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# INVESTIGATION OF HEAVY RADIATION-SHIELDING CONCRETE AFTER ACTIVATION BY FAST NEUTRONS AND GAMMA RADIATION

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*The induced activity of materials based on iron-barium-serpentinite cement stone and the developed iron-magnetite-serpentinite cement stone in the composition of radiation-shielding units of a high-power circuit-type reactor has been calculated, and radiation factors specified by the activation process have been analyzed.*

The problem of ionizing radiation shielding in a nuclear-power complex has attracted a good deal of attention [1–4]. Biological shielding of nuclear reactors is meant for reducing the power of doses of $\gamma$ radiation and neutrons to limiting permissible values.

Radiation shielding of the reactor is a very complex and expensive installation; therefore, the problem of designing simple, easily mountable, and inexpensive shielding is of great current interest. Such shielding is most frequently made of special heavy concrete that absorbs ionizing radiation [5, 6]. The water content in concrete should be no less than 1%, which improves the attenuation of neutron fluxes [2].

In the radiation-shielding units of high-power circuit-type reactors use is made of iron-barium-serpentinite cement stone (IBSCS). This material was manufactured from a mixture of barium-serpentinite cement and pig iron powder through tempering it with water [7]. The mixtures were produced at the Leningrad cement pilot plant of the Institute "Giprotsement." The production rate was no lower than 15 thousand tons per year, which fully met the demands of all nuclear power plants with high-power circuit-type reactors under construction. After the accident at the Chernobyl nuclear power plant, the construction of plants with such reactors was discontinued, which immediately led to the suspension of the production of IBSCS mixtures, and the pilot plant was closed. However, starting in 1996, at operating nuclear power plants the question has been raised as to resuming IBSCS production for overhauling the failed units. Investigations of this problem at the Federal State Unitary Enterprise "Dollezhal Scientific Research and Design Institute of Electrical Engineering" showed that resumption of IBSCS production is at present unfeasible. Therefore a decision was made to develop the technology and begin the manufacture of a new alternative material not inferior to IBSCS in its properties. On order of the Scientific Research and Design Institute of Electrical Engineering, a new alternative material — iron-magnetite-serpentinite cement stone (IMSCS) — was produced at the V. G. Shukhov Belgorod State Technological University.

The design was based on the principle of not exceeding the operative norms for external and internal irradiation of the personnel and population and on the content of harmful chemical substances in objects of the surrounding medium at normal operation, in the case of failures, and over the entire time of operation of high-power circuit-type reactors.

The manufacture of the mixtures, heavy radiation-shielding concretes, and articles made of them can be organized under the conditions of the production of reinforced concrete or on specialized industrial sites. Concrete produced from the IMSCS mixture is manufactured from a homogeneous mixture of plasticized portland cement, additives with chemically bound water (as serpentine and others), an activated iron ore filler, and iron shot. The developed concrete has the following characteristics:

| | |
|---|---|
| Volume mass of freshly laid concrete (State Standard 12730.1-78) | $4130 \pm 30$ kg/m$^3$ |
| Density of concrete dried at 110$^o$C (State Standard 12730.1-78) | $4000 \pm 20$ kg/m$^3$ |

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TABLE 1. Chemical Composition of Shielding Materials (wt. 5%)

| Material | Components | | | | | | | | | | Density, g/cm$^3$ |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | $SiO_2$ | $Al_2O_3$ | $Fe_2O_3$ | CaO | MgO | $R_2O^*$ | BaO | $SO_3$ | $H_2O$ | Fe | |
| IMSCS | 3.26 | 0.66 | 13.73 | 9.96 | 0.31 | 0.16 | — | 0.28 | 1.00 | 70.64 | 3.9 |
| IBSCS | 3.85 | 0.38 | 1.08 | 1.81 | 2.3 | — | 28.64 | 1.24 | 0.7 | 60 | 3.8 |

*$R_2O = K_2O + Na_2O$, supposedly in the same weight proportion.

TABLE 2. Nuclide Composition of Materials (wt. %)

| Nuclide | Material | | Nuclide | Material | |
|---|---|---|---|---|---|
| | IMSCS | IBSCS | | IMSCS | IBSCS |
| H | $1.11 \cdot 10^{-3}$ | $1.92 \cdot 10^{-3}$ | Ca | $7.12 \cdot 10^{-2}$ | $5.80 \cdot 10^{-3}$ |
| O | $1.02 \cdot 10^{-1}$ | $8.91 \cdot 10^{-2}$ | Cr | $2.12 \cdot 10^{-3}$ | $1.82 \cdot 10^{-3}$ |
| Na | $6.32 \cdot 10^{-4}$ | — | Mn | $4.59 \cdot 10^{-3}$ | $3.95 \cdot 10^{-3}$ |
| Mg | $1.87 \cdot 10^{-3}$ | $1.39 \cdot 10^{-2}$ | Fe | $7.88 \cdot 10^{-1}$ | $5.96 \cdot 10^{-1}$ |
| Al | $3.50 \cdot 10^{-3}$ | $2.02 \cdot 10^{-3}$ | Co | $1.34 \cdot 10^{-4}$ | $1.03 \cdot 10^{-4}$ |
| Si | $1.63 \cdot 10^{-2}$ | $1.79 \cdot 10^{-2}$ | Ni | $2.12 \cdot 10^{-3}$ | $1.82 \cdot 10^{-3}$ |
| S | $1.47 \cdot 10^{-3}$ | $4.98 \cdot 10^{-3}$ | Cu | $2.12 \cdot 10^{-3}$ | $1.80 \cdot 10^{-3}$ |
| K | $7.06 \cdot 10^{-4}$ | — | Ba | — | $2.56 \cdot 10^{-1}$ |

| | |
|---|---|
| Ultimate bending strength (State Standard 310.4-81) | $7.7 \pm 0.3$ MPa |
| Ultimate compression strength after hardening for 28 days | $45 \pm 5$ MPa |
| Compression strength after drying at 110°C | $47 \pm 5$ MPa |
| Compression strength after thermal treatment at 300°C | $30 \pm 4$ MPa |
| Compression strength class (State Standard 26633-91) | V 35 |
| Quantity of water in concrete after hardening for 28 days | $2.5 \pm 0.25$ wt. % |
| Quantity of water in concrete after drying at 110°C | $2.2 \pm 0.2$ mass % |
| Quantity of chemically bound water at 300°C | $1.00 \pm 0.1$ mass % |
| Coefficient of linear expansion at 300°C | $8 \cdot 10^{-6}$ m·deg$^{-1}$ |
| Thermal conductivity at 300°C | 3.85 W/(m·K) |
| Operation temperature | up to 300°C |
| Combustibility group (State Standard 30244-94) | NG (noncombustible) |

Experimental investigations of the radiation-shielding properties of the material IMSCS conducted at the Dollezhal Scientific Research and Design Institute of Electrical Engineering showed that for $\gamma$ radiation sources $^{137}$Cs and $^{60}$Co the relaxation length of the power of the dose of $\gamma$ radiation is $4.37 \pm 0.1$ and $5.7 \pm 0.1$ cm, respectively, and for fast neutrons it is $10.6 \pm 0.1$ cm; at $\gamma$ radiation energies of 661 and 1332 keV the total attenuation coefficient is, respectively, 0.347 and 0.240 cm$^{-1}$, i.e., the developed material IMSCS is superior to the previously used material IBSCS in its radiation shielding characteristics.

Activation is calculated in two steps:

1. The space-energy distribution of neutrons in the considered material is determined. To this end, the ANISN program [8] is used, which implements the solution of a unidimensional transport equation by the method of discrete ordinates with account for anisotropic scattering. The neutron spectrum is calculated for a 12-group splitting of the energy interval.

2. The specific activity of radionuclides after irradiation of the considered material is calculated. For this, the SAM program [9] is used, which calculates the induced activity of the material and its radionuclide composition as a function of the spectrum of activating neutrons, irradiation mode, and holding time.

The chemical composition of the materials IMSCS and IBSCS after heating at 300°C is presented in Table 1. In calculating the nuclear concentrations of nuclides in the shielding materials, as the "Fe" component representing a metallic filler, consideration was given not to pure iron, which is almost impracticable, but instead to steel of the St3

TABLE 3. Nuclear Reactions of Nuclides in the Materials IMSCS and IBSCS

| Nuclide | Reaction | Reaction products | Nuclide | Reaction | Reaction products |
|---|---|---|---|---|---|
| $^{23}$Na | (n, γ) | $^{24}$Na | $^{59}$Co | (n, p) | $^{59}$Fe |
| $^{24}$Mg | (n, p) | $^{24}$Na | $^{59}$Co | (n, α) | $^{56}$Mn |
| $^{27}$Al | (n, γ) | $^{28}$Al | $^{59}$Co | (n, 2n) | $^{58}$Co |
| $^{27}$Al | (n, α) | $^{24}$Na | $^{58}$Ni | (n, γ) | $^{59}$Ni$^{*}$ |
| $^{28}$Si | (n, ð) | $^{28}$Al | $^{58}$Ni | (n, p) | $^{58}$Co |
| $^{32}$S | (n, ð) | $^{32}$P$^{*}$ | $^{58}$Ni | (n,α ) | $^{57}$Fe$^{*}$ |
| $^{33}$S | (n, p) | $^{33}$P$^{*}$ | $^{58}$Ni | (n, 2n) | $^{57}$Ni → $^{57}$Co |
| $^{34}$S | (n, γ) | $^{35}$S$^{*}$ | $^{58}$Ni | (n, d) | $^{57}$Co |
| $^{50}$Cr | (n, γ) | $^{51}$Cr | $^{60}$Ni | (n, p) | $^{60}$Co |
| $^{52}$Cr | (n, 2n) | $^{51}$Cr | $^{60}$Ni | (n, 2n) | $^{59}$Ni$^{*}$ |
| $^{55}$Mn | (n, γ) | $^{56}$Mn | $^{62}$Ni | (n, γ) | $^{63}$Ni$^{*}$ |
| $^{55}$Mn | (n, 2n) | $^{54}$Mn | $^{62}$Ni | (n, α) | $^{59}$Fe |
| $^{54}$Fe | (n, γ) | $^{55}$Fe$^{*}$ | $^{64}$Ni | (n, 2n) | $^{63}$Ni* |
| $^{54}$Fe | (n, p) | $^{54}$Mn | $^{63}$Cu | (n, γ) | $^{64}$Cu |
| $^{54}$Fe | (n, α) | $^{51}$Cr | $^{63}$Cu | (n, p) | $^{63}$Ni$^{*}$ |
| $^{56}$Fe | (n, p) | $^{56}$Mn | $^{63}$Cu | (n, α) | $^{60}$Co |
| $^{56}$Fe | (n, 2n) | $^{55}$Fe$^{*}$ | $^{63}$Cu | (n, α) | $^{60m}$Co → $^{60}$Co |
| $^{58}$Fe | (n, γ) | $^{59}$Fe | $^{65}$Cu | (n, 2n) | $^{64}$Cu |
| $^{59}$Co | (n, γ) | $^{60}$Co | | | |

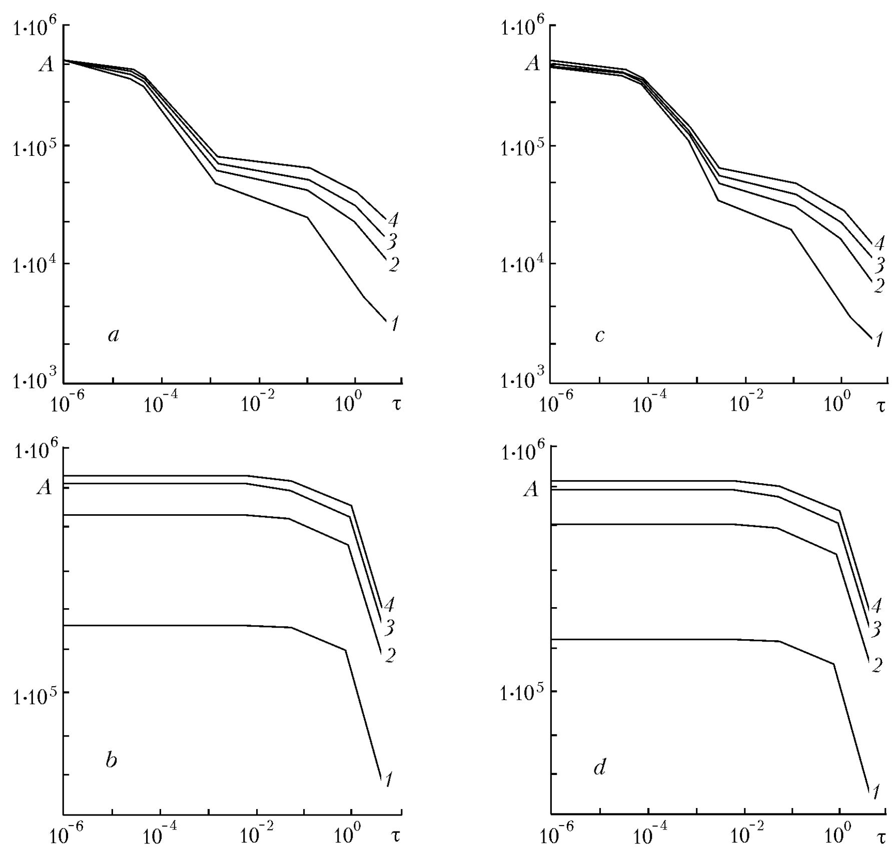


Fig. 1. Activity $A$ of γ radiation (a, b) and β radiation (c, d) of units made of IMSCS (a, b) and IBSCS (c, d) vs. the holding time τ for the following durations of irradiation: 1) 1 year, 2) 5, 3) 10, and 4) 30 years. $A$, Bq; τ, year.

TABLE 4. Average Values of the Power of the Dose on the Side of the Unit, μZv/h

| Material of the unit | Distance from the units, cm/h | | |
|---|---|---|---|
| | 1 | 10 | 100 |
| IMSCS | 1.3 | $6.4 \cdot 10^{-1}$ | $3.8 \cdot 10^{-2}$ |
| IBSCS | 1.2 | $6.1 \cdot 10^{-1}$ | $3.6 \cdot 10^{-2}$ |

TABLE 5. Comparison of the Specific Activity $A_{sp}$ of Materials with the Minimum Significant Specific Activity (the Norms NRB-99) (the irradiation time is 30 years and the holding time is 0)

| Nuclide | Minimum significant specific activity, Bq/year | $A_{sp}$, Bq/year | | $A_{sp}$, Bq/year | |
|---|---|---|---|---|---|
| | | IMSCS | IBSCS | IMSCS | IMSCS |
| $^{24}$Na | $1.00 \cdot 10^{1}$ | $3.25 \cdot 10^{-2}$ | $4.62 \cdot 10^{-4}$ | $3.25 \cdot 10^{-3}$ | $4.62 \cdot 10^{-5}$ |
| $^{28}$Al | — | $6.52 \cdot 10^{-2}$ | $5.02 \cdot 10^{-2}$ | 0 | 0 |
| $^{27}$Mg | — | $1.85 \cdot 10^{-4}$ | $1.17 \cdot 10^{-4}$ | 0 | 0 |
| $^{32}$P* | $1 \cdot 10^{3}$ | $8.62 \cdot 10^{-4}$ | $3.07 \cdot 10^{-3}$ | $8.62 \cdot 10^{-7}$ | $3.07 \cdot 10^{-6}$ |
| $^{33}$P* | $1 \cdot 10^{5}$ | $1.44 \cdot 10^{-5}$ | $5.46 \cdot 10^{-5}$ | $1.44 \cdot 10^{-10}$ | $5.46 \cdot 10^{-10}$ |
| $^{35}$S* | $1 \cdot 10^{5}$ | $2.23 \cdot 10^{-3}$ | $9.61 \cdot 10^{-3}$ | $2.23 \cdot 10^{-8}$ | $9.61 \cdot 10^{-8}$ |
| $^{51}$Cr | $1 \cdot 10^{3}$ | $6.37 \cdot 10^{-2}$ | $7.23 \cdot 10^{-2}$ | $6.37 \cdot 10^{-5}$ | $7.23 \cdot 10^{-5}$ |
| $^{56}$Mn | $1 \cdot 10^{1}$ | 2.53 | 2.82 | $2.53 \cdot 10^{-1}$ | $2.82 \cdot 10^{-1}$ |
| $^{54}$Mn | $1 \cdot 10^{1}$ | $1.94 \cdot 10^{-2}$ | $1.56 \cdot 10^{-2}$ | $1.94 \cdot 10^{-3}$ | $1.56 \cdot 10^{-3}$ |
| $^{55}$Fe* | $1 \cdot 10^{4}$ | 5.28 | 5.18 | $5.28 \cdot 10^{-4}$ | $5.18 \cdot 10^{-4}$ |
| $^{59}$Fe | $1 \cdot 10^{1}$ | $1.05 \cdot 10^{-1}$ | $1.03 \cdot 10^{-1}$ | $1.05 \cdot 10^{-2}$ | $1.03 \cdot 10^{-2}$ |
| $^{60}$Co | $1 \cdot 10^{1}$ | 1.94·10–1 | $1.91 \cdot 10^{-1}$ | $1.94 \cdot 10^{-2}$ | $1.91 \cdot 10^{-2}$ |
| $^{58}$Co | $1 \cdot 10^{1}$ | $8.00 \cdot 10^{-4}$ | $7.30 \cdot 10^{-4}$ | $8.00 \cdot 10^{-5}$ | $7.30 \cdot 10^{-5}$ |
| $^{59}$Ni* | $1 \cdot 10^{4}$ | $7.45 \cdot 10^{-5}$ | $8.28 \cdot 10^{-5}$ | $7.45 \cdot 10^{-9}$ | $8.28 \cdot 10^{-9}$ |
| $^{57}$Co | $1 \cdot 10^{2}$ | $3.09 \cdot 10^{-6}$ | $2.98 \cdot 10^{-6}$ | $3.09 \cdot 10^{-8}$ | $2.98 \cdot 10^{-8}$ |
| $^{63}$Ni* | $1 \cdot 10^{5}$ | $7.18 \cdot 10^{-3}$ | $8.0 \cdot 10^{-3}$ | $7.18 \cdot 10^{-8}$ | $8.07 \cdot 10^{-8}$ |
| $^{64}$Cu | $1 \cdot 10^{2}$ | $2.17 \cdot 10^{-1}$ | $2.43 \cdot 10^{-1}$ | $2.17 \cdot 10^{-3}$ | $2.43 \cdot 10^{-3}$ |
| All nuclides | | 8.52 | 8.70 | $2.91 \cdot 10^{-1}$ | $3.16 \cdot 10^{-1}$ |

type as a widely used grade of low-alloy steel. Steel of the St3 type comprises 98% Fe, 0.3% Cr, 0.65% Mn, 0.3% Ni, 0.17% Si, 0.05% S, and 0.3% Cu, which allows a variation of the number of activated nuclides. Along with this, a possible content of cobalt in the considered material is taken into account proceeding from the fact that it occurs as additives in iron (0.015%), chromium (0.04%), and nickel (0.7%). Table 2 supplies weight concentrations of nuclides in the shielding materials calculated from the data in Table 1 (with account for the above reasoning).

The space-energy distributions of neutron fluxes in the shielding materials were obtained using compositions consisting of the reactor core, structural elements of the reactor and reflector, the bed of the charge of serpentinite chips, and the bed of the considered material with a thickness of 60 cm. The absolute normalization of neutron fluxes is based on experimental values of the fluxes at the level of the lower surface of radiation shielding units of the high-power circuit-type reactor: $2 \cdot 10^{3}$ cm$^{-2} \cdot$sec$^{-1}$ for neutrons with energy over 1 MeV and $5 \cdot 10^{4}$ cm$^{-2} \cdot$sec$^{-1}$ for slow neutrons.

Activation is calculated for four values of the irradiation time: 1 year and 5, 10, and 30 years, and for eight values of the holding time after irradiation: 0 h (the instant of the cessation of irradiation), 0.5 h, 1 h, 6 h, 1 day, 1 month, 1 year, and 5 years. The main nuclear reactions of nuclides, which are taken into account in the activation calculations by the SAM program (for the selected irradiation modes and values of the holding time), are presented in Table 3 (in Table 3 and thereafter, the symbol "*" denotes radionuclides which are actually only β radiators). Results calculated using the SAM program are given in Fig. 1.

It should be noted that the material activation in the lower part of the unit is much higher than in its upper part, the difference being larger than two orders of magnitude. The specific activity averaged over the volume of the unit is about 20 times smaller than the maximum value in the lower part of the unit. Estimates of the levels of γ radiation from a just removed unit (without holding) presented in Table 4 give values of the power of the dose near it and on the side of its lateral surface. The maximum values of the power of the dose on the side of the lower part of the unit will be larger by approximately an order of magnitude.

The results obtained show that γ- and β-ray activity of the materials as a function of the holding time after irradiation ($T_{1/2}$ is the half-life period) is mainly determined by the following radionuclides:

a) in the first 24 h period — $^{56}$Mn ($T_{1/2}$ = 2.6 h), $^{64}$Cu ($T_{1/2}$ = 12.7 h), $^{51}$Cr ($T_{1/2}$ = 27.7 days), $^{54}$Mn ($T_{1/2}$ = 312.5 days), $^{55}$Fe ($T_{1/2}$ = 2.7 years), $^{59}$Fe ($T_{1/2}$ = 44.5 days), and $^{60}$Co ($T_{1/2}$ = 5.27 years);

b) after the lapse of a 24 h period during first months — $^{51}$Cr, $^{55}$Fe, $^{54}$Mn, $^{59}$Fe, and $^{60}$Co;

c) after the lapse of 1 year — $^{54}$Mn, $^{55}$Fe, and $^{60}$Co;

d) after the lapse of 5 years — $^{55}$Fe and $^{60}$Co.

If as the "Fe" component in the materials use is made of steel or pig iron, in which alloying additives are absent or contained in much smaller quantities that in steel of the St3 grade adopted in the calculations, γ- and β-ray activity of the materials will be determined by radionuclides $^{55}$Fe, $^{59}$Fe, and $^{60}$Co (with $^{59}$Fe contributing only in the first months). The power of the dose of γ radiation from a just removed will in this case be approximately 5 times lower than the values supplied in Table 4.

The degree of radioactivity of the irradiated materials can be evaluated by comparing their specific activity $A_{sp}$ with values of the minimum significant specific activity specified by the radiation safety norms NRB-99. Table 5 presents values of the minimum significant specific activity and the specific activity of the materials averaged over the volume of the unit for individual radionuclides. Clearly, as based on individual nuclides the values of $A_{sp}$ of the materials are mostly much smaller than the corresponding values of the minimum significant specific activity. According to the norms NRB-99, if only several nuclides are present, the sum of ratios of the activities of their tabular values should not be larger than unity. This requirement of the norm NRB-99 is fulfilled. Thus, for the unit as a whole the value of the average specific activity of the materials is smaller than the criterion of the minimum significant specific activity.

## CONCLUSIONS

1. The induced activity of the materials IMSCS and IBSCS in the composition of radiation shielding units of a high-power circuit-type reactor has been calculated and radiation factors specified by the activation process have been analyzed.

2. The maximum value of the induced activity of the unit after 30 years of irradiation is ~$1.2 \cdot 10^6$ Bq immediately on its removal and decreases to $2.3 \cdot 10^5$ after 5 years of holding.

3. For the considered irradiation and holding modes, the main contribution to the induced activity of the materials is made by activation of iron and a cobalt additive contained in it. With a holding time of up to 1 year, a noticeable contribution is made by additives such as manganese, chromium, nickel, and copper, which can be presented in a metallic filler.

4. Immediately on removal of the unit irradiated for 30 years, the average power of the dose of γ radiation around it is lower than 12 μZv/h at the surface; at a distance of 1 m from the unit the power of the dose is not higher than 0.5 μZv/h in any direction.

5. For the unit as a whole, the value of the average specific activity of the considered materials based on both individual radionuclides and their totality is smaller than the criterion of minimum significant specific activity specified by the norms NRB-99.

6. The material IMSCS is identical to the material IBSCS in the degree of activation by neutrons and can successfully supercede it in the manufacture of radiation-shielding units of high-power circuit-type reactors.

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