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CHANGE OF QUALITY OF A RADIOACTIVE WASTE UNDER LONG STORE TERMS

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By calculation methods, the dose rate of the radioactive waste, behind concrete protection, was evaluated in current work. Parameters, which were taken in account in the calculations, are geometry of the protection shell, size of the source and its isotopic composition. As model geometrical parameters the spent fuel assembly’s size and thickness of the concrete wall of the ventilated storage container (VSC)-VVER were taken. The computer program that does numerical calculation was composed in the Wolfram Alpha environment. The program takes into account change of the isotopic composition and spectra of gamma-radiation with time. Calculation results were compared to the known data on the spent nuclear fuel heat dissipation. Approach described in this work can be used for fast estimation of change in the quality of radioactive waste (RAW) in the long-term storage without recycling, for different initial isotopic composition. Obtained results were analyzed on the matter of change in gamma-radiation of RAW.

KEYWORDS: radioactive waste, dose rate, long terms storage, concrete protection, isotopic composition

Production and accumulation of the radioactive waste (RAW) are one of the major problems associated with the use of nuclear energy in any of its forms. According to the existing data, there is about 300 thousand tons of accumulated spent nuclear fuel (SNF) with total activity of ~10^{20}Bk, and by year 2030, this quantity is predicted to be 500 thousand tons. In particular, importance of this problem is reflected by the fact, that in the last years several monographs [1-5] on the problems of dealing with RAW were published.

Leaving aside the cosmic origin and associated with uranium mining radionuclides, in this paper we will consider 2 forms of RAW: 1) contained in SNF; 2) the so-called operational waste of nuclear plant. According to the established terminology, in countries, where radioactive or other recycling is not intended (for example - Ukraine), it belongs to the high-level RAW (HLW).
Currently, from 32 countries, oriented at nuclear power, 11 are developing radiochemical recycling, 6 - designing geological burial of SNF and HLW, and the rest (including Ukraine) implementing a temporary solution, consisting in the organization of temporary (30-100 years) safe storage of radioactive waste (spent fuel) without processing [3].

The composition of the radionuclides SNF studied in sufficiently detailed and is found in most nuclear databases. The composition of the radionuclides of the operational RAW is determined by the set of measures used on the nuclear plant for isolating, quite widely varies with time and treatment technology (recycling) [3].

Due to the currently accepted procedure of transformation of physical and chemical forms, and the isotopic composition of RAW, of particular interest is to trace the properties of the RAW(SNF) and other - solid radioactive waste (SRW) and liquid radioactive waste (LRW), for long-term storage without further processing.

The purpose of this work - to track changes in the quality of stored RAW for the first 30-50 years, and on this basis assess the possibility of improving the technology of handling RAW. Of particular interest in this case is the cooling interval 3-10 years, when overload, certification, and sorting operations are being done and there is possibility of operations with open SNF.

**PROPERTIES OF THE RADIOACTIVE WASTE QUALITIES**

Following the recommendations of [6] by a quality of waste (spent nuclear fuel) we mean the following criteria for storage and disposal: the total activity, radionuclide composition and specific activity, the equivalent dose rate, the energy release, the content of nuclear (fissile) materials, other physical and chemical parameters. For the standard SNF these parameters are presented on Fig. 1,2.

**Energy release**

Energy release of spent fuel was investigated from the very first years of operation of the reactors.

Way and Wigner (1946) published a formula for calculating the energy release up to the $10^5$ seconds [7], which was used until the 70s. In 1963 B.G. Pologikh [8] proposed to modify the formula and extend its validity up to $10^7$ seconds (3 years). McNeir [9] published the most accurate experimental results of residual heat release and offered their approximation as integral and series with tabulated coefficients.

Modern data on heat generation of WWER SNF [10] are consistent with other known settlements within ±10% (with the oldest (+28/-19)%) and made valid until the times of ~$10^8$ seconds.

![Energy release of the SNF during long term storage without recycling][1]

![Index of danger or the relative toxicity of the radioactive products of SNF (quantity in m³ of water to bring it to the allowable concentration)][2]

**Total activity and dose rate**

In this article, the total activity of the source does not affect the result, let's consider the initial time it is equal to 1. Since in RAW there are several different isotopes, the total activity is the sum of activities.

\[
A(t) = \sum_{i=1}^{n} A_i(t)
\]

Hereinafter, $i$ - denotes the number of isotopes in the mixture.

Since activity of the individual isotope decays exponentially, the total activity decreases as the sum of exponents.
The calculation uses the percentage value of the contribution to the total activity of the isotope source at a time

$$a_j = \frac{A_j(t)}{A(t)}$$  \hspace{1cm} (2)

Renormalizing condition according to (1)

$$\sum_j a_j = 1.$$  \hspace{1cm} (3)

Data on the total activity of SNF and its behavior were investigated and presented in [12]. The dose rate can be calculated based on the isotopic composition.

**CALCULATION OF GAMMA-CONSTANT**

**Rationale for a common approach**

When working with the real sources of ionizing radiation such as SNF four factors should be accounted for: 1) the geometry of the problem, 2) self-absorption in the source, 3) the effect of shielding, 4) the isotopic composition of the emitter. Since the isotopic composition of the radionuclide mix changes over time, factors 2 and 3 does not remain constant. In this context, it is convenient to use, as a characteristic of the source, a generalized gamma constant (GGC) [13,14], which takes into account the self-absorption in the volume source and the possible presence of protection and is numerically equal to the ratio of gamma-radiation at the observation point to the activity of the source. Generalization means that in calculations several factors are accounted for such as different radioisotopes in the sources, each with its differential gamma constant and account self-absorption in the body of the source and possible protective screen, absorption of which differernt for each energy of radiation.

As a model for the calculation of the source fragment of the standard VVER-1000 has been selected, presented in the form of a cylinder with diameter of 250 mm and height of 2.5 meters with the closest to the real filling, consisting of 322 kg of uranium dioxide, 109 kg of zirconium and 14 kg of steel, located in the air. The disposition of fuel and concrete protection is shown on the Fig. 3.

Radionuclide composition of the SNF was taken according to 2000 y. [15] data and is given in Table 1 below. There are also differential values of gamma constant of the radionuclides components.

The composition of fission products from Table 1 after 3 years of aging was selected for calculation. For smaller aging time the SNF data was not considered because the problems, discussed in this article, related to the quality of the radiation protection of the concrete, where the fuel is stored after 3 years of aging. Another reason is the intense radiation of short-lived isotopes in fresh spent fuel, making it difficult to measure its composition.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Activity %</th>
<th>$\Gamma$, aGy×m²/(s×Bq)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{90}$Sr</td>
<td>19,20</td>
<td>7,85E-07</td>
</tr>
<tr>
<td>$^{106}$Ru</td>
<td>16,95</td>
<td>7,48E+00</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>7,25</td>
<td>5,76E+01</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>26,29</td>
<td>2,11E+01</td>
</tr>
<tr>
<td>$^{144}$Ce</td>
<td>18,55</td>
<td>5,28E-01</td>
</tr>
<tr>
<td>$^{147}$Pm</td>
<td>9,65</td>
<td>1,06E-04</td>
</tr>
<tr>
<td>$^{85}$Kr</td>
<td>0,77</td>
<td>8,43E-02</td>
</tr>
<tr>
<td>$^{154}$Eu</td>
<td>0,74</td>
<td>4,34E+01</td>
</tr>
<tr>
<td>$^{155}$Eu</td>
<td>0,61</td>
<td>1,57E+00</td>
</tr>
</tbody>
</table>

The attenuation coefficient for concrete protection can be taken from known tables. And the attenuation coefficient due to self-absorption for the spent fuel assembly (SFA), consisting of several components with total amount of (uranium – 450 kg, 55 kg oxygen and zirconium 200 kg). For calculations instead of zirconium the data for attenuation coefficient were taken for the nearest element with known tables of absorption coefficients, in this case molybdenum. It was assumed that these elements are distributed uniformly over the volume. Accordingly, knowing the density of each element and the mass attenuation coefficient [13] it is possible to calculate the total linear attenuation coefficient.

**Method of calculation of the GGC**

The dose rate of gamma radiation was calculated by the standard method [13] at the point opposite the mid-cylinder power at a distance of 1 m from its axis, also with placing concrete wall with thickness of 30 or 60 cm between
the source and the observation point. Gamma constant of the j-th isotope containing several gamma lines can be calculated as follows:

$$\Gamma_j = \sum_i n_{ij} \Gamma(E_{ij}).$$  \hspace{1cm} (4)$$

where $$\Gamma(E_{ij})$$ - gamma constant for a certain line of energy $$n_{ij}$$ - quantum yield of photons of a given energy. Here, j - the index of the isotope in the mixture, i - the index of the gamma line.

Total dose rate of a mixture of isotopes can be expressed as:

$$\dot{G} = \sum_j \dot{G}_j = \sum_j \frac{\Gamma_j A_j}{\Delta_j},$$ \hspace{1cm} (5)

where j is the index of isotope, r is the distance from the source to the observation point.

Generalized gamma constant - the ratio of gamma radiation and the activity of A at a distance r, which can be written as follows:

$$\Gamma = \frac{\dot{G} r^2}{A}.$$ \hspace{1cm} (6)

Substituting in (6) the expression (5) yields:

$$\Gamma = \sum_j \frac{A_j}{A} \Gamma_j.$$

This expression was used by us to calculate the GGC for ideal point source.

For the volume cylindrical source dose rate calculated according to [13]:

$$\dot{G}_{ji} = 2 \frac{A_j}{V} \Gamma_j R G_{ji} \left( \frac{b}{R}, \frac{b}{R}, \mu_1(E_{ij}) R, d \mu(E_{ij}) \right).$$ \hspace{1cm} (8)

here h is the height of the cylinder, b is the distance from its axis to the observation point, R is the radius of cylindrical source, and d is the thickness of the protection screen.

Reduction factors for self-absorbing cylinder ($$\mu_1$$) and protection ($$\mu$$) depends on the energy of photons, therefore, the dose must be calculated separately for each gamma line. The total dose rate - the sum of the partial dose powers.

For radiation of one isotope, we have:

$$\dot{G}_j = \sum_i \dot{G}_{ji} = \sum_i 2 \frac{A_j}{V} \Gamma_j R G_{ji} \left( \frac{b}{R}, \frac{b}{R}, \mu_1(E_{ij}) R, d \mu(E_{ij}) \right).$$ \hspace{1cm} (9)

And GGC of the self-absorbing cylinder with one emitting isotope with a protective screen we get:

$$\Gamma_j = \frac{\dot{G}_j r^2}{A_j} = \sum_i 2 \frac{A_j}{\Delta_j} \Gamma_j \frac{r^2}{V} G_{ji} = \sum_i 2 n_{ij} \Gamma_j \frac{r^2}{V} G_{ji}. \hspace{1cm} (10)$$

As you can see, this value does not vary with time as it does not contain time dependent arguments. For GGC isotope mixture, the total activity A can be obtained:

$$\dot{G} = \sum_j \dot{G}_j = \sum_j 2 \frac{A_j}{A} \Gamma_j R G_{ji},$$ \hspace{1cm} (11)

$$\Gamma = \sum_j \frac{A_j}{A} \Gamma_j \frac{r^2}{V} G_{ji}.$$ \hspace{1cm} (12)

In this expression activity and gamma constant of individual elements are multiplied, making it impossible to separate them into two separate sum (sum of the activities and the amount of gamma constant). In this case, the value of the GGC is time dependent because for different isotopes half-life is different, thus the percentage $$\frac{A_j}{A}$$ contribution of the j-th mixture isotope will vary.

Expression (12) was used to calculate the generalized gamma constant (GGC) cylindrical self-absorbing source with protection.

The results of calculations of external radiation SNF

Authors have developed a unique computer program, which calculated gamma constant according to the eq. (12). The decay of the SNF isotopes was also taken inti account as a change to the energy distribution of the gamma spectrum.

Fig. 4,5 show the calculated dependence of the GGC versus time for an ideal point source and a source with self-absorption accordingly. Accounting for self-absorption in the source characterizes the heat release caused by gamma radiation of the SFA. Obtained GGC is consistent with the data from [11], if the activity is calculated for the data of the Table 1. Also, from this graph, you can conclude that the dose rate of the ideal source decreases slower than activity with time the first five years, and then decline is at the same speed. This is due to the rapid depletion of the isotopic composition of gamma emitters in the first 3-5 years of age and accompanied by an increase of GGC. Values of the GGC curve on Fig. 5 about an order of magnitude smaller than on Fig. 4 due to self-absorption power. Also, this graph proves the rule, which says that the total activity of the SNF mixture the first 30 years is decreasing faster than the decay heat.
The result of the calculation model of the GGC source with self-absorption and the flat protection of concrete 30 cm and 60 cm is shown on Fig. 6, 7. Having a shield leads to a weakening of the flow of low-energy photons, which are dominating in spectrum at the beginning of decay and after 30-40 years of aging. The observed peak is formed mainly from emissions of the $^{134,137}$Cs. The first five years of constant GGC is growing rapidly, due to the decay of the low-energy sources, and then falls down as exponent. From this we can conclude that the weakening multiplicity of gamma-radiation of protective storage container increases with time. At the same time, Fig. 7 reflects the fact that the dose rate of external radiation of SNF decays faster than its activity. For convenience the calculation results for the GGC are presented in the Table 2.

![Fig. 4. Point source’s GGC without protection](image1)

![Fig. 5. Self-absorbing cylindrical source’s GGC without protection](image2)

![Fig. 6. GGC for source with protection 30 cm](image3)

![Fig. 7. GGC for source with protection 60 cm](image4)

<table>
<thead>
<tr>
<th>SNF age, yrs</th>
<th>Point source</th>
<th>Cylindrical source without protection</th>
<th>Cylindrical source with flat protection wall d=30 cm</th>
<th>Cylindrical source with flat protection wall d=60 cm</th>
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<tr>
<td>0.5</td>
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<td>4.3915E-03</td>
<td>4.9752E-05</td>
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<td>1</td>
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<td>3.1912E+00</td>
<td>8.6897E-03</td>
<td>3.1683E-05</td>
</tr>
</tbody>
</table>

Table 2.
CONCLUSION

To produce the best technology of radioactive waste (SNF) management, accurate and reliable information on the properties of the SNF during prolonged storage is needed. Accumulated to date, research results and authors’ assessments allows to establish some laws relating to the physical properties of the RAW (SNF) for long-term storage without recycling. Approach of the GGC proposed by the authors to assess the quality of RAW (SNF) proved effective and showed characteristic changes in the quality of spent nuclear fuel. It is, first of all, the backlog in decrease of activity from the decrease of the energy release and dose power of radiation, then the establishment of the equilibrium spectrum and exponential decay of dose power of γ-rays, and, finally, reducing the role of radiation fission products and actinides dominance radiation after 30-50 years of aging.

Unique computer program that calculates SNF gamma radiation parameters was developed. The program takes into account isotope decay, geometry of the source, and concrete protection.

In this paper, for various reasons of such an important factor in assessing the quality of spent nuclear fuel as the neutron radiation, especially for spent nuclear fuel with a high level of burnout, remained outside the consideration. Along with the problem of dealing with nuclear operating waste these questions waiting for the optimal solution.

REFERENCES