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Some problems of the new radiation-resistant rubber mechanics in vibrating machines at hard γ-radiation

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Annotation. Today, the sources of ionizing radiation are present in various spheres of human activity: during the uranium ore extraction, processing and benefication, in medicine, at exploration of outer space, in nuclear power plants, etc. Here rubber is used as insulating and protective materials, sealers, elastic links, dampers and wear-resistant linings of machines. Therefore, a problem of creating new radiation-resistant rubbers capable to operate in a radiation zone for a long time with no significant changes in their physical and mechanical parameters and durability is of great importance. In this work, some aspects of creating such rubber for use in vibrating machines (screens, feeders, conveyors) operating under extreme conditions while mining and processing of uranium ores are discussed. In our case, the extreme conditions include high long-term stationary cyclic loads, high and low temperatures and exposure to hard γ-radiation. Under such conditions, rheological and fatigue characteristics of rubber differ from those demonstrated under normal conditions, and such factors as dissipative heating, changes in characteristics over time (aging effects), development of damageability, etc., become essential and must be taken into account when choosing a proper type of rubber.

1. Introduction

Ukraine is among the top ten world uranium producers; a specialized enterprise in the city of Zhovti Vody produces natural uranium concentrate that meets all requirements of international standards. As of today, 15 energetic blocks of the nuclear power plants are in operation in Ukraine with total capacity of about 15 million kW, which generate more than half of the total electricity volume produced by the country [1].

Various vibrating machines are widely used for the uranium ore extraction, processing and benefication: vibrating feeders are used for underground uranium mining and conveying; ball mills and vibrating screens are used in benefication processes; one- and two-pipe vibrating conveyors – for processing beneficiated raw materials (including reprocessing of the spent fuel cells from the nuclear power plants). In all these machines, rubber elements are used as elastic links, vibration insulators, protective linings, elastic elements of drive couplings and sealers.

Below is an example of two-pipe vibrating conveyors of the KB2T type used for transporting radioactive (including dusty and toxic) materials. The KB2T conveyor is a two-mass near-resonance system consisting of two transporting pipes (figure 1) interconnected by the links – the rubber-metal blocks (RMB). The pipes are connected to the conveyor frame by means of the rubber-metal hinges installed in the support units. An eccentric drive with an elastic connecting rod is connected to the lower pipe. In order to ensure a leak-free transportation of aggressive materials, special rubber sealers are installed in the loading and unloading points. Usually, the conveyors are mounted on the vibration insulators [2] in order to protect their foundation against dynamic impacts.

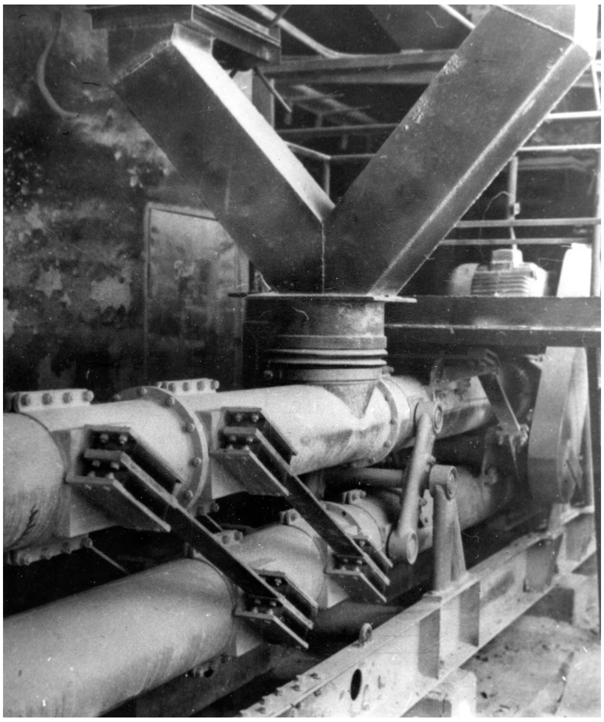


Figure 1. Vibrating conveyor of the KB2T type.

Due to the specificity of the processed mineral raw materials, which feature a rather high radioactivity, stricter requirements are stipulated for the conveyors both for their rubber elements durability and reliability and stability of their elastic-viscous characteristics, and for stability of amplitude-frequency characteristics of the machines during their operation. In addition, it is required to provide stable operation of the vibrating machines without constant presence of human operators, especially in conditions of hard γ-radiation. Besides, rubber elements should operate for at least 20-25 thousand hours before their failure and, at the same time, their rigidity characteristics should be stable within the range of 15-25% under the following intense cyclic loading mode: relative shear deformation of the main elastic links should be not less than 0.20-0.25 at loading frequency of up to 10-12 Hz and amplitude of the machine's work members vibrating of more than 10-12 mm. These working conditions are explained by specific character of the technological processes.

Under such operating conditions (with no irradiation), the rubbers known in engineering practice did not meet these requirements at all: their mean time between failures (MTBF) was not more than 800-1200 hours, and their rigidity was increased during this time by 60-80 % [2].

When these rubbers were irradiated (by doses of 0.3-0.6 MGy), their durability decreased significantly.

Therefore, it is very important to create the new radiation-resistant rubbers with stable physical and mechanical parameters during long-term operation of machines in extreme conditions.

Researches in this direction were begun in the second half of the last century [3-6]: recently, they have been intensified due to the expansion of space researches [7, 13], increased volume of uranium ores mining and benefication [1] and operation of nuclear power plants [8, 9, 12]. In these studies, the emphasis is on the creation of the new radiation-resistant materials for further manufacture of such products as sealing elements, electricals for various technological purposes [8-12], and others. Publications on this issue testify to the relevance of the works and good progress in the field of mechanochemistry of composite materials.

At the same time, there are no studies in the field creating of radiation-resistant rubbers for massive elements of machines operating in extreme conditions; in the literature known, there are no publications on this problem for the last forty years. To some extent, this article is intended to fill the gap.

The purpose of our work is to create a new radiation-resistant rubber for vibrating machines operating under extreme conditions of loading and action of hard γ-radiation; and to prove resistance of the new rubber to radiation by the long-term industrial tests.

2. Methods

2.1. Simulation model

In context of our research, we consider rubber as a linear viscoelastic medium with high dissipation and large reverse deformations; in this case, the viscoelastic behavior is determined by the presence of time effects, and linearity is based on the fact that deformation is proportional to the stress for any fixed moment in time. Further, we also assume that elastomers refer to elastic-hereditary media, for the calculation of which classical theories of elasticity are not always applicable due to the presence of viscous component, memory of previous influences and instability of physical and mechanical characteristics over time, the so-called aging effects. In order to avoid these disagreements, we use the Boltzmann-Volterra principle, according to which relationship between the stress σ and deformation ε is most fully determined by integral relations with the kernels of relaxation and aftereffect [14].

2.2. Mathematical model

Mechanics of rubber as an elastic-hereditary medium is well studied; therefore, below are given the final expressions used for calculating the material rheological characteristics and determining an over-time changes in vibration amplitude of the KB2T vibrating conveyor with elastic links irradiated or not irradiated by a certain dose of γ-radiation.

For the case when rubber is described by the Boltzmann-Volterra integral relations with relaxation and aftereffect kernels, the equation of vibrating mass flow can be written in an operator form in the following way [14]:

$$\ddot{x} + C_t x = q_1 \sin \omega t,$$

where x is coordinate; C_t is operator of rigidity of the conveyor elastic suspension, N/m; q_1 is force of inertia per unit of vibrating mass, N/kg.

$$C_t = C_0 \left[1 - \chi \Re_\alpha^* \left(-\beta \right) \right], \tag{1}$$

$$\Re_\alpha^* (-\beta) \varepsilon(t) = \int_0^t \Re_\alpha \left(-\beta, t - \tau \right) \varepsilon(\tau) \, \mathrm{d}\tau , \tag{2}$$

$$\Re_\alpha(-\beta, t-\tau) = (t-\tau)^\alpha \sum_{n-0}^{\infty} \frac{(-\beta)^n (t-\tau)^{n(n+2)}}{\Gamma[(n+1)(1+\alpha)]}, \tag{3}$$

where C_0 is instantaneous value of elastic suspension rigidity, N/m; $\Re_\alpha(-\beta, t\text{-}\tau)$ is the Yu. Rabotnov exponential function of fractional order; α, β, λ are rheological parameters of rubber; Γ is gamma-function.

In [14], the following basic relationships were obtained for determining the rubber rheological parameters:

$$\psi = 2\pi B(\omega); \tag{4}$$

$$\frac{G(\omega)}{G_0} = 1 - A(\omega); \tag{5}$$

$$A = \frac{\omega^{1+\alpha}\cos\delta + \beta}{\omega^{2(1+2)} + 2\omega^{1+\alpha}\beta\cos\delta + \beta^2}; \tag{6}$$

$$B = \frac{\omega^{1+\alpha}\sin\delta}{\omega^{2(1+\alpha)} + 2\omega^{1+\alpha}\beta\cos\delta + \beta^2}; \tag{7}$$

$$\lambda = \frac{G_0 - G_\infty}{G_0}; \quad \alpha = 1 - \frac{4}{\pi}\operatorname{arctg}\frac{\psi_{\max}}{\pi\lambda}; \tag{8}$$

$$t_0 = \left[\omega(\psi_{\max})\right]^{-1}; \quad \beta = \left[t_0^{1+\alpha}\right]^{-1}; \quad \chi = \lambda \cdot \left[t_0^{1+\alpha}\right]^{-1},$$

where A and B are rheological characteristics of rubber (the Fourier sine- and cosine- transforms of the fractional exponential function); ψ is coefficient of energy dissipation; $G(\omega)$ is current value of the shear modulus, N/m^2; G_0 is instantaneous value of the shear modulus, N/m^2; t_0 is the generalized time period of relaxation, s.

With taking into account the above relationships, the expression for the vibration amplitude for a fixed value of time will have the form:

$$a = q_1 \left\{ \left[\omega_{0u}^2 (1 - \chi A) - \omega^2 \right]^2 + \chi B^2 \omega_{0u}^4 \right\}^{-1/2}, \tag{9}$$

where ω_{0u} is frequency of natural vibrations of an ideally elastic system, rad/s.

2.3. Samples and experimental facilities

In our studies, we used standard double-sided blades and full-scale rubber-metal elements RMB102. Standard samples were used for determining the filler effects on the properties of the vulcanizates and on the choice of rubber composition; the tests were carried out in accordance with current standards. The RMB102 elements were used for determining rheological and fatigue parameters of rubber; the samples were selected by value of the conditionally equilibrium shear modulus; parameters variety in the lot consisting of 8-64 elements did not exceed 2.5 %. Parameters of rigidity and dissipation were registered by the original dynamic testing stands, including the universal stand “Instron 1126” (England) with recording accuracy of ±0.3 % for load and ±2.5 % for deformation [14].

The RMB102 fatigue characteristics were studied during the long-term tests (from 2 years to 5 years) on the KB2T conveyor stands at their industrial operation on transporting of loose, dusty and toxic materials in the vibration mode: vibration amplitude was 10.5 mm (relative shear was 0.21) and frequency was 11.0 Hz.

The RMB102 elements were exposed to the 0 MGy, 0.1 MGy, 0.3 MGy, 0.6 MGy and 1.0 MGy doses of radiation in special equipment with using the compound C_0^{60}; dose of the absorbed radiation was determined by the known spectrophotometric methods.

The absorbed dose of radiation, at which the key information characteristics of the material were within the permissible values, was taken as a quantitative indicator of the rubber resistance to radiation. According to the GOST 9.701-79, these characteristics include tensile strength, relative elongation at break, hardness and endurance under repeated tension for the standard samples, and dynamic shear modulus, coefficient of energy dissipation, temperature of dissipative heating and durability for the massive elements of the RMB type. Dose of 0.3 MGy was accepted as the maximum absorbed radiation dose as it mostly corresponded to the conditions where the considered vibrating machines operated.

2.4. Creation of the radiation-resistant rubber

The work on creation of the radiation-resistant rubber was carried out in three stages. At the first stage, an effect of the fillers on physical and mechanical properties of the rubber based on the synthetic isoprene caoutchouc SKI-3 was investigated. The tested mixtures had the following composition (in mass parts) per 100 mass parts of caoutchouc: 1.5-2.0 of sulfur; 0.8-1.0 of CBS (N,N-dicyclohexyl-2-benzothiazolesulphenamide); 5-25 of carbon black P803; 1.0 of stearic acid; 5.0 of zinc oxide; 2.0 of neozone D (phenyl-2-naphthylamin); 2.0 of 4010NA (N-isopropyl-N'-phenyl-p-phenylene). A uniform vulcanization of the samples throughout their volume was achieved at a temperature of 140-146 °C.

The effect of carbon black on the properties of vulcanizates (standard double-sided blades) is shown in table 1. As it can be seen, vulcanizate No. 1 had the best combination of dynamic and fatigue properties. From these mixtures, rubber-metal blocks RMB102 were made and then were tested on the KB2T conveyor stands under operating conditions with vibration amplitude of 10.5 mm (relative shear of the elements was 0.21) and frequency of 11.0 Hz.

Table 1. Influence of the carbon black type on the properties of vulcanizates made of SKI-3.

	Carbon black P803		
Indicators	No. 1 5 mass parts	No. 2 15 mass parts	No. 3 25 mass parts
Breaking strength, MPa	29.0	28.0	27.0
Relative elongation, %	780	760	650
Residual elongation, %	14	18	18
Shore A hardness	38	44	46
Dynamic modulus at impact tension, MPa	2.17	2.40	3.60
Internal friction modulus at impact tension, MPa	0.21	0.30	0.40
Heat generation, °C	38-40	46	51
Multiple stretching endurance, thousand cycles	960	840	725

The research results are shown in table 2. As one can see, the best dynamic and fatigue properties were shown by the rubber-metal elements made of the rubber No. 1. At the same time, these elements featured insufficient stability of their mechanical characteristics over time: for example, dynamic rigidity of the elements was increased by 80 % after 15,000 hours of loading, which does not meet technological requirements prescribed for the KB2T conveyors operating in extreme conditions. The elements made of rubbers No. 2 and No. 3 with high content of carbon had shorter durability, and their dynamic characteristics were less stable over time.

At the second stage of the research, in order to improve dynamic endurance and reduce heat generation, 0.5 mass parts of the elastopar (N-methyl-N,4-dinitrosoaniline) was added into the low-filled vulcanizate No. 1 (5.0 mass parts of P803 carbon black). The RMB102 blocks made of this rubber were tested for 4 years; their initial dynamic and fatigue characteristics were as follows: dynamic shear modulus was 0.58 MPa; steady-state temperature of dissipative self-heating in central area of the RMB block was 50 °C; energy dissipation coefficient was 0.35. At the end of the stage, durability accounted more than 35,000 hours without any visible signs of destruction and increase of rigidity characteristics was within the range of 17-25 %.

Table 2. Dynamic characteristics and durability of the RMB102 blocks.

	Rubber		
Indicators	No. 1 Carbon black P806 (5 mass parts), zinc oxide (5 mass parts)	No. 2 Carbon black P803 (15 mass parts), zinc oxide (5 mass parts)	No. 3 Carbon black P803 (25 mass parts), zinc oxide (15 mass parts)
Dynamic shear modulus, MPa	0.57	1.02	0.89
Energy dissipation coefficient	0.35	0.37	0.30
Steady-state temperature inside the RMB, °C	50-55	75-80	70-75
Durability, h	15000	6000	4700
Product characteristics	No visible signs of destruction	were withdrawn from testing due to instability of dynamic rigidity	were withdrawn from testing due to the rapid growth of fatigue cracks

At the third stage, an effect of radiation on the dynamic and fatigue properties of rubbers with different rate of filling with carbon black was investigated (table 3). The RMB102 blocks were made of these rubbers, then they were exposed to the γ-radiation by doses of 0 MGy, 0.1 MGy, 0.3 MGy, 0.6 MGy, 1.0 MGy and tested under operating conditions on the KB2T conveyor stands for 5000 hours. The results of measurements of dynamic shear odulus and coefficient of energy dissipation depending on the radiation dose are presented in table 4 and table 5. As it can be seen, up to the dose of 0.3 MGy, rigidity and dissipative characteristics of the blocks have changed insignificantly: for rubber No. 1, this change was less than 8 %. All blocks were in workable condition.

Table 3. Content of fillers in the studied rubbers, in mass parts.

	Rubber No.		
Fillers	No. 1	No. 2	No. 3
Carbon black P803	5.0	15.0	25.0
Zinc oxide	5.0	5.0	15.0
Elastopar	0.5	0.5	0.5

Table 4. Dependence of dynamic shear modulus in MPa on the radiation dose for the RMB102 blocks at relative shear of 0.21 and loading frequency of 11.0 Hz.

	Dose, MGy				
Rubber	0.00	0.10	0.30	0.60	1.00
No. 1	0.58	0.59	0.60	0.64	0.67
No. 2	1.02	1.06	1.08	1.20	1.29
No. 3	0.89	0.95	0.97	1.01	1.12

Table 5. Dependence of energy dissipation coefficient on the radiation dose $\psi(D)$ for the RMB102 blocks at relative shear of 0.21 and loading frequency of 11.0 Hz.

	Dose, MGy				
Rubber	0.00	0.10	0.30	0.60	1.00
No. 1	0.35	0.33	0.30	0.25	0.23
No. 2	0.37	0.30	0.25	0.15	0.12
No. 3	0.30	0.25	0.20	0.15	0.11

Comparative tests of the RMB102 blocks made of rubber with no irradiation and rubber with elements irradiated by dose of 0.3 MGy were also carried out; the blocks were tested in operational conditions for about 5 years during the transportation of dusty and toxic materials. The test results showed that durability of the blocks before their failure did not differ significantly. Therefore, rubber No. 1 was recommended as the most appropriate to the technological requirements stated for elastic elements in vibrating machines operating under conditions of hard radiation.

3. Discussion of the results

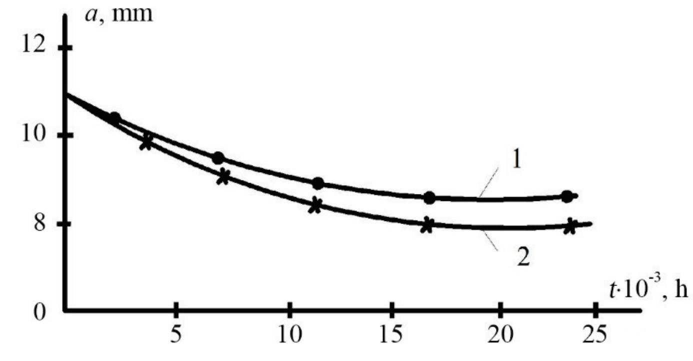
The experimental studies of the rubber No. 1 dynamic characteristics (table 6) and calculation of its rheological parameters by using formulas (1-8) show that irradiation increases the rubber rigidity and reduces dissipation of energy: this indicates that structurization reaction predominates in the material. In vibrating machines, action of the γ-radiation leads to a change in the amplitude-frequency characteristics: in this case, drift of the resonance frequency peak and decrease of the vibration amplitude over time are recorded.

Table 6. Rheological parameters of rubber No. 1 (RMB102).

	Rheological parameters of rubber				
Rubber No. 1	α	β	λ	ψ	G_d
No radiation	-0.64	0.89	0.35	0.35	0.58
Absorbed dose	-0.64	0.91	0.39	0.30	0.60

In [2], a method is given for calculating the time dependence of amplitude of the KB2T conveyor vibration with taking into account damages developing in the elastic links. By using this method and equation (9), the time dependence was calculated for a similar conveyor, elastic links of which received a one-time radiation dose of 0.3 MGy; a damage caused by the rubber aging was also taken into account. The calculation results show that for a time interval of 25000 hours the final vibration amplitude in the two considered cases differed by 10-12 % (figure 2).

Comparative performance data obtained under industrial conditions for transporting dusty and toxic materials confirms the analytical calculations.



1 – without irradiation; 2 – dose of 0.2 MGy of hard γ-radiation

Figure 2. Dependence of amplitude of the KB2T vibrating conveyor vibration on the duration of its operation.

As one can see, hard γ-radiation negatively impacts on the performance of machines with rubber elements: damage of rubber caused by irradiation changes its structure, increases its rigidity and reduces its ability to dissipate energy; the rubber aging and growth of fatigue cracks in it are accelerated. In addition, air is decomposed and, consequently, ozone is formed, which contributes to the oxidation of metal parts in machines and to occurrence of cracks on the surface of the elements. As a ult, the amplitude-frequency characteristics of machines are changed, and amplitude of their vibrations decreases, which is unacceptable for a number of technological processes, for example, during reprocessing of spent fuel cells from the nuclear power plants.

Though the created radiation-resistant rubber does not fully eliminate the above noted affects, it, nevertheless, allows to stabilize amplitude of the machine vibrations within the range of 10-12 % at certain absorbed radiation doses (in the considered case, it is up to 0.3 MGy).

In practice, the low-modulus rubber No. 1 is widely used in vibrating equipment operating under long-term cyclic loading when vibration amplitude is up to 12.5 mm (relative shear is up to 0.4) at loading frequencies of 10-12 Hz; it is also used in vibrating machines, which, due to technological conditions, operate in aggressive environment, including hard γ-radiation, and, therefore, should feature stability of amplitude-frequency characteristics in the course of time. As for stability of the rubber No. 1 mechanical parameters, long-term industrial tests show that in case of no radiation blocks of the BRM type installed in the conveyors of the KB2T type (at $a = 10.5$ mm, $f = \omega/(2\pi) = 11$ Hz) have the MTBF of at least 35,000 hours, i.e. for about 4-5 years, and at least 25000 hours in case of irradiation by dose of up to 0.3 MGy. The rubbers No. 2 and No. 3 are also used in industry as elastic links in vibrating machines operating in less intense loading mode (at $a = 6\text{-}8$ mm, $f = 10\text{-}12$ Hz) and with no hard γ-radiation.

Rubber No. 1, with carbon black P803 replaced by carbon black N220, was patented in Ukraine [15] with the following ratio of components, in mass parts: 100 of caoutchouc SKI-3 (cis-1,4-Polyisoprene); 1.5-2.0 of sulfur; 0.8-1.0 of CBS (N,N-dicyclohexyl-2-benzothiazolesulphenamide); 5-15 of carbon black N220; 1.0 of stearic acid; 5.0 of zinc oxide; 2.0 of neozone D (phenyl-2-naphthylamin); 2.0 of 4010NA (N-isopropyl-N'-phenyl-p-phenylene); 0.5 of elastopar (N-methyl-N,4-dinitrosoaniline).

4. Conclusions

1. A new radiation-resistant rubber was created for vibrating machines operating under extreme conditions of loading and exposure to hard γ-radiation.

2. Results of the long-term laboratory and industrial tests made it possible for the first time to reveal a very important regularity: rubbers, which are resistant to aging under long-term dynamic loads, are also resistant to the action of γ-radiation, which indicates the similarity of the mechanisms of changing the structure of the material and the predominance of the structurization reactions.

3. It is established that γ-radiation affects the amplitude-frequency characteristics of vibrating machines: during long-term operation, a drift of the resonance frequency peak and a decrease of vibration amplitude are observed over time leading to decrease of the machine productivity and, in some cases, to a breakdown in processes. In practice, this undesirable phenomenon is eliminated by resetting of the machine dynamic parameters or replacing elements of its elastic suspension.

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