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RADIATION RESISTANT COMPOSITE FOR BIOLOGICAL PROTECTION OF THE PERSONNEL

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ABSTRACT

Results of scientific and practical justification of use as a part of designs of biological protection of polymer concrete on the basis of non - isocyanates polyurethane are presented. Influence of a ratio polymer is established: radiation - resistant filler on strength and protective properties of a polymer concrete. Regression dependences of change of the main properties of the specified polymer concrete on the power of the absorbed radiation dose are given. The synergetic effect in weakening of radiation when sharing a hydrogenous polymeric basis and radiation resistant powders is shown.

Key words: *polymer concrete, composite, radiation, biological protection, filler*

INTRODUCTION

Development of nuclear power, undoubtedly, is the evidence of progress. Atomic energy is rather cheap and it's receiving at the correct organization of technological process quite safely. Nevertheless, long operating experience of nuclear installations by various countries showed that along with increase of reliability of reactors it is necessary to take measures for increase of efficiency of biological protection of the personnel of stations. This article is devoted to development of the composite providing the solution of the specified task.

STATEMENT OF A PROBLEM IN A GENERAL VIEW AND ITS COMMUNICATION WITH IMPORTANT SCIENTIFIC AND PRACTICAL TASKS

Traditionally at the enterprises of the nuclear industry apply various metals and concrete with a density up to 4500 kg/m^3 with radiation resistant fillers [1] to biological protection of the personnel.
Besides, widely use combinations of the specified materials depending on the power of gamma radiations. At the same time, the standard methods of protection are sometimes insufficiently effective. It is caused by that penetration of the particles which are formed at nuclear reactions is not identical. So, for example, electrons, protons, the alpha particles and splinters of division having electric charge are quickly broken in rather thin layers of substance. Run the most getting from the listed particles – an electron with energy 5…6 MeV in aluminum no more than 1 cm. The main danger is constituted by neutral particles – neutrons and γ- quanta. At disintegration of elementary particles arise γ- quanta with energy of 70 MeV. When passing fast neutrons through substance and at their braking in a Coulomb field of kernels there can be photons with energy to several tens of GeV.

It is known that most intensively slow down fast neutrons of high energy to average energy elements which possess ability of inelastic dispersion of neutrons – for example, barium and iron [1]. Besides, availability of hydrogen as a part of material which not only slows down is necessary, but also takes slow neutrons.

Because at delay of fast neutrons heat is allocated, it is necessary to provide in addition thermal stability of material that is reached by introduction of radiation resistant filler.

At all clear advantages of the high density concrete which is used as a part of dry of protection of reactors there are also essential shortcomings: permeability for a neutron stream, considerable water absorption, and tendency to a Formation of cracks in the stretched zone [2].

Experience of use of reactors on fast neutrons showed that it is necessary to include ability of delay and absorption of various energy neutrons in the list of the additional requirements imposed to the developed materials [2].

The majority of polymers on the basis of which it is possible to receive various composites with the complex congenial properties [3] belong to such materials.

In this regard at a construction of protection against radiations all technical and economic advantages and shortcomings of various materials have to be carefully weighed. The preference has to be given to the materials possessing the properties solving for this design and conditions of construction. Owing to development of materials science by such materials composites on a polymeric basis are represented. The available experience of use of polymer concrete for protective designs showed their advantages in comparison with concrete on the basis of mineral knitting [3,4].

It is caused by the high durability, crack resistance and chemical firmness of the cured composites and the reduced terms of input of constructions in operation. The characteristics of the constructional materials providing the appropriate level of protection demanded in modern conditions cause the necessity of management of processes of their structuration for possibility of quality control.

Besides, at design of compounding of composites the integrated approach as besides providing the greatest possible linear coefficient of easing the developed material has to have the sufficient radiation firmness providing the set term of operation of a design of protection is necessary for biological protection.

In total the specified researches provide the solution of a scientific task – creation of effective composite materials due to the use of polymeric raw materials and modern technologies changing the mechanism of radiation protection and improving operational characteristics of the products made of these materials.

Practical task thus is the increase in service life of construction designs and reliability at their operation and providing the set extent of protection of the personnel.

THE ANALYSIS OF RESEARCHES AND PUBLICATIONS IN WHICH SOLUTIONS OF THIS PROBLEM ARE BEGUN AND ON WHICH THE AUTHOR LEANS

Modern scientific approaches to development of radiation resistant composites are based on rational use of a chemical composition and structure of polymeric and not polymeric materials.

Radiation firmness of polymeric materials depends on their chemical structure. Bigger firmness polymers with double communications and aromatic cycles, in comparison with polymers with saturated communications possess. It is explained by effect of dispersion of energy structural elements of macromolecules. This phenomenon is especially characteristic for polymers with the aromatic groups possessing a big set of power levels therefore ionizing radiation disperses on communications.

Besides, in such macromolecules due to existence of free electrons energy redistribution takes place. Lower radiation firmness polymers with communications of *C-F, C-Si, C-O* [3,5] possess.

There is an experience of use in radiation protection of glass [6], sulfur [7,8] and liquid rubbers [9]. It is shown that when it is desirable that protective materials possessed insulating properties, apply polymeric compositions with introduction to them of metal powders with a big density, such as tungsten, lead, pine forest, cadmium, iron [1].

Also there are enough examples of use as a polymeric basis of epoxies and the vulcanized rubber providing solution of the problem of corrosion, ultra-violet radiation, radiation and other adverse natural and techno genic factors [4,10].

In spite of the fact that now there is rather wide range of the materials capable a long time to resist to influence of radiation, it is represented to the most perspective polymer concrete, possessing a set of operational characteristics from which are distinguished high, almost universal chemical and radiation firmness, technological effectiveness and small shrinkage.
The available results on development of polymer concrete allow allocating them in row radiation resistant materials [11].

ALLOCATION OF PARTS OF A COMMON PROBLEM UNRESOLVED BEFORE

As all types of radiations cause chemical changes as a result of which collapse available in polymers and new communications are formed, use as a basis of a radiation protective composite of the polymer which is characterized by increase in hardness, durability and heat resistance at an initial stage of radiation with ensuring protection against a neutron stream is necessary.

Weakening of a neutron stream cannot achieve, having increased density and atomic number of substance that is traditionally used for weakening of gamma radiation – at the specified change the linear coefficient of absorption increases. For weakening of a neutron stream of such dependence does not exist.

The technology of delay and absorption of various energy neutrons consists of several stages. First, fast neutrons need to be slowed down to thermal as kernels badly absorb fast neutrons, and easy elements slow down neutrons of average energy to small energy more intensively. Then capture of slow neutrons is necessary. It is known that hydrogen not only slows down, but also takes slow neutrons [5].

Pine forest, cadmium and lithium are also effective for capture and absorption of slow neutrons [5].

Besides the requirements stated above the materials applied to the device of protection against radiations have to provide: minimum education and minimum energy of secondary radiations; in particular, the minimum exit γ- radiations with the minimum energy arising at capture of photons; the low induced radioactivity in protection; mechanical durability of material, first of all, on compression; rather low module of elasticity promoting reduction of tension by stretching from heating of inside layers of protection; the minimum thermal expansion providing solidity of a design and reducing tension; water tightness and gas-tightness, corrosion resistance; technological effectiveness (simplicity of installation and dismantle) and low cost [9].

The majority of these requirements are considerably mutually opposite that complicates optimization of structure of a composite on one criterion function.

In this regard we conducted the researches on specification of compounding aimed at providing long preservation of initial properties of polymer concrete for biological protection in the conditions of influence of radiation at the set durability.

PROBLEM DEFINITIONS

Effective radiation protection requires receiving the composite combining a polymeric basis with enough the hydrogen communications weakening and absorbing neutron radiation, and the radiation protective filler providing preset values of physic mechanical characteristics.

Thus special attention should be paid on compatibility of a filler and polymeric basis for prevention of a bulging and stratifying of ready mix.

Besides experience of the previous researchers showed that introduction to structure of polymer concrete only of a fine-grained filler as at production of designs of biological protection the molding technology and the corresponding equipment are applied is most effective is not adapted for supply of mix with large filler.

MATERIALS, METHODS OF RESEARCH AND MAIN SCIENTIFIC RESULTS

Receiving radiation - a protective composite agrees to the scientific hypothesis made by us perhaps at combination of the hydrogenous basic polymeric basis possessing sufficient radiation firmness and the high density radiation protective filler ground to a necessary state. For this purpose it is necessary to combine the available polymeric basis with the high content of hydrogen with radiation resistant filler. The rational ratio of the specified components will provide protection, both from neutron, and from gamma radiation at the set durability.

Besides, it is necessary to ensure appropriate ecological safety, both by production of a design of biological protection, and at long operation.

Basis of selection of a compounding of a composite on a polymeric basis for production of designs of biological protection are known provisions of the polystructural theory of V. I. Solomatov and the concept of system and structural approach in material quality management assuming transition from the principle of a fragmentariness to complexity at which structure of material, technology of products and designs are presented in the form of the interconnected systems.

At a choice of a polymeric basis the class of the connections possessing set of the properties providing the requirements given above – the polyurethane received by a cold Concreting of oligocyclocarbonate is revealed by the conducted researches (trademarks Laprolat 803, Laprolat 301, Laproksid 503M). Laprolat represent products of carbonization of epoxy derivative polyoksypropylen (laprol) and have trailer cycle carbonate groups. Laprolat are cured on reaction of an urethane – cycle carbonate – amine with primary aliphatic or cycle aliphatic amines, forming hydroxyl urethane structure [5,12].

Properties of the received non - isocyanates polyurethane are caused by existence of interactions of specific character (hydrogen communications, communications of ionic type) and nonspecific (a dipole - dipolar, van-der-vaals of interactions, and also crystallization) which total contribution to formation of a complex of properties of polyurethane is defining. At formation of hydrogen communications by donors of protons Hydrogen atoms of urethane groups, serve in a case polyurethane-urea and polyamidourethane – Hydrogen atoms of the relevant functional groups. Acceptors of protons are carbonyls of the listed groups [13].

Existence of a large amount of hydrogen as a part of polymer promotes effective capture of fast neutrons and decrease in density of a fluency of the specified particles. At the same time, the specified process is followed by radiation self-heating that negatively affects strength characteristics of material. It is obvious that it is necessary to provide at the same time both effective biological protection, and resistance to temperature influences. These issues are resolved in common as introduction of the filler having high linear coefficient of easing promotes decrease in heat generation in a body of a polymer concrete. By preliminary researches it is established that the most perspective fillers are fine radiation resistant powders [14,15].

The key characteristic of the developed material is the radiation linear coefficient of easing (LCE):

$$\mu = n \cdot \sigma, \qquad (1)$$

where n – number of atoms of material of an absorber in unit of its volume, σ – section of interaction of radiation with one atom of substance, depends as on properties of material, and energy of radiation.

Size σ(Z, E) depends on Z – atomic number of substance and energy of radiation – E.

For difficult substances LCE it is possible to present expression:

$$\mu = \mu 1 \cdot a1 + \mu 2 \cdot a2 + \ldots + \mu i \cdot ai, \qquad (2)$$

where a1, a2, ai – mass fractions of each of components.

Values σ can be calculated on the known formulas which are coordinated with experimental data within three percent. The main contribution to the calculated error of LCE is made uncertainty of values α_i and absorber material density variations. Obviously, this factor is especially essential in case of the considered composite materials.

In this regard it is important to receive the values of LCE which are directly measured for the interesting range of energy of radiation. In this work the interval of values made 0,1 … 1,5 MeV as the main radioactive isotopes defining a radiation dose have energy of radiation in this interval.

At design of radiation protective composites it is necessary to consider that fact that introduction of fillers on the one hand, provides composite durability, with another – cuts a consumption of the polymeric component containing hydrogen communications.

It is obvious that it is necessary to receive the balanced combination of the specified components providing the greatest possible linear coefficient of easing it agrees (2) at the set durability.

We carried out the comparative analysis of physical and chemical characteristics of a ground serpentines, barite, limonite, magnetite and glass powder of Sodium boron silicate structure, degree of their compatibility with polymeric binding is estimated. The specified powders were chosen owing to that they are traditionally used in radiation protection.

Ranging of fillers in size of a specific surface of specific surface (Sss), oil-absorption power and value pH a water extract is presented in Table 1.

Table 1. Physical and chemical characteristics of fillers

Name	Average density, kg/m^3	S_{ss}, m^2/g	The chemical structure	Oil-absorption g / 100 g	*pH* water extract
serpentinit	2300 … 3200	300 … 350	$Mg_3\ (OH)\ 4Si_2O_5$ or $3MgO2SiO_22H_2O$	28…60	9,1…10,3
barite	2900 … 3600	280 … 310	$BaSO_4$	30…50	6…8
limonite	2400 … 3700	310 … 340	$Fe_2O_3\ H_2O + Fe_2O_3\ 2H_2O$	40…56	5…5,5
magnetite	3250 … 4100	290 … 340	Fe_3O_4	35…50	5,5…6,2
powder glass	Bulk density 600 … 900	microspheres 15 … 200 microns	$Na_2O \approx 7{,}5\%$ $B_2O_3 \approx 12\%$ $SiO_2 \approx 80{,}5\%$	9…11	6,6…7,4

At selection of a subtlety of a grinding of particles of filler meant the following: tendency of particles to agglomeration which increases with increase in a specific surface of a filler; sedimentation of particles which is accelerated with reduction of a specific surface, increase of density of a filler and decrease in viscosity of the binding.

Establishment of a rational ratio of a polymeric basis and filler was carried out taking into account three indicators: the set viscosity providing a molding method of application, durability at compression (reinforced concrete durability is not lower), radiation firmness.

In researches used two-component polymeric binding [16].

Component A - epoxy dyane - ED-20 (ED-22) in accordance with GOST 10587 with EDOS a thinner – softener on TU 2493-003/3004749-93, with a density of 1,16 … 1,25 kg/m^3. A component B - adduct laprolat-803 (TU 6-05-221-995-88 with a mass fraction of cycle carbonate groups – 21 … 31, epoxy groups - 2,5%, *pH* 3,5…5,5) with amines.

Test of radiation firmness of samples was carried out on the GURH-1000 installation with energy of gamma radiation of a radioactive source of Co^{60} *1,25* MeV.

Change of durability of samples on compression depending on various absorbed gamma radiation doses was the controlled parameter defining radiation firmness. The necessary total absorbed doses were gathered depending on exposition time in γ-quanta a bunch. Temperature of samples in the course of a dose set due to radiation heating did not exceed 45 °C.

Experimental measurements of linear coefficient of easing were taken a linear 4096-channel semiconductor gamma spectrometer with diffusive *Ge-Li* the semiconductor detector DGDK-160V.
Tests carried out according to the technique stated to Perekalsky O. E. in the corresponding qualification work [9].

At the first stage researches of a contribution of fillers to change of viscosity of mixes are conducted. It is established that introduction of ground fillers significantly increases viscosity of mix, even in the conditions of course of exothermic reaction between polymer and components of the curing group. As introduction of softeners of the chemical nature to composition of radiation protective materials irrationally, we considered a question of use of softener of physical type – glass powder.

The choice of the specified softener is caused by a variety of reasons. First, existence in composition of powder of microspheres provides considerable decrease in resistance of mix to viscous shift. Secondly, the chemical composition of powder conforms to requirements for radiation protection.
The analysis of results of experiment allowed drawing a conclusion that at joint introduction of fine fillers viscosity η changed according to dependence:

$$\eta = \eta_0 + Ax_1 + Bx_2, \qquad (3)$$

where η_0 – initial viscosity of polymer, Pa · s; x_1, x_2 –quantity of the entered components, mass. %; *A*, *B* – the coefficients characterizing a contribution of each filler.

On the basis of (3) counted combinations of various fillers to glass powder. As a result of calculation of possible combinations borders of a variation of the volume maintenance of fillers at the set quantity of the binding were defined.

Considering difficult structure of a polymeric basis of a composite it is rather difficult to judge chemise of the processes proceeding at interaction of the specified powders with polymer. Nevertheless, the comparative analysis of the obtained data allowed to reveal the most acceptable filler – limonite.

Due to the above, we made the assumption of decrease in viscosity of mix at complex introduction of limonite and glass powder as glass powder, even at big concentration; owing to a spherical form of particles and small oil-absorption power plays a sliding layer role between the main filler and binding.
As a result of the made experiments it is established that at combination of fillers with binding in the conditions of a big difference in *pH* (for example, at introduction of a serpentine) perhaps formation of

pores a composite owing to allocation of gaseous products of collateral reactions in an interface. It is established that fillers have to meet a condition *8> pH > 5*.

At the second stage we conducted researches on establishment of ratio binding – filler. It should be noted that in reference books there are no the absorption given about degree glass powder of radiation. In this regard LCE assessment not actually of powder, and a composite in general was carried out. As criterion functions were appointed extent of easing scale - radiations, durability at compression (R_c) and viscosity of mix (η).

Applied two-factorial two-level experiment to establishment of a rational combination of components. As the factors influencing the composite key properties were appointed: quantity of a powdery filler of the corresponding dispersion (x_1), quantity a glass spheres (x_2), in mass fractions from the mass of the polymeric binding.

Taking into account that combination of components of the curing group and a polymeric basis is exothermic reaction, mix temperature at introduction of powders was fixed at the level of 30 ^{0}C. Control of decrease in temperature was exercised spirit thermometers. Measurements of viscosity were taken at two values of temperature: 30 and 20 ^{0}C.

Establishment of a rational ratio "binding – filler" was carried out by means of two-factorial, three-level experiment. Data of the previous experiments allowed us to establish the maximum and minimum extents of filling corresponding to threshold value of viscosity. The plan of experiment with the received values of criterion functions is presented in Table 2. In respect of designation "+", "-", "0" correspond to the top, lower and average level of factors, respectively.

Table 2. The plan of carrying out two-factorial three-level experiment
with the received values of criterion functions

No. of a line	x_1	x_2	R_c *, MPa	LCE *, cm^{-1}	η* (Pa·s) at a temperature:	
					20^{0}C	30^{0}C
1	+/3	-/1	47,2/47,61	0,665/0,651	141/140	70/72
2	+/3	+/3	58,5/58,77	0,890/0,921	177/171	78/77
3	-/1	+/3	47,5/47,33	0,335/0,393	88/92	55/58
4	0/2	+/3	53,6/53,05	0,689/0,657	131/128	64/68
5	0/2	-/1	41,3/41,89	0,369/0,387	97/98	55/59
6	+/3	0/2	53,4/53,19	0,775/0,786	135/133	74/73
7	-/1	0/2	42,5/41,75	0,265/0,258	70/68	51/57
8	0/2	0/2	46,8/47,47	0,538/0,522	110/108	60/66
* over line experimental values, behind line – the regressions received on the equations						

As a result of processing of experimental data the regression equations for change of viscosity, linear coefficient of easing and durability were received at compression:

– at a temperature of 30^{0}C: $\eta=28,66+11,8x_1+4,5x_2;$ (4)

–at a temperature of 20^{0}C: $\eta= - 3,633+40,18x_1+17,35x_2;$ (5)

$$LCE = - 0,276+0,264x_1+0,135x_2; \quad (6)$$

$$R_c = 24,87+5,72x_1+5,58x_2. \quad (7)$$

Check of the received equations by Fischer's criteria and criterion of Stjudenta showed their adequacy and reliability.

Owing to that the regression equations are linear to optimize structure in any chosen parameter rather difficult. In this regard optimization was performed in common, on *LCE* and viscosity.

As appears from the equation (6), the contribution of fillers to value of *LCE* is ambiguous. Extent of weakening of gamma radiation powder of limonite is slightly higher, than at powder glass. At the same time, the received values of *LCE* of a composite in general considerably exceed total values of *LCE* of the specified fillers. It is obvious that sharing of powders of limonite and glass as a part of the developed polymer concrete causes synergetic effect, mutually strengthening effect of easing γ-

radiation. In our opinion the specified circumstance is explained by the contents as a part of a composite various by the nature and structure of the components providing in total effective radiation protection.

The comparative analysis of influence of concentration of the entered powders on durability of the cured composite showed that in general the contribution of the specified powders to durability is identical. Coefficients of the equation (5) are almost equal: 5,72 and 5,58 respectively.

It is obvious that mechanical hardening is realized generally due to streamlining of structures of a composite that is by formation of clusters round the active centers.

In general the tendency of increase in durability at increase in extent of filling is observed. Along with growth of durability value of *LCE* increases, that is «the law of an alignment» is realized. It is theoretically possible to receive extent of filling to 90% on volume, however introduction of such quantity of filler requires temperature increase binding up to the sizes at which the speed of a concreting will increase in hundreds of times that will not allow providing viability of mix.

As process of mixture of a ground filler and polymeric basis happens at a temperature up to 300C, the quantity of the entered filler is much less greatest possible. However, as it was specified already earlier, temperature increase of reactionary mix inevitably leads to acceleration of a concreting. In this regard influences of temperature of mix on its viability conducted researches. The specified parameter was estimated on change of viscosity of mix in time at the fixed temperature.

So at a temperature of 50^0C reaction of a concreting began already 17 minutes later after the end of hashing, and at a temperature of 40^0C the concreting began from 34 minutes. As a result of the conducted researches the rational range of temperature of mix – from 30 to 35^0C is established.

Taking into account ensuring the set viscosity the quantity of the entered components made: limonite 30 –and powder glass Sodium boron silicate structure – of 30 mass. %. By results of tests of three series on nine samples of the received composition of material in the conditions of short-term loading the calculated and standard values of strength characteristics of a radiation resistant composite presented in Table 3 are established.

Table 3. Standard and settlement characteristics radiation - a resistant composite

Characteristics	Value, MPa
The cubical durability at compression with security 0,95	58
Durability on stretching at a bend with security 0,95	5,7
Standard prismatic durability at compression	46,4
Standard module of elasticity	18720
Settlement resistance to compression	52,7
Settlement module of elasticity	15730

The average density of the tested samples made 1280 kg/m^3.

For the developed composite values of basic characteristics in the conditions of influence of ionizing radiation are defined.

First of all defined easing coefficient γ - radiations, as the qualitative characteristic.

Radiation firmness was defined by keeping of samples 2,5 cm thick to the set dose of ionizing radiation with the subsequent control of change of values of basic characteristics: durability at compression of R_c, the elasticity module at compression E, linear coefficient of easing μ.

The changes at radiation exposure of the specified characteristics expressed through the corresponding coefficients K_{rd} and K_E, indirectly testify to weakening of communication between a filler and organic binding owing to degradation of properties of the last. Results of the conducted researches are presented in Table 4.

Table 4. Radiation dependent change μ, strength and the module of elasticity at compression

Dose γ-quant, $P \cdot 10^6$	$K_{rd} = R_c/R_{c, initial}$	$K_E = E/E_{initial}$	μ, cm^{-1}
10	1,0	1,0	0,26 ± 0,02
50	1,0	1,0	0,14 ± 0,01
250	0,94	1,0	0,12 ± 0,01
500	0,91	0,98	0,11 ± 0,01

CONCLUSIONS AND PROSPECTS OF FURTHER RESEARCHES

Influence of gamma radiation and neutron radiation affects first of all organic part of a polymer concrete that is confirmed by emergence on the strip IR spectrum at 1730 cm^{-1} indicating destruction of double communication and oxidation of unsaturated hydrocarbon with formation of $S=O$ – of communication. Thus, it is possible to believe that there is a structural modification binding, providing an additional stitching of molecular chains and hardening of a composite in general.

At the same time, material is considered radiation resistant if its indicators decrease no more than by 25% of the initial value. The endurance of samples of a non - isocyanates polymer concrete set by us corresponded about 7…8 years of operation in the conditions of rigid radioactive effects. Taking into account that values of strength characteristics for the first two-three years increased, it is possible to predict reliable biological protection of the personnel against radiation exposure at least for not less than 10 … 12 years.

In further researches detailed studying of heat physical characteristics of the developed composite for ensuring rational design of construction designs is supposed.

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