 Nm), corresponding to crystalline phase of SiO_2 in the form of quartz and feldspar (d = 0.322 Nm).

On the X-ray diagram, reflecting phase composition of mineral additive, consisting of binding agent, a range of diffraction reflections is be observed, relating to crystalline phases of quartz (d = 0.424; 0.228 Nm), feldspar (d = 0.652; 0.424; 0.356; 0.346; 0.326; 0.322; 0.299 Nm) and montmorillonite (d = 0.242; 0.168; 0.150 Nm).

On diffractograms of cured samples of binding agent lines of crystal new formations are fixed with d = 0.707; 0.404; 0.318; 0.268 Nm, corresponding to zeolite ($Na_2Ca_2Al_6Si_9O_{30}\times9H_2O$) and alkali aluminosilicates of albite type – $NaAlSi_3O_8$ (d = 0.374;0.321; 0.292 Nm), and also a range of diffraction reflections relating to crystalline phases of SiO_2, in the form of low temperature quartz modifications (d = 0.424; 0.334; 0.228 Nm).

The research of dependence of strength from quantitative content of aqueous solution of sodium hydrate and mineral additive and also the type of the last in composition has been carried out. It has been determined that the better properties have such compositions, in which as mineral component a finely dispersed powder of expanded clay is used.

On the basis of the developed binding agents the compositions of solutions and concretes are obtained and their physicotechnical properties are investigated. The main characteristics of the materials are given in table 1.

Table 1 – Physicotechnical parameters of building materials based on alkali glass binding agent

Parameter	Mortar	Heavy concrete	Light concrete	Foamed concrete	Concrete with aggregates from microspheres
Compressive strength, MPa	18	25	16	0.5-0.9	20
Mean density, kg/m^3	2000	2400	1400	500	650
Heat conduction coefficient, W/m °C	–	–	0.43	0.13	0.19
Elasticity modulus, MPa	6000	9750	4600	400	6500
Thermal-expansion coefficient	0.897×10^{-5}	1.558×10^{-5}	0.427×10^{-5}	–	–
Linear shrinkage, %	0.13	0.12	0.24	–	–
Water absorption for 24 h, % by weight	0.3-0.6	0.2-0.3	1.5-4.5	30-50	0.2

One of the distinctive features of new materials is that sands with argillaceous admixtures can be used for their filling. The hydration of clay minerals is carried out at curing of solutions and concretes on such sands in conditions of superalkalinity of liquid phase, as a result of which alkali hydroaluminosilicates are formed, contributing to compaction of their structure. Thus it has been found out on the basis of experimental analysis that at the use of sand with the content of clay of 7 % for the preparation of mortar its strength after thermally wet treatment was on 18 % higher than in analogous content, the filling material in which was pure quartz sand.

Thus, the use of binder based on broken glass allows applying of sands with high content of clay impurities for the preparation of concretes, which are not recommended for cement concretes. It is important to note that resources of such sands in Russia are sufficiently large, while in many regions expensive procedures for the enrichment of native sands are carried out.

In recent years the growth of diversity and population of microorganisms is registered, causing biological damage of materials and constructions. The aggressiveness of the known species is grown. The introduction of new materials to the building contributes only at the beginning to the limitation of such failure mode. It has been calculated that the harm caused to the objects in the result of biological damage makes up a number of ten billion dollars [2, 14]. The processes of biodeteriorations are in progress year by year. The sample observation of buildings and constructions in Moscow, Saint Petersburg, Nizhni Novgorod, Vladivostok, Yakutsk, Saransk and in other towns has shown that a great their number has been damaged by microorganisms of many species [2, 14].

Literature data show that more than 40 % of total number of biodeteriorations is connected with microorganism activity – bacteria and fungi. Bacteria generate at the excessive moisture content in materials, for example, at their contact with liquid (cooling towers, tanks, pipelines, collectors, etc.). At the absence of liquid spray medium the development of bacteria is suppressed, and they give their place to fungi, which are also developed at moisture higher than 75 %.The optimum moisture for them is 95–98 %. On the other hand it is known that fungi and spores of lot of bacteria retain their life also in dried condition. Some hundreds of fungi are known, causing damages of different industrial and building materials. In addition the operational experience of materials of constructions, buildings and structures is shown that the greatest damaging effect from microorganisms is caused by filamentous fungi, in comparison with bacteria and actinomycetes. Dominant in the processes of biodeterioration species of filamentous fungi, relating to the class of hyphomycetes are the following: *Aspergillus niger, Aspergillus flavus, Aspergillus terreus, Penicillium cyclopium, Penicillium funiculosum, Penicillium chrysogenum, Paecilomyces varioti, Chaetomium globosum, Trichoderma viride* [1]. In the temperate climate fungi cause damages of industrial materials in the course of their preparation, when the production is interconnected with high temperature and moisture, and also at violation of terms of keeping and exploitation, transportation. In tropical and subtropical climate the growth of fungi is more intensive, and therefore the damage caused by fungi is more significant.

Filamentous fungi damage practically all natural and many synthetic materials, and also steel and reinforced concrete structures.

Growth and live activity of microorganisms are closely connected with the conditions of the environment, in which they live. The external environment can initiate or suppress the growth of biodestructors. Damage of materials by fungi depends on their composition. Primarily materials are damaged, containing nutrients for fungi.
Organic compounds, being the source of nutrition for fungi, are the part of many building and industrial materials, and this is the cause of their stocking by fungi. In addition the source of organic substances can be different kinds of pollutions, getting on materials, which by their chemical composition are not the source of energy and carbon (metals, polymers, etc.). In table 2 are given obtained results, showing comparative data of biological stability of different kinds of binders.

Table 2 – Biostability of binders after maturing

Material	Method 1	Method 3	Result
Alkali glass binder	0	0 (R* = 45 mm)	fungicidal
Portland cement stone	0	3	funginert
Gypsum rock	4	5	not funginert
Cured epoxy resin	2	5	funginert
Mortars based on liquid glass	0	0 (R = 8 mm)	fungicidal

*R – Radius of inhibition zone of fungi growth

The results show that only samples of alkali glass binder and mortar based on liquid glass possess fungicidal properties. High biostability of binder based on broken glass is explained apparently by the increased level of hydrogen value of the system. In composites based on liquid glass the same effect is explained by the fact that as a curing agent sodium silicofluoride is used (20 parts by weight to 100 parts by weight of liquid glass), which has a great fungicidal action. Then biostability of samples of materials, given in table 2 after their dry curing for three or nine months (table 3) has been investigated.

Table 3 – Results of biostability of binders

Composition	Method 1	Method 3	Result
after dry curing for 3 months			
Alkali glass binder	0	0(R=15 mm)	fungicidal
Portland cement stone	0	3	funginert
Gypsum rock	4	5	not funginert
Cured epoxy resin	2	5	funginert
Mortars based on liquid glass	0	0(R=15 mm)	fungicidal
after dry curing for 9 months			
Alkali glass binder	0	0	fungicidal
Portland cement stone	2	4	funginert
Gypsum rock	5	5	not funginert
Cured epoxy resin	4	5	not funginert
Mortars based on liquid glass	0	0	fungicidal
after dry curing for 12 months			
Alkali glass binder	0	2	funginert
Portland cement stone	0	4	funginert
Gypsum rock	4	5	not funginert
Cured epoxy resin	4	5	not funginert
Mortars based on liquid glass	0	0	fungicidal

The obtained in the course of the experiment results show that the decreasing of biological stability of materials is carried out. Samples of alkali glass binder and mortar based on liquid glass retain fungicidity, though in the course of time the radius of inhibition zone of fungi growth is fallen to zero (at tests to fungicidity the area around the samples is slowly overgrown). As for the samples of portland cement stone they remain funginert. Samples of cured epoxy resin after dry curing for 9 months become not funginert.

With the purpose of revelation of potential biodestructors of binder of organic and inorganic origin after dry curing the investigations for the determination of species composition of microorganisms have been carried out. The task of the research was the establishment of the genus of fungi from the present in space air, able to use binding agents as energy source, and also the determination of concrete species – the representatives of the genus.

In the course of performance of the research genus and species of fungi are established, stocking on samples of materials in air-dry conditions for three, nine and twelve months after maturing strength. Species composition of fungi is given in table 4.

After maturing of samples for 3 months 1 genus of fungi has been found on portland cement stone – *Aspergillus ustus*. On samples of gypsum rock – 5 genuses of fungi (*Aspergillus, Penicillium, Mucor, Chaetomium, Verticillium*) able to grow on their surface. On samples of cured epoxy resin 3 genuses of fungi have been found (*Aspergillus*, *Penicillium*, *Mucor*). As for the number of fungi, found on the surfaces of the investigated compositions after dry curing for 9 months, it was in all cases grown.

Colonies of fungi are not observed on alkali glass binder and samples of mortar based on liquid glass at curing in normal conditions for 3 months. After 9 months colonies of fungi even if have been observed, but there are few and develop slowly and are in suppressed state. On compositions fungi grew on stamps from samples and on medium near the samples.

After 12 months of curing total number of species of fungi, found out on the surfaces of the investigated samples has been increased. It should be noted that on the surfaces of all compositions (except samples of mortar based on liquid glass) similar species of fungi are found out: *Aspergillus niger, Aspergillus ustus, Cladosporium elatum*.

Table 4 – Species composition of fungi colonies, stocking binders

Item of material	Species of fungi	Total number of species of fungi / number of genuses
after dry curing for 3 months		
Alkali glass binder	Fungi are not found	0
Portland cement stone	Aspergillus ustus	1/1
Gypsum rock	Aspergillus ustus, Penicillium nigricans, Mucor corticola, Chaetomium globosum, Verticillium nigrescens	5/5
Cured epoxy resin	Aspergillus niger, Aspergillus ustus, Penicillium nigricans, Mucor corticola	4/3
Mortars based on liquid glass	Fungi are not found	0
after dry curing for 9 months		
Alkali glass binder	Aspergillus niger, Penicillium cyclopium, Penicillium claviforme, Penicillium notatum, Penicillium lanoso-griseum, Alternaria dianthi, Chaetomium globosum	7/4
Portland cement stone	Aspergillus niger, Aspergillus oryzae, Alternaria alternata, Alternaria dianthi, Penicillium notatum, Penicillium chrysogenum, Penicillium lanosum, Penicillium urticae, Penicillium puberulum, Gliocladium roseum	10/4
Continuation of the table 4		
1	2	3
Gypsum rock	Aspergillus niger, Aspergillus oryzae, Aspergillus ustus, Aspergillus clavatus, Alternaria brassicae, Alternaria alternata, Cladosporium macrocarpum, Cladosporium elatum, Chaetomium globosum, Chaetomium dolichortrichum, Penicillium lanosum, Penicillium notatum, Penicillium nigricans	13/5
Cured epoxy resin	Aspergillus niger, Aspergillus ustus, Penicillium lanosum, Penicillium notatum, Penicillium palitans, Penicillium urticae, Penicillium puberulum, Cladosporium macrocarpum, Mucor corticola, Mucor circinelloides	10/4
Mortars based on liquid glass	Aspergillus niger, three colonies of bacteria	1/1
after dry curing for 12 months		
Alkali glass binder	Aspergillus niger, Aspergillus ustus, Penicillium notatum, Penicillium claviforme, Penicillium cyclopium, Penicillium ochrochloron, Penicillium nigricans, Fusarium moniliforme, Cladosporium elatum	9/4
Portland cement stone	Aspergillus niger, Aspergillus ustus, Alternaria brassicae, Cladosporium elatum	4/3
Gypsum rock	Aspergillus niger, Aspergillus ustus, Aspergillus fumigatus, Aspergillus clavatus, Aspergillus oryzae, Penicillium notatum, Cladosporium elatum, Fusarium sambucinum, Chaetomium dolichortrichum, Mucor corticola, Mucor circinelloides, Alternaria brassicae	12/7
Cured epoxy resin	Aspergillus niger, Aspergillus ustus, Aspergillus fumigatus, Aspergillus ruber, Penicillium notatum, Alternaria brassicae, Alternaria pluriseptata, Fusarium moniliforme, Cladosporium elatum	9/5
Mortars based on liquid glass	Aspergillus ustus, Aspergillus versicolor, Aspergillus fumigatus, Alternaria dianthi	4/2

II. Discussion And Conclusion

The results of the carried out experiments have been also confirmed that even containing in space air spores of microfungi may stocking on the surfaces of building materials and articles and use them, or contaminants on them as nutrient substrate. Growing on the surface of building materials and structures, microorganisms together with destructive effect deteriorate ecological situation in buildings and structures (lead to the appearance of moldy smell in the building and extract toxic products, allergic agents) [2, 5, 6, 8-16]. Growing on materials fungi extract a mass of spores and different products of vital functions, which are able to cause a number of serious diseases [primarily, mycoses – affection with fungal infection (for example, aspergillosis – disease caused by *Aspergillus* fungi)], which are heavily detected and more heavily treated. It is also known that 50 % of diseases of bronchial asthma in particular are connected with myxomycetes. They cause also penicillose, often accompanied by arthritis and ostitis. Therefore the growth of colonies of fungi affects not only building and industrial materials but also people, being in contact with them. This once more confirms the necessity of selective approach in sampling of building materials depending on specific operation conditions and implementation of preventive measures, eliminating or minimizing the possibility of stocking of microorganisms on them.

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