

# Gamma-Ray Self-Absorption Corrections in Stainless Steel 12X18H10T for the Needs of Non-Destructive Isotopic Differentiation of Shielded Actinides

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**Abstract:-** One of the main tasks of nuclear science and technology is related to the development of methods of countermeasures, circulation, non-proliferation, and safe use of shielded nuclear materials - actinides. To solve this problem, information about their isotopic and quantitative composition is necessary. One of the main methods of non-destructive differentiation of shielded actinides, which is constantly being developed, is based on the use of their characteristic or stimulated gamma rays. For its implementation, information on the self-absorption of gamma rays of a wide energy range in screens (combinations of the elements from which they are made) is required. The results of calculations of the dependence of the self-absorption values of gamma rays in stainless steel 12X18N10 on their energy (100 keV ÷ 3000 keV) at fixed values of the screen thickness (0.1 ÷ 20 mm) are presented in the article. It was established that the self-absorption of gamma rays has smaller values for the energy range of spectrometric measurements of stimulated gamma rays compared to the energy range of spectrometry of characteristic gamma rays during the differentiation of shielded actinides. The energy range of gamma rays from 1000 to 3000 keV was determined, which can be considered optimal for the spectrometry of stimulated gamma rays from shielded actinides during their differentiation.

**Keywords:-** Shielded Actinides Differentiation; Gamma-Rays; Stainless Steel 12X18H10T; Mass Attenuation Coefficient; Self-Absorption.

## I. INTRODUCTION

Information on the isotopic and quantitative characteristics of fertile and fissile nuclear materials (actinides) is necessary to solve the current problems of nuclear technology related to their safe use and the development of methods to control their circulation and non-proliferation [1]. One of the main non-destructive methods for determining the isotopic and quantitative composition of unshielded and shielded nuclear materials (actinides) is based on gamma -spectrometric measurements of their characteristic or stimulated gamma rays [1,2].

To carry out nondestructive analysis of the isotopic and quantitative composition of unshielded samples of uranium (<sup>234,235,238</sup>U) and plutonium (<sup>239,240,241</sup>Pu), passive differentiation methods have been developed based on spectrometric measurements of their characteristic gamma rays [3-5]. This technique also determines the isotopic and quantitative composition of spent nuclear fuel (actinides and accumulated fission products of <sup>144</sup>Ce, <sup>134</sup>Cs, <sup>106</sup>Ru, <sup>125</sup>Sb, <sup>154</sup>Eu) [6].

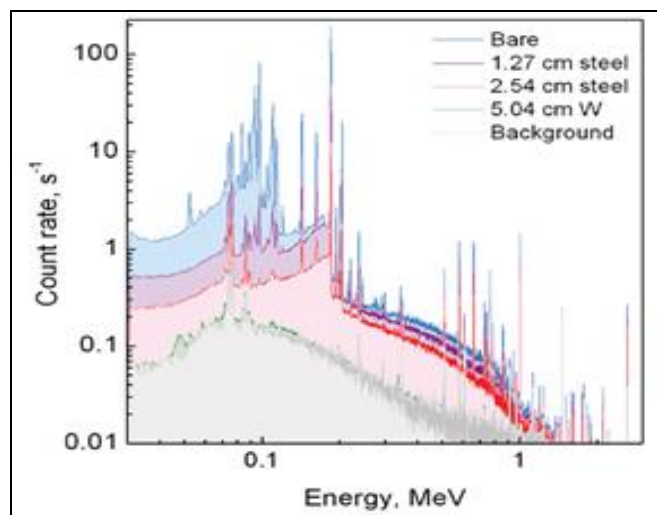


Fig 1 Gamma Spectra of Enriched Uranium with and without Absorbing Screens [11]

Also, this method is widely used to analyze shielded nuclear materials (particularly those located in sealed steel containers) - isotopes of uranium and plutonium [7-9]. Although with the help of passive spectrometric measurements under certain conditions, it is possible to detect actinides with light protection (for example, 5 mm of lead) by their characteristic gamma rays, it is not an effective detection method with heavier protection [10,11]. As an illustration, Fig. 1 shows the gamma spectra of enriched uranium, measured without a screen and with screens made of steel (thickness – 12.7 mm, 25.4 mm) and tungsten (thickness – 50.4 mm). As can be seen from Fig. 1, shielding with steel up to 25.4 mm thick still makes it possible to detect the characteristic gamma lines of enriched uranium with energies of 185.0 keV and 1001.0 keV, but a tungsten shield 50.4 mm thick is already opaque for these gamma lines [11].

Recently, for the isotopic differentiation of actinides and determination of their quantitative composition, methods of analysis based on their stimulated delayed gamma radiation (gamma rays from the products of their photo- and neutron fission) have been developed and used [1,11-17], which are more sensitive compared to methods of analysis of characteristic gamma-rays [10,11]. The photo-splitting differentiation of shielded (placed in metal containers) actinides has an advantage over neutron stimulation due to the threshold nature of photonuclear reactions [15,16]. For example, Fig. 2 shows characteristic gamma spectra of stimulated gamma rays of a sample of enriched uranium placed in a hermetic container made of stainless steel 12X18N10T and an empty activated container [16]. As can be seen from Fig. 2, gamma rays from the activation products of the chemical elements from which the container is made do not affect the analysis procedure when using stimulated gamma rays, even in the case when the mass of enriched uranium is approximately two orders of magnitude less than the mass of the container (the container is 70 g,  $^{235}\text{U}$  – 0.514 g) [16].

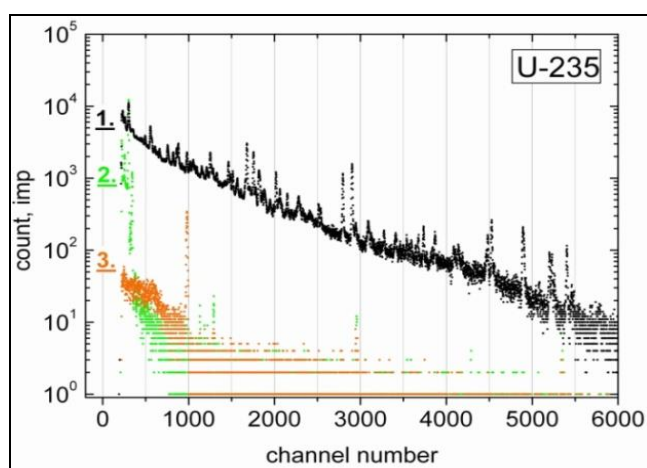


Fig 2 Gamma-Spectra: 1 – an Activated Sample of Enriched Uranium, 2 – its Characteristic Gamma Lines, 3 – Activation Products of the Chemical Elements from which the Container is made [16]

It should be noted that when applying the photofission reaction of shielded (in stainless steel containers) actinides to analyze their isotopic and quantitative composition, it is possible to use gamma rays of their fission products with energies greater than 1 MeV [16,17]. This expands the possibilities of this actinide differentiation method.

When differentiating actinides, measuring high-intensity gamma quanta from unactivated (characteristic gamma rays) and activated (gamma rays of nuclear reaction products) samples under investigation is necessary. Therefore, there is a need to weaken the intensity of gamma rays to reduce detector miscalculations (its dead time) and, accordingly, corrections for cascade summation [18]. To weaken the intensity of gamma rays during spectrometric measurements, the distance between the sample and the working surface of the detector is increased, which leads to a weakening of the effect of cascade summation of gamma quanta due to a decrease in the geometric efficiency of the detector [19,20], or absorbing

screens are used, which are installed between the sample and the working surface of the detector (which also allows reducing the intensity due to the absorption and scattering of gamma rays) [19,20]. It should be noted that the number of experimental studies on the effectiveness of measurements with absorbing screens is quite limited [19-25]. For example, screens made of Al (thickness – 20.0 mm) [21], Cu (1.0 mm) [22], Cd (1.5 mm) [23], Pb (1.0 and 2.0 mm), and stainless steel (1.0 mm, 9.1 mm, 20 mm) [19-21,25] are used to calibrate detectors according to energy efficiency [24].

It is known that the accuracy of spectrometric measurements depends on the accuracy of detector calibration in terms of energy efficiency, which must take into account corrections related to the measurement geometry and the absorption of gamma rays by the screen material [19,26-28].

Therefore, to optimize the experimental determination of the isotopic and quantitative composition of shielded actinides (or their differentiation) by their stimulated delayed gamma rays, information on the absorption properties (factors affecting the mass absorption coefficient) of the materials from which the screens (containers) are made is necessary.

Calculating the correction coefficient of self-absorption of gamma rays from actinide fission products by screens (containers) during spectrometric measurements is needed to forecast research results.

The purpose of the presented work is to calculate the coefficient of the self-absorption correction factor of gamma rays by stainless steel of the 12X18N10T brand for the energy range suitable for photo-splitting differentiation of actinides and to estimate the self-absorption depending on the energy of the gamma rays and the thickness of the screen.

## II. MATERIAL AND METODS

The use of stainless steel as a material for the manufacture of hermetic containers for packaging actinides or screens to weaken their characteristic stimulated gamma rays during spectrometric measurements to solve the problems of their isotopic differentiation [16,19,20] requires research on the mechanism of the processes of interaction of gamma rays with chemical elements, which contain stainless steel, in particular the 12X18H10T brand.

The interaction of gamma rays with electrons, nucleons, the electric field surrounding nuclei/electrons, and the meson field surrounding nucleons in matter results in complete absorption or scattering (elastic or inelastic) of gamma rays. The combination of these four types of interaction and the resulting mechanism implies twelve mechanisms of photon interaction with matter. The probability of the occurrence of a particular type of mechanism depends on the energy of the incident gamma rays, the scattering angle, the target material, and the experimental conditions [27,28].

For the energy range from 10 to 10,000 keV, there are four main interaction mechanisms: the photoelectric effect, Compton scattering, Rayleigh scattering, and vapor formation.

Other known interaction mechanisms, namely Thomson scattering, Delbrück scattering, nuclear resonance scattering, photodisintegration, and the formation of mesons do not affect the general process of interaction of gamma rays with matter [27,28].

In photoelectric absorption, a photon (gamma quanta) interacts with a tightly bound electron of an atom (electron of the K or L shell), resulting in its complete absorption. The probability of the appearance of this effect increases rapidly with an increase in the electron's binding energy, provided that the energy of the incident photon significantly exceeds the electron's binding energy in the atom. This effect dominates in the energy range from 1 keV to 500 keV (low energy). The cross-section of this process is inversely proportional to the photon energy (as  $1E^{3-5}$ ) and directly proportional to the atomic number of elements (as  $Z^{4-5}$ ). This indicates that this process is dominant for elements with a high Z number at photon energies  $< 500$  keV [27,28].

In Compton scattering, photons (gamma quanta) interact with an electron (assumed to be unbound and at rest). As a result, the photon's energy is partially transferred to the electron, and the photon is scattered along with the rest of the energy. Therefore, this mechanism leads to the degradation of gamma photon energy, resulting in inelastic/incoherent scattering. Compton scattering is the primary mechanism of interaction of gamma quanta in the energy range from 100 keV to 10 MeV (intermediate). The Compton scattering cross section slowly decreases with increasing gamma quanta energy [27,28].

In Rayleigh scattering, the atom is generally considered as a target. When a photon interacts with an atom, it scatters and changes direction to preserve momentum. In this process, the atom "recoils" to maintain momentum (that is, before and after scattering), and the recoil energy obtained in this way is very small and can be neglected due to the large mass of the atom. Therefore, the photon's energy remains unchanged before and after the collision. Thus, this process is coherent/elastic scattering of gamma rays from bound atomic electrons even at energies from 100 keV. This scattering mechanism dominates at low energies, high Z values, and small scattering angles (narrow beam geometry) [27,28].

Pair formation is a type of interaction in which photons (gamma rays) are completely absorbed. When photons interact with the electric field of electrons/nucleus, an electron-positron pair is formed. The threshold energy of the process for this interaction mechanism is 1022 keV. Electrons of atoms do not participate in this process except for the shielding function of the nuclear field. When the energy of gamma rays is above the threshold, it (as kinetic) is transmitted to electrons and positrons. This mechanism of interaction of photons with heavy elements dominates at photon energies greater than 1022 keV. The interaction cross-section depends on the atomic number of the element (as  $Z^2$ ) and the photon energy (as the logarithm of the energy) [27,28].

Thus, the abovementioned processes will affect the absorption properties of stainless steel to gamma rays (characteristic and stimulated) during spectrometric measurements of shielded actinide samples.

The change in the flux of gamma rays in a substance, which is associated with absorption and scattering, is characterized by such parameters as the mass absorption coefficient ( $\mu_m$ ), electron density ( $N_e$ ), and effective atomic number ( $Z_{eff}$ ).

The mass absorption coefficient ( $\mu_m$ ) is one of the main parameters characterizing the measure of the number of photons interacting (absorption, scattering) with a substance.

The effective atomic number ( $Z_{eff}$ ) is a parameter that represents the attenuation of gamma rays in a complex material. It is analogous to the atomic number (Z) of elements, which is given to complex materials (compounds and mixtures) to visualize photon interactions since it is not possible to provide a single value for photon interactions in the case of a compound/mixture containing different types of elements over a wide range of energies [27]. It is defined as the ratio of the total atomic cross-section to the total electronic cross-section.

Electron density ( $N_e$ ) is the average number of electrons per unit mass, measured in the number of electrons per gram. The greater the electron density value, the greater the probability of photon interaction [28].

The build-up factor is a parameter used to describe the scattering of the photon flux in the target material. In the case of "bad geometry" (for a wide beam of gamma quanta), a gamma ray passing through an absorbing screen generates two components of radiation - inside or outside the screen, namely gamma quanta that have undergone collisions and gamma quanta that have not. Therefore, it is necessary to introduce a correction factor (coefficient) that takes into account this process and significantly depends on the energy of gamma rays and the absorption properties of the screen material [19,20,27,29,30].

The mass absorption coefficient for a compound or mixture is obtained according to the Beer-Lambert law at photon energies from 1 keV to 100 GeV [31]:

$$\mu_m = \left(\frac{\mu}{\rho}\right) - \frac{\ln(I_0/I)}{\rho t} \quad (1)$$

$$I = I_0 e^{-\mu x} = I_0 e^{-\mu_m t} \quad (2)$$

Where  $\mu_m$  ( $\text{cm}^2 \cdot \text{g}^{-1}$ ) and  $\mu$  ( $\text{cm}^{-1}$ ) are mass and linear absorption coefficients, respectively;  $x$  (cm) and  $t$  ( $\text{g} \cdot \text{cm}^{-2}$ ) – thickness and mass thickness (mass per unit area), respectively;  $\rho$  ( $\text{g} \cdot \text{cm}^{-3}$ ) is the density of the material;  $I_0$  and  $I$  – initial and attenuated due to absorption and scattering intensities of gamma rays.

The total mass attenuation coefficient  $\mu_m$  for any composition of elements and/or compounds are determined by the mixture rule [31]:

$$\mu_m = \left(\frac{\mu}{\rho}\right) = \sum_i w_i (\mu/\rho)_i \quad (3)$$

$$w_i = n_i A_i / \sum_i n_i A_i \quad (4)$$

Where  $w_i$  is the mass fraction of the  $i$ -th constituent element;  $A_i$  is the atomic weight of the  $i$ -th element;  $n_i$  is the number of atoms of the  $i$ -th component of the element in the composite [31].

Theoretically, the mass absorption coefficient is calculated for a wide energy range of gamma rays, taking into account the general cross sections for several interactions in elements, mixtures, and compounds, using programs such as Xcom, Phy-X/PSD, and others [32-34].

The values of the mass absorption coefficient calculated in this way will reflect the dependence on the energy of the incident photon and the nature of the material interacting with it.

However, it should be noted that these calculations are valid only if certain specific conditions are met, namely monoenergetic gamma rays, narrow beam geometry, and thin screens. Therefore, these calculations are of an estimated nature [27,28].

Calculations of the self-absorption correction factor  $F_s$  of gamma rays in samples depending on their thickness were carried out according to equation (5) [35]:

$$F_s = \frac{1 - e^{-\mu_m t}}{\mu_m t} \quad (5)$$

The value of self-absorption in percent was calculated according to the equation:

$$S_A = (1 - F_s) \cdot 100\% \quad (6)$$

Xcom [33] and Phy-X/PSD [34] were used to calculate  $\mu_m$ .

Calculations were made for stainless steel of the 12X18H10T brand, which is used in nuclear engineering for packing radioisotopes and making screens to attenuate gamma rays during spectroscopic measurements [16,17,19,20].

A spectral analysis was performed to determine the content of chemical elements in a steel sample [36,37] since in the reference books of standards [38,39], only the percentage limits of the content of chemical elements in different steel grades are given. The content of chemical elements in 12X18H10T steel was: 6 C – 0.07 ( $\leq 0.12$ ), 14 Si – 0.6 ( $\leq 0.8$ ), 15 P – 0.03 ( $\leq 0.035$ ), 16 S – 0.02 ( $\leq 0.02$ ), 22 Ti – 0.8 ( $\sim 0.4 \div 1$ ), 24 Cr – 17.6 (17  $\div$  19), 25 Mn – 1.6 ( $\leq 2$ ),

26 Fe – 70.06 (65.73  $\div$  70.33), 28 Ni – 9.2 (9  $\div$  11), 29 Cu – 0.02 ( $\leq 0.3$ ). The values of the limit content of chemical elements in steel according to the reference books of standards [38,39] are given in parentheses. The density of stainless steel 12X18H10T – 7.9 g/cm<sup>3</sup> was determined by hydrostatic weighing according to the method [40].

### III. RESULT AND DISCUSSION

Fig. 3 shows the results of determining the mass absorption coefficients during the interaction of photons with stainless steel 12X18H10T depending on the energy of gamma rays calculated by the Xcom [33]. The main contribution for the energy range from 1000 to 3000 keV, which covers the energy range of spectrometric measurements of stimulated gamma rays during isotopic differentiation of actinides [16,17], is given by the Compton process scattering, namely incoherent scattering. The obtained data correlate with the results of calculations for other similar stainless steels in chemical composition [41].

Fig. 4 shows the results of calculating the dependence of the values of the mass absorption coefficient on the energy of gamma rays both for stainless steel 12X18H10T and, for comparison, for steels similar in chemical composition SS304, SS304L, SS316L, SSMn4N, MRGMoCrTi [42] by Xcom [33] and Phy-X/PSD [34]. The results obtained during calculations separately by each of these programs are consistent [32,33]. For the gamma-ray energy range of 1000  $\div$  3000 keV, the values of the mass coefficients for the abovementioned grades of steel match within the uncertainty of the calculations (inset in Fig. 4).

The parameters that depend on the attenuation of gamma rays in complex materials (stainless steels) are their effective atomic numbers and effective electron densities. Their energy dependences for a number of stainless and carbon steels were calculated in [41]. The results of the calculations [41] are shown in Fig. 5 and Fig. 6, respectively. Fig. 5 additionally presents the results of calculations of the dependence of the effective atomic number for stainless steel 12X18H10T on the energy of gamma rays. Calculations were performed using the Phy-X/PSD [34].

The results of the calculations show that the values of effective atomic numbers and effective electron densities for various grades of stainless (in particular, 12X18H10T) and carbon steels are practically constant (do not change within wide limits) for the energy range of gamma rays 1000  $\div$  3000 keV [41]. At the same time, for the energies of gamma rays, which cover the range of energies up to 200 keV [7-9], which is used for passive isotopic differentiation of shielded actinides, significant changes in the energy dependence of effective atomic numbers and effective electron densities for different grades of steel are observed [28-30,41].

It should be noted that the accumulation coefficient increases with increasing energy of photons (gamma rays) due to multiple scattering in the region of Compton scattering. In the area of high energies ( $> 3$  MeV), the formation of electron-positron pairs turns into the process of Compton

scattering. This effect may be associated with the dominance of the formation process of electron-positron pairs at energies above 1.22 MeV, which increases with increasing gamma-ray energy [29,30,43].

Thus, the range of gamma-ray energies from 1000 to 3000 keV can be considered optimal for spectrometric measurements of stimulated gamma rays from fission products of shielded actinides during their isotopic differentiation [14-17].

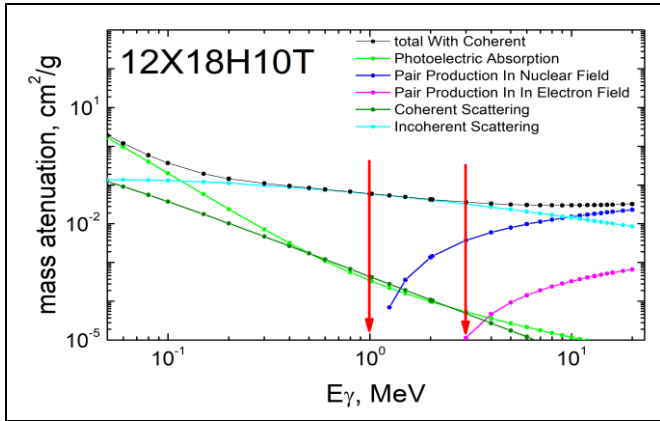


Fig 3 Dependence of Mass Absorption Coefficients for Partial Interactions of Photons with Stainless Steel 12X18H10T on their Energy

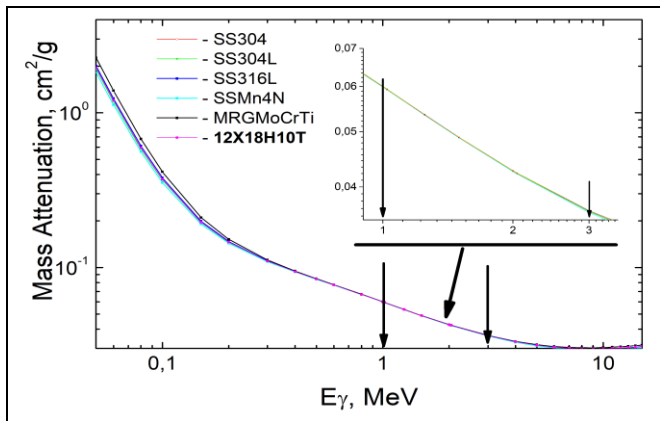


Fig 4 Mass Absorption Coefficients for Stainless Steels of Different Grades [42]

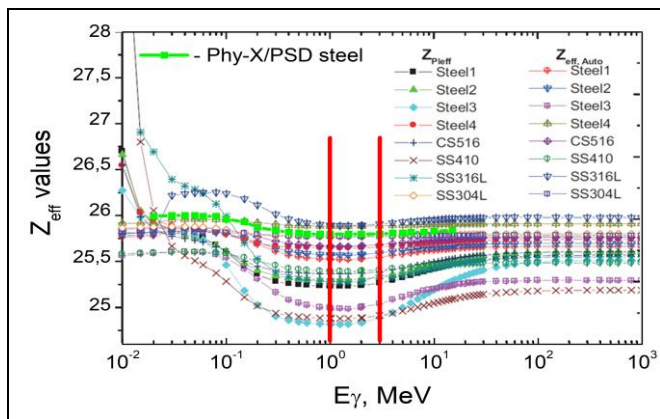


Fig 5 Variation in Effective Atomic Numbers of Stainless Steels with Photon Energy [41]

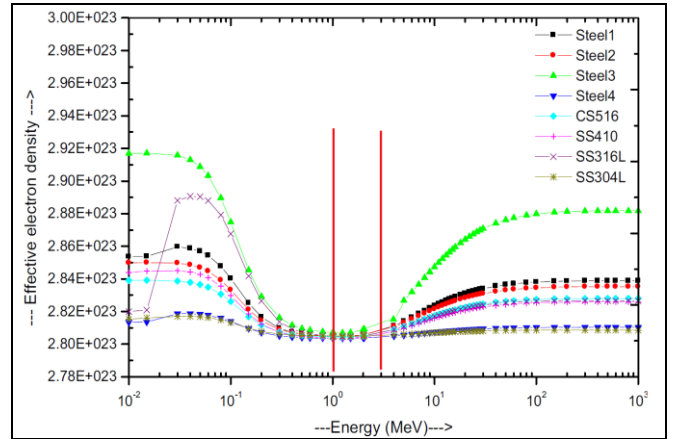


Fig 6 Variation in Effective Electron Densities of Stainless Steels with Photon Energy [41].

Fig. 7 shows the results of calculations of the self-absorption correction coefficient for gamma rays in stainless steel 12X18H10T depending on their energy (100 ÷ 3000 keV) at fixed thickness values (from 0.1 to 20 mm). Calculations were performed according to formula (5) [35] using the numerical values of the mass absorption coefficient obtained above (Fig. 4).

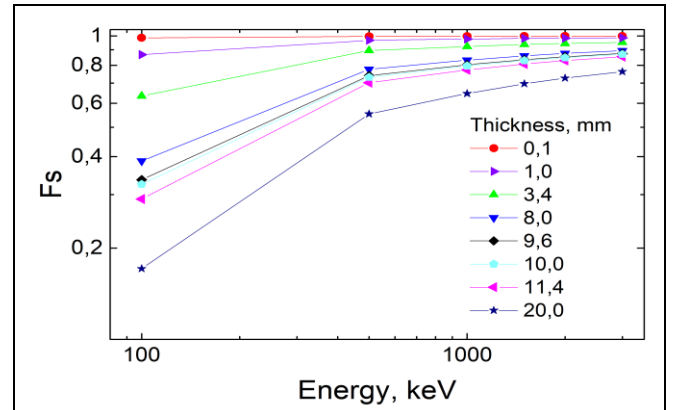


Fig 7 The Coefficient of Correction for the Self-Absorption of Gamma Rays in Stainless Steel 12X18H10T Depending on the Energy at Fixed Thickness Values

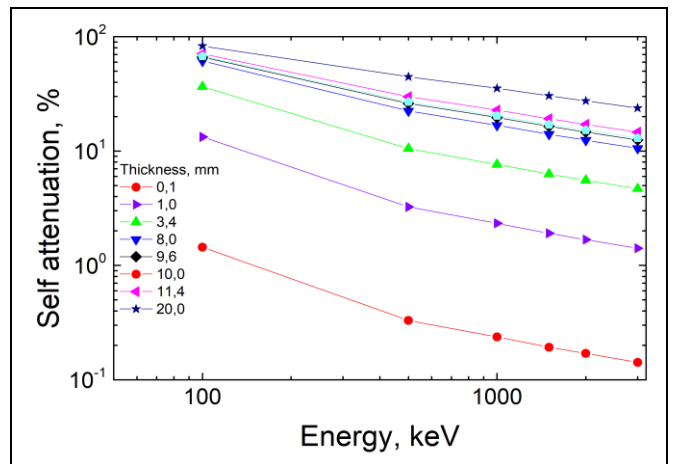


Fig 8 Dependence of the Self-Absorption Value of Gamma Rays in Stainless Steel 12X18H10T on their Energy at a Fixed Thickness

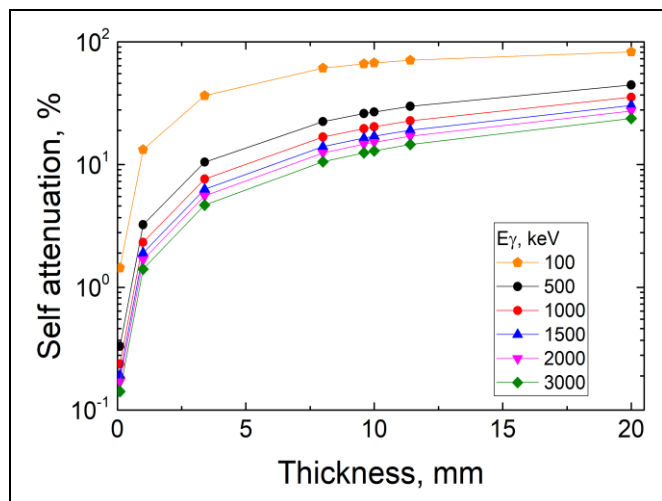


Fig 9 Dependence of the Self-Absorption Value of Gamma Rays in Stainless Steel 12X18H10T on the Thickness at Fixed Energy Values

The obtained values were used for calculating: a) the dependence of the self-absorption of gamma rays (as a percentage) of the value of the correction factor for self-absorption according to formula (6) in stainless steel 12X18H10T on the energy of gamma rays (100 ÷ 3000 keV) at a fixed screen thickness (from 0.1 to 20 mm); b) dependence of the self-absorption of gamma rays as a percentage of the screen thickness (0.1 ÷ 20 mm) at fixed values of the energy of gamma rays (100 ÷ 3000 keV).

Fig. 8 and Fig. 9 show the results of calculations of the self-absorption of gamma rays (in percent) in stainless steel 12X18H10T depending on their energy and the thickness of the absorber (screen).

It was established that the quantitative value of gamma-ray self-absorption in 12X18H10T steel at fixed screen thicknesses decreases with increasing gamma-ray energy (Fig. 8).

It was established that the quantitative value of self-absorption of gamma rays in 12X18H10T steel for fixed values of gamma-ray energy increases with increasing thickness (Fig. 9). However, with increasing gamma-ray energy, the value of self-absorption of gamma rays in 12X18H10T steel will have smaller numerical values.

It should be noted that the decrease in the values of self-absorption of gamma rays in steel is greater for the range of energies used for spectrometry of stimulated gamma rays [16,17] in comparison with the range of energies of spectrometry of characteristic gamma rays [7-9] during isotopic differentiation of shielded actinides.

#### IV. CONCLUSION

The results of calculations of the dependence of the self-absorption values of gamma rays in stainless steel 12X18N10 on their energy (100 keV ÷ 3000 keV) at fixed screen thickness values (0.1 ÷ 20 mm) are presented.

It was established that the self-absorption values of gamma rays have lower values for the energy range used for spectrometry of stimulated gamma rays compared to the energy range of spectrometry of characteristic gamma rays during isotopic differentiation of shielded actinides.

Based on the results of the analysis of the mass coefficients of gamma-ray absorption in various grades of stainless steel and the parameters on which the attenuation of gamma rays depends, the energy range of gamma rays (from 1000 to 3000 keV) was determined, which can be considered optimal for carrying out spectrometric measurements of stimulated gamma rays from fission products of shielded actinides during their isotopic differentiation.

The obtained values of gamma-ray self-absorption in stainless steel 12X18N10 can be used to predict the results of isotopic differentiation of shielded actinides during the nondestructive analysis of their characteristic and stimulated gamma rays and to determine the effectiveness of shielding gamma radiation of actinides for their safe use.

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