ELECTRICAL CONDUCTIVITY OF MAGNESIA INSULATION FOR HEAT-RESISTANT CABLES IN THE PRESENCE OF INTENSE RADIATION AND HIGH TEMPERATURE

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Emission detectors (ED) for neutron and γ radiation and small ionization chambers are widely used in order to control power distribution in reactors. A signal from the ED and the ionization chambers within the active zone and the biological shield is transmitted along cables which have a core and a casing consisting of magnesium oxide [1, 2]. When developing ED and chambers for a specific reactor, it is often necessary to know how the resistance of the cable insulation in the communication lines depends on temperature, irradiation intensity, and operational lifetime in the active zone for a choice of optimal operational regimes for the detectors and on estimates of the error in monitoring the power distribution. Let us examine the mechanism for electrical conductivity in irradiated and unirradiated mineral cable insulation in order to clarify the laws that govern such behavior.

The mechanism of electrical conductivity in magnesium oxide outside of radiation fields has been studied experimentally by the EMF method and by Turbandt's method [3, 4]. The electrical conductivity consists of bulk and surface components. The bulk electrical conductivity of magnesium oxide at a temperature 500-1400°C and high partial oxygen pressure (exceeding 10^{-5} Pa) consists of intrinsic and ionic conductivity, i.e., the charge is transferred by magnesium ions [5]. For polar crystals, including magnesium oxide, the following temperature dependence is characteristic for the bulk electrical conductivity [6]:

$$\sigma_V = A_1 \exp\left(-\frac{W_i}{kT}\right) + A_2 \exp\left(-\frac{W_i + 0.5W_d}{kT}\right), \tag{1}$$

where A_1 and A_2 are coefficients that depend weakly on temperature; W_i, activation energy required for drift of weakly bound atoms (ions) and vacancies; W_d, activation energy necessary for the formation of a defect.

At low temperatures, the contribution of the first term in formula (1) to the electrical conductivity is much greater than that of the second, and at high temperatures the opposite occurs. For this reason, the dependence of σ_V on T for high and low temperatures is exponential. This stems from the fact that the concentration of Frenkel' and Schottky defects in a crystalline lattice increases exponentially with temperature.

Thus, the temperature dependence of the resistance of cable insulation R_{ins} in the absence of irradiation can be represented in terms of the variables $\log R_{ins}$ and $1/T$. In this case, the graph of the function will contain straight-line segments.

The mechanism of electrical conductivity with irradiation was studied primarily theoretically for electrical conductivity stemming from γ radiation. It is assumed that the band model of a solid with traps distributed in the forbidden band is valid for mineral insulation, i.e., free and trapped charge carriers, electrons, exist [7]. The action of the radiation involves the transfer of electrons from the valance band into the conduction band in a quantity that is determined by the γ-quanta flux density. An electron can return into the valance band and recombine only via a trap. The temperature dependence of γ radiation-induced electrical conductivity arising from γ radiation is determined, on the one hand, by the fact that the probability for an electron to return from the energy level of a trap into the conduction band increases exponentially with temperature and the probability for recombination decreases correspondingly, while on the other hand, the temperature dependence of the electronic mobility is of the form $\mu \approx$ const $T^{3/2}$, obtained from Rutherford's formula for scattering of electrons.

Calculation of the electrical conductivity of mineral insulation in accordance with the considerations presented above gives for γ radiation-induced electrical conductivity the following expression [7]:

$$\sigma_r = P_\gamma G T^{3/2} \exp(-W/kT), \tag{2}$$

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where P_γ is the intensity of the γ-radiation dose; T, absolute temperature; G and W, coefficients that do not depend on temperature and radiation intensity.

In performing the calculation, it was tacitly assumed that the number of trapped electrons does not depend on the γ-radiation dose, from which it follows that the electrical conductivity is proportional to the irradiation intensity. Such an approach is simplified and does not permit explaining the experimentally observed dependence of σ_r on P_γ, which is of the form $\sigma_r \sim P_\gamma^\Delta$, where Δ differs from unity. In order to understand the physical mechanism of the phenomena leading to this difference and to establish limits on the variation of Δ, let us use some of the results of theoretical investigations of the phenomenon of photoconductivity [8, 9].

It is possible to change the number of trapped electrons depending on the irradiation intensity by using the concept of the Fermi level and the energy distribution of traps in the forbidden zone. The number of trapped electrons equals the number of traps located below the Fermi level [10]. Two types of distributions of traps with respect to energy are considered [9]: uniform and exponential, i.e., such that the concentration of traps having energy E decreases exponentially with increasing distance of E from the bottom of the conduction band. It turned out that the functional dependence of the number of free electrons n in the conduction band having the form $n \sim P_\gamma^{1/2}$, which is valid for substances that do not contain traps, changes into a dependence of the form $n \sim P_\gamma$ for a uniform distribution of traps. If the energy distribution of traps is an exponential of the form $n_E dE = A \exp(-E/kT_1)dE$ and the Fermi level is situated in the center of the region where the energy distribution of traps is exponential (the case most often encountered in practice), then a dependence of the form $n \sim P_\gamma^\Delta$, where $\Delta = T_1/(T + T_1)$ and $T < T_1$, is often valid [9]. Here, T is the absolute temperature of the insulation, while T_1 can be viewed as an equilibrium temperature corresponding to the given trap distribution, i.e., the temperature for which the traps are frozen when the substance cools after it crystallizes. When the energy distribution of traps is very sharply exponential, as T_1 approaches T, the parameter Δ approaches 0.5 [9]. The parameter Δ is a characteristic of the substance of which the insulation is made. However, theoretical calculation of the parameter Δ for MgO is at present difficult due to the absence of information on the energy distribution of traps in this substance.

Special conditions, apparently, exist in ED insulation. When the emitter is irradiated by neutrons, ED insulation is irradiated by fast electrons from the emitter, which, in slowing down, create a large number of secondary electrons with energy exceeding 1 eV. In this case, the expression $\Delta = T_1/(T + T_1)$, strictly speaking, is not applicable, since the injection of high-energy electrons into insulation was not taken into account in deriving it. However, this effect can be taken into account empirically by introducing into the expression for Δ some effective temperature T_{eff} instead of T. Injection of a large number of high-energy electrons into the insulation by secondary forces can, probably, strongly increase the effective temperature of the electron gas in the insulation, as a result of which it will turn out that $T_{eff} > T_1$. In this case, we obtain $\Delta < 0.5$, i.e., for ED the dependence of the insulation resistance on the irradiation intensity can turn out to be significantly weaker than for a cable. In this case, however, it should be noted that, as evident from the experimental results in [2], the radiation-induced electrical conductivity σ_r is greater for ED than for a cable, even though it depends weakly on the irradiation intensity.

The resistance of cable insulation and ED insulation depends relatively weakly on the operational lifetime in the active zone of the reactor in comparison with the dependence on temperature and irradiation intensity [2, 11]. The operational lifetime of cables in the active zone is conveniently expressed in terms of the fluence of fast neutrons F_f, while the operational lifetime of ED is conveniently expressed in terms of the fluence of thermal neutrons F_T.

Thus, the dependence of the resistance of cable insulation on temperature, irradiation intensity, and operational lifetime in the active zone can be sought in the form

$$R_r = G_1 P_\gamma^{-\Delta} T^{-3/2} \exp(W_1/kT + HF_f, \quad (3)$$

where G_1, W_1, Δ, and H are constants determined from the experimental data, and furthermore, it may be expected that the parameter Δ falls into the range 0.5-1. It is clear that the following four dimensional variables will be convenient for such an analysis: $1/T$, $\ln P_\gamma$, F_f, and $\ln(R_r T^{3/2})$.

A similar dependence is also obtained for ED, but the expected value of Δ in this case, as follows from what was said above, will be 0-0.5. In order to analyze the data on the resistance of ED insulation, it is convenient to use four dimensional variables of the type $1/T$, $\ln \varphi_T$, F_T, and $\ln(R_r T^{3/2})$.

TABLE 1. Confidence Intervals for the Coefficients of Empirical Formulas with a Confidence Probability of 0.98

Formula	No. of exptl. points analyzed	Common logarithm of the first coeff.	Δ	Coeff. multiplying 1/T	Coeff. multiplying $F_f \cdot 10^{-20}$
5	50	13,7±1,4	0,87±0,16	5200±2200	0,18±0,15
6	51	—2,6±1,7	—	21300±3500	—
7	51	17,6±0,2	1,0±0,05	—530±390	0,18±0,17
8	32	—1,1±1,2	—	16300±1700	—

The electrical conductivity of insulation with irradiation consists of thermal σ_t and radiation-thermal σ_r components, i.e., the resistance of the insulation at a given temperature and irradiation intensity is expressed as follows:

$$R_{\mathrm{ins}} = \left(\frac{1}{R_r} + \frac{1}{R_t}\right)^{-1}, \qquad (4)$$

where R_t is the resistance of the insulation for a given temperature and in the absence of radiation.

The general analytic expression of R_r for the cable is obtained by a statistical analysis of numerical information on the resistance of cable insulation with magnesia insulation, and in addition, we simultaneously analyze the data from our experiments [2, 11] on the IVV-2 water cooled—water moderated research reactor for cables produced in this country, as well as the results obtained by researchers in other countries [12, 13], for tests in a reactor and γ radiation from a source, for cables manufactured in the USA and in Canada.

All tests are obtained for coaxial cables with a single layer of insulation (external cable diameter varying from 1.0 to 6.3 mm, casing thickness varying from 0.2 to 0.5 mm, and core diameter varying from 0.2 to 0.7 mm). Most of the results are for a cable with an external diameter of 3.0 mm, and for this reason, the resistance of insulation in cables with other diameters was referred to a cable of this size.

Data from four different experiments were analyzed: in three of the experiments, the dependences of the resistance of cable insulation on temperature and irradiation intensity were obtained with independent variations in the latter, while in the fourth experiment the dependence on the operational lifetime of the cable in the active zone was obtained. These experimental results, apparently, reflect all of the published information (up to mid-1978) on the resistance of cable insulation with MgO insulation under intense irradiation and temperature varying from 20 to 750°C.

The experimental data were analyzed using two statistical methods: a multidimensional least-squares method (MMLS) and the method of grouping arguments (MGA) [8, 14]. For the analysis, the experimental results were represented in terms of the coordinates indicated above. The data corresponding to a temperature less than and above 450°C were analyzed separately, since at approximately this temperature there is transition from impurity electrical conductivity (at a temperature up to 450°C) in magnesium oxide to the intrinsic ionic electrical conductivity. It should be expected that with this separation in doing the analysis using the indicated variables, the multidimensional analytical expression obtained will be close to linear.

A distinguishing feature of MGA in comparison with MMLS is the possibility of separating out arguments according to the degree to which they effect the function. In carrying out the MGA analysis, it turned out that the function $\ln(R_r T^{3/2})$ is most strongly affected by $\ln P_\gamma$, less strongly by $1/T$, and most weakly by F_f. At a temperature 30-450 °C, the effect of the latter argument was generally not discernible in the range of irradiation intensities and neutron fluence investigated.

A general analytic expression for R_t for a cable is obtained by simultaneous statistical analysis in the variables $1/T$ and $\log R_t$ using the least-squares method on data from three experiments [2, 12, 13] for an unirradiated cable for temperatures below and above 450°C, separately.

The following general analytic expressions were obtained using MMLS for R_r and R_t:

for temperatures above 450°C

$$R_r = 3{,}6 \cdot 10^{13} T^{-3/2} (10^2 P_\gamma)^{-0{,}87} \exp(5200/T + 0{,}18 F_f 10^{-20}) \ln \frac{b}{a}, \tag{5}$$

$$R_t = 2{,}5 \cdot 10^{-3} \exp(21300/T) \ln \frac{b}{a}; \tag{6}$$

for temperatures up to 450°C

$$R_r = 4{,}2 \cdot 10^{17} T^{-3/2} (10^2 P_\gamma)^{-1{,}0} \exp(-530/T + 0{,}18 F_f 10^{-20}) \ln \frac{b}{a}; \tag{7}$$

$$R_t = 0.087 \exp(16300/T) \ln \frac{b}{a}, \tag{8}$$

where T is the absolute cable temperature, in °K; R_γ, dose of γ radiation, in g-R/sec; F_f, fluence of fast neutrons ($E > 1$ MeV), in neutrons/cm^2; a and b, inner and outer diameters of the cable insulation, in mm. R_r and R_t are referred to a 1-m length of cable and expressed in $\Omega \cdot$ m.

The limits of applicability of formulas (5) and (7) are determined according to each of the variable ranges investigated in the experiments, on the basis of which these formulas were obtained, i.e., for temperatures from 30 to 720°C, for γ-radiation dosage from 0.8 to $1 \cdot 10^5$ g-R/sec, and for fast neutron fluences from 0 to $9 \cdot 10^{20}$ neutron/cm^2.

We estimated the errors in the analytic expressions starting with a check of the normality of the distribution of deviations in the experimental values of $\log R_r$ from the values computed according to formulas (5) and (7) using the criterion [15]. It was shown that this distribution was close to normal (using a χ^2 criterion with a confidence probability $1 - p \approx 0.7$). A similar result was also obtained for $\log R_t$. It was verified that the experimental results on the resistance R_r, obtained in different experiments, can be viewed as belonging to a single general set of results. The results were checked using Bartlett's criterion with a confidence probability of 0.98.

We also checked the normality of the distribution of deviations in the experimental values of $(\log R_{ins})$ from the values of $(\log R_{ins})_{comp}$, obtained from formula (4) using formulas (5)-(8), and it was established that the distribution is close to normal (χ^2 confidence probability of 0.7). The mean-square deviation of $(\log R_{ins})_{exp}$ from $(\log R_{ins})_{comp}$ was computed. It corresponds to an increase or decrease in the resistance of the cable insulation by a factor of 2. It should be noted that a similar mean-square deviation obtained in analyzing data of one of the experiments [2] corresponds to a change in R_{ins} by a factor of 1.6.

In addition, we computed the mean-square errors in determining the coefficients in the formulas (5)-(8) as a result of errors in measuring the temperature, γ-radiation dose, fast neutron fluence, and insulation resistance. The calculation was done using a Monte Carlo method and consists of a repeated (100 times) calculation of these coefficients using the MMLS method, and in addition, in making the transition from one cycle of computations to another, the values of t, P_γ, F_f and R_r for the experimental points are changed randomly according to a normal distribution within the limits of error in the measurements. The mean-square deviation of the values of each coefficient in the 100 computations gives the error sought, which, as it turned out, is too small to correspond to the observed spread in experimental points for the analyzed data on the resistance of cable insulation.

In order to determine the errors in the coefficients corresponding to this spread, the Monte Carlo calculation was carried out in such a way that in passing from one cycle of computations to another the value of $\ln(R_r T^{3/2})$ was changed randomly according to a normal distribution, and in addition, the mean-square deviation, characterizing this change, equaled the mean-square deviation of the experimental values of $\ln(R_r T^{3/2})$ from the values obtained according to formulas (5) and (7). In this case, the spread in the values of each coefficient characterizes the error stemming mainly from systematic errors in each experiment due to the use of cables made by different manufacturers and from different batches, as a result of performing measurements in different reactors, different techniques in setting up the experiment, and errors of a subjective nature. The confidence intervals obtained by the latter of the methods indicated above, for a confidence probability of 0.98 for the coefficients in formulas (5)-(7), are presented in Table 1.

The values of the coefficients in the empirical formulas characterize, generally speaking, the properties of magnesium oxide as an insulator, functioning in the presence of radiation and at high temperatures. However, at the same time, the values of these coefficients and their errors are determined to a large extent by the technological level achieved in preparing magnesium oxide, in manufacturing and processing cables, as well the state of all of the techniques in setting up the experiment for studying electrical conductivity.

We analyzed data on the resistance of the insulation for an emission neutron detector with an emitter consisting of silver and magnesium oxide insulation in a similar manner. The results of three different experiments with ED manufactured in this country were analyzed: in two of the experiments, the dependence of the resistance of the ED insulation on temperature and irradiation intensity was analyzed, while in the third experiment, the dependence on the operational lifetime of the ED in the active zone was analyzed [2, 11]. The following analytic expressions were obtained for R_r and R_t for ED using MMLS:

for temperatures exceeding 450°C*

$$R_r = 3{,}8\cdot 10^6 T^{-3/2} (\varphi_T \cdot 10^{-13})^{-0{,}31} \exp(6100/T) \ln \frac{b}{a}; \tag{9}$$

$$R_t = 2{,}8 \exp(12600/T) \ln \frac{b}{a}; \tag{10}$$

for temperatures up to 450°C

$$R_r = 1{,}5\cdot 10^9 T^{-3/2} (\varphi_T 10^{-13})^{-0{,}50} \exp(2500/T + 0{,}024 F_T 10^{-20}) \ln \frac{b}{a}; \tag{11}$$

$$R_t = 2.4\cdot 10^3 \exp(7700/T) \ln \frac{b}{a}. \tag{12}$$

As can be seen from the results of the analysis, the experimental values of the parameter Δ for a cable and for ED correspond to the theoretically predicted values. It was shown that the deviations of the experimental values of $\log R_r$, $\log R_t$, and $\log R_{ins}$ from the values obtained from formulas (4) and (9)-(12) are distributed almost in a normal manner (confidence probability $1 - p$ using a χ^2 check not less than 0.5). The mean-square deviation of the experimental values of $\log R_{ins}$ from those obtained according to (4) using formulas (9)-(12) correspond to an increase or a decrease in R_{ins} for ED by a factor of 1.7. This value for ED is less than that for a cable, since the analysis involved data from tests in a single reactor and only ED manufactured in this country were used.

The limits of applicability of formulas (9) and (11) are: for temperature, from 80 to 800°C; for flux density of thermal neutrons, from $1.7\cdot 10^{12}$ to $8.5\cdot 10^{13}$ neutron/cm$^2\cdot$sec; for fluence of thermal neutrons [only for formula (11)], from 10^{18} to $1.2\cdot 10^{21}$ neutrons/cm^2.

The method described above for analyzing the experimental and computational results, roughly similar to the method for analyzing physical reactor experiments [16], permits, having obtained an analytic expression, representing all available experimental computational information on the quantity being examined in a clear and easily understood form. The relations obtained for cables and ED can be used for developing new designs for ED and ionization chambers.

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BLISTERING OF 0Kh16N15M3B STAINLESS STEEL UNDER SIMULTANEOUS IRRADIATION BY DEUTERIUM AND HELIUM IONS

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The basic laws governing radiation-induced blistering have by now been studied experimentally [1-4]. However, it should be noted that most of the investigations were carried out for a single type of ion, even though under real conditions in a nuclear reactor the first wall will be subjected to irradiation by ions of hydrogen and helium isotopes, simultaneously. It is already clear that the synergistic effect occurring with simultaneous action of different components of the plasma emission will effect blistering to a greater extent than other processes occurring on the surface of the wall. In particular, the first experiments on simultaneous implantation of D^+ and He^+ ions in nickel revealed a decrease in the critical dose that causes the formation of blisters [5].

We studied the radiation damage to the surface of 0Kh16N15M3B stainless steel with simultaneous and sequential irradiation by deuterium and helium ions with energies of 20 and 40 keV, respectively, for irradiation temperatures of 500 and 200°C. The specimens for the irradiation were cut out of foil obtained by repeated rolling with intermediate annealing at 850°C for 30 min. After the final rolling, the foil was annealed at 1050°C for 30 min. The size of a grain after working was 30-50 μm. Electrically polished specimens of austenitic 0Kh16N15M3B stainless steel were irradiated by He^+ and D^+ ions in an ILU-3 accelerator [6]. Five series of experiments were carried out in order to clarify the role of synergism:

1) irradiation by 20 keV D^+ ions;

2) irradiation by 40 keV He^+ ions;

3) simultaneous irradiation by 20 keV D^+ ions and 40 keV He^+ ions;

4) sequential irradiation by 20 keV D^+ ions and by 40 keV He^+ ions;

5) sequential irradiation first by He^+ ions and then by D^+ ions.

In all of the experiments, the irradiation dose was 10^{18} and $6 \cdot 10^{18}$ cm^{-2} for helium and deuterium ions, respectively. The rate at which the dose was accumulated was $5.4 \cdot 10^{14}$ for D^+ ions and $1.8 \cdot 10^{14}$ cm^{-2} sec^{-1} for He^+ ions. It should be noted that D^+ ions simultaneously irradiated three specimens from runs 1, 4, and 5, while He^+ ions irradiated two specimens from runs 2 and 4. Each of the runs was carried out for two values of the target temperature, 200 and 500°C, monitored by a thermocouple. As is well-known, bombardment of Fe–Cr–Ni steels and alloys with helium ions at $T = (0.1-0.4)\ T_{melt}$ leads to the development of flaking [4] and, for $T \approx (0.4-0.45)\ T_{melt}$, blistering is observed. A Stereoskan-180 scanning electron microscope was used to study the surfaces of irradiated targets.

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