Contribution of Compton Scattering in Problems Associated with Measuring the Surface Density of Radiation Protection Coatings

B I Kapranov¹ and M A Mudrov²

¹ Section Head, Laboratory No. 40, National Research Tomsk Polytechnic University, Tomsk, Russia
² Principal Engineer of Accreditation, Monitoring And Diagnostics Regional Centre, National Research Tomsk Polytechnic University, Tomsk, Russia
E-mail: mudrov@tpu.ru

Abstract. The paper analyses the ratio between the number of Compton-scattered quanta excited in elements of radiation protection coatings by isotope Am-241 and the number of fluorescence quanta excited in the same elements, taking into account the fluorescence yield coefficient and the distance between the energy of primary radiation quanta (60 keV) and the energy of absorption K-edge. The paper shows that the number of scattered quanta exceeds the number of fluorescent quanta from K-level in all elements. The obtained values define the ratio of scattered and fluorescent quanta in any radiation protection coatings with a known chemical composition.

1. Problem of Controlling Protective Properties of Radiation Protection Coatings

Radiation protection coatings are widely used in the aviation and space engineering for protection against ionizing radiation. This issue is especially important for instrumentation, in particular for improving the operational reliability of electronics exposed to external X-ray radiation.

Instrument modules are protected with a special coating to effectively attenuate X-ray radiation in the energy range of up to 100 keV. The standard requirements for such coatings include mechanical strength, maximum attenuation efficiency and minimum weight. In order to fulfill these requirements, special coating compositions [1], production or application methods [2], and methods for measuring attenuating properties [3] are developed. Such coatings are most often made of RZK-6 (rare-earth concentrate) and RZO-4 (rare-earth oxide), which contain oxides of rare-earth elements [4, 5]. They are efficient due to the fact that absorption K-jumps occur in the energy range of up to 100 keV. The resulting attenuation coefficient is higher than the attenuation coefficient for these energies in lead, while the density of such materials is lower than lead density.

In order to control the quality of such coatings, the fluorescent method is most often used. This method is aimed at irradiating the coating-base combination with primary γ- or X-rays and recording the intensity of the fluorescent quantum flux from elements of the coating. In this case, the measured parameter is the surface density ρ expressed in g/cm². With the increase in the surface density, the intensity of the fluorescent quantum flux grows. The devices developed for such control are based on the one-sided access geometry [6–9]. The long-lived isotope Am-241 is most often used as a primary radiation source. The development of methods and equipment for control of such coatings is always associated with the problem of the ratio between intensities of fluorescence and Compton scattering radiation fluxes. The Compton radiation is generated both in elements of the coating and in the base...
covered with such a coating. The resulting secondary flux has a complex energy spectrum. The type of the spectrum depends on a number of factors, including geometrical thickness of the coating and the base; additional excitation of elements by radiation scattered from the base; subexcitation of K-lines of elements with a lower atomic number by K-lines of elements with a higher atomic number; scattering of fluorescent quanta by the base from the elements.

This paper studies the contribution of Compton scattering to the total secondary flux occurring in elements of the coating excited with γ-quanta of Am-241. To solve this problem, let us consider the geometry shown in figure 1.

**Figure 1.** Diagram for calculation of fluorescence radiation and scattered radiation fluxes.

$N$ is the source of radiation (monochromatic, energy $E_0$); $n_0$ is the number of photons per unit of time emitted by the source into a solid angle $4\pi$ (intensity); $D$ is the detector; $H$ is the distance from the “source – detector” base to the object; $h$ is the thickness of the coating; $d$ is the thickness of the base; $\alpha$ is the incidence angle of the primary beam; $\beta_s, \beta_0$ are the emergence angles of the secondary radiation from the coating and the base, respectively; $Q_{ss}, Q_{so}$ are the scattering angles of the secondary radiation on the detector from the coating and the base, respectively; $x_s, x_0$ are the coordinates of the points of interaction between the primary beam in the coating / the base, respectively; AB is the line of interaction between the primary beam and the coating; BC is the line of interaction between the primary beam and the base.

### 2. Monochromatic Source of Isotope Am$^{241}$

The radiation from source N (assume the flux to be “narrow”) is incident on the object at angle $\alpha$. Primary radiation quanta pass through the coating with thickness $h$ along line AB where they are attenuated. Then, they get into the base with thickness $d$ where they are scattered on line BC. The total secondary flux in detector D is composed of fluorescent quanta (only k-fluorescence is taken into account) from elements of the coating, Compton-scattered quanta in elements of the coating, Compton-scattered quanta in the base, with allowance for attenuation of the primary flux and the secondary flux caused by the coating and other factors. We assume that fluorescent quanta from the element of coating $dx_s$ at point $x_s$ escape into the detector at an angle $\beta_s$ to the normal.
Radiation source \( N \) and detector \( D \) are at distance \( H \) from the test object in positions \( y_N \) and \( y_D \). Let us designate the surface of the object exposed to the primary beam as \( S \). We also assume that the linear dimensions of coating \( h \) and area \( S \) are small as compared to \( H \). The interaction between the primary beam and the coating is assumed to occur along line \( AB \), the length of which is determined as \( \frac{h}{\cos \alpha} \).

The intensity of the primary quantum flux at point \( x_s \) of the coating is determined by the geometric divergence and attenuation on the way from point \( A \) to the interaction point:

\[
n(x) = \frac{n_o}{4\pi} \left( \frac{H}{\cos \alpha} + \frac{x}{\cos \alpha} \right) \cdot e^{-\mu_s \frac{x}{\cos \alpha}},
\]

where:

\( \mu_s \) is the linear attenuation factor of the primary radiation in the coating material;

\( \frac{x}{\cos \alpha} \) is the path of the primary radiation in the coating to the interaction point.

Passing through the layer \( dx_s \), the intensity of the primary beam is reduced by the value \( dn_s(x_s) \):

\[
dn_s(x_s) = -\mu_s \cdot \frac{1}{\cos \alpha} \cdot n_s(x_s) \cdot dx_s,
\]

provided that \( h < H \).

In this expression, let us designate the intensity of the primary quantum flux at the entry to the product as \( n_1 \). If we consider the radiation source to be a point source,

\[
n_1 = \frac{n_o}{4\pi \left( \frac{H}{\cos \alpha} \right) \cos \alpha}.
\]

Then

\[
dn_s(x_s) = -n_1 \cdot \frac{\mu_s^s \rho_s \cos \alpha}{\cos \alpha} \cdot e^{-\mu_s^s \rho_s \frac{x_s}{\cos \alpha}} \cdot dx_s,
\]

where:

\( \mu_s^s \) is the mass attenuation factor of the primary beam in the coating material;

\( \rho_s \) is the density of the coating material.

The total number of quanta in the primary beam absorbed in the coating on line AB is determined by integrating \( dn_s(x_s) \) throughout the thickness of coating \( h \).

\[
\Delta n_s(h) = n_1 \cdot \mu_s^s \rho_s \cdot \frac{1}{\cos \alpha} \left( 1 - e^{-\mu_s^s \rho_s \frac{h}{\cos \alpha}} \right).
\]

The relative share of quanta in the primary beam absorbed in the coating will be:

\[
\delta n_s(h) = \frac{\Delta n_s(h)}{n_1} = \mu_s^s \rho_s \cdot \frac{1}{\cos \alpha} \left( 1 - e^{-\mu_s^s \rho_s \frac{h}{\cos \alpha}} \right).
\]

Therefore, the quantum flux of the primary beam passed through the coating, i.e. at the entry to the base, will be equal to \( n_2 = n_1 - \Delta n_s(h) \) or:

\[
n_2 = n_1 \cdot \left[ 1 - \mu_s^s \rho_s \cdot \frac{1}{\cos \alpha} \left( 1 - e^{-\mu_s^s \rho_s \frac{h}{\cos \alpha}} \right) \right].
\]

Quanta of primary radiation in the coating material are subject to two processes: photoabsorption and scattering. The ratio between probabilities of these processes is given the photoabsorption section \( \tau \) and the scattering section \( \sigma \) \([10]\). The total mass section of the interaction in the coating is determined as:

\[
\mu_s^s = \tau_s^s + \sigma_s^s.
\]

Hence, the intensity of the primary radiation quantum flux which has interacted with photoabsorption will be:

\[
\Delta n_s(h)_{\text{absorb}} = n_1 \cdot \tau_s^s \rho_s \cdot \frac{1}{\cos \alpha} \left( 1 - e^{-\mu_s^s \rho_s \frac{h}{\cos \alpha}} \right),
\]

and the intensity of the primary radiation photon flux which has Compton-interacted will be:

\[
\Delta n_s(h)_{\text{Compt}} = n_1 \cdot \sigma_s^s \rho_s \cdot \frac{1}{\cos \alpha} \left( 1 - e^{-\mu_s^s \rho_s \frac{h}{\cos \alpha}} \right) \tag{1}
\]
The fluorescence radiation yield is associated only with photoabsorption $\tau$. Each photoabsorbed photon of primary radiation excites one atom in the coating substance [11], and the number of the atoms that have emitted fluorescence quanta is defined by the probability $\omega_k$ of the fluorescence yield:

$$\Delta n_{fl}^k = \omega_k \cdot \Delta n_{\text{absorb}}(h).$$

$\omega_k$ is determined by experiment as a ratio of the fluorescent quantum flux $n_{fl}^k$, to the primary quanta flux photoabsorbed in the material $\Delta n_{\text{absorb}}(h)$. The most frequently used formula by Stephenson [11]. The calculated values of the fluorescence yield coefficient $\omega_k$ of the elements included in the coating are shown in Table 1 [11].

If we consider only the fluorescence yield during the excitation of $K$-level of elements, doublet $K\alpha_1 K\alpha_2$ is excited. Its line energies $E_{K\alpha_1}$ and $E_{K\alpha_2}$ are defined by the energies of transitions from $K$-level to level $L_{III}$ and from $K$-level to level $L_{II}$, respectively. The numerical values for these elements are also shown in Table 1.

3. The ratio between Intensities of Lines

The ratio between intensities of lines belonging to the same multiplet is calculated by the Burger and Dorgelo rule [11]: If we neglect splitting of the initial multiplet levels (which is permissible for $K$-level of all elements since they differ only by the spin number), the intensities of the line of multiplet $K\alpha_1 K\alpha_2$ will be proportional to the statistical weights of the final levels $L_{III}$ and $L_{II}$ respectively. Since the top level of multiplet $K\alpha_1 K\alpha_2$ is common, then:

$$j_{\alpha_1} j_{\alpha_2} = \frac{\tau_{\alpha_1}}{\tau_{\alpha_2}} = \frac{2}{1}.$$

The energies of lines $K\alpha_1$ and $K\alpha_2$ differ insignificantly and cannot be resolved with a scintillation spectrometer. Each doublet will be recorded in aggregate. In order to determine the energy of $K$-doublet, we can use the concept of effective energy defined as the sum of the weighted energies of lines $K\alpha_1$ and $K\alpha_2$. The quantitative ratio of the energy contribution of lines $K\alpha_1$ and $K\alpha_2$ is defined by the relative intensity of these lines. According to [11], for the elements with Z of 23 to 90, the ratio $j_{\alpha_1} j_{\alpha_2} = 2.00$ with a deviation of not more than $\pm 0.05$. Therefore, this paper assumes the weight of line $K\alpha_1$ to be 2, and the weight of line $K\alpha_2$ to be 1. Then, the effective energy of the doublet will be determined as:

$$E_{\text{eff}} = \frac{2}{3} \cdot E_{K\alpha_1} + \frac{1}{3} \cdot E_{K\alpha_2}$$

Table 1 shows the effective energies of $K$-doublets for elements of coatings calculated from this ratio. The weighted sum of the effective energies of $K$-doublets in multicomponent coatings will define the effective energy of the fluorescence radiation spectrum excited in a coating of a specific composition.

Another factor that defines the fluorescent quantum yield is the dependence on the energy of exciting radiation. The largest fluorescence yield from the $K$-level will be caused by the energy of primary radiation that is probably closer to the energy of the absorption $K$-edge $E_k$. But the condition $E_0/E_k > 1$ must be satisfied. The dependence of the excitation probability on the ratio of these energies is described by the function [11]:

$$\varphi \left( E_0/E_k \right) = \left( \frac{E_0}{E_k} \right)^3 \sin \alpha + \sin \beta.$$

where $S_k$ is the absorption $K$-jump.

The numerical values $S_k$ for elements of the coating are also presented in Table 1.

The value of the function $\varphi \left( E_0/E_k \right)$ is maximum at the ratio of energies $E_0/E_k = 1$ and given by $S_k^{\text{eff}}/S_k$ when angles $\alpha$ and $\beta$ are equal. As $E_k$ moves away from $E_0$ towards lower values, the excitation probability value $\varphi$ decreases. If isotope Am-241 is used as a source, the excitation probability for $K$-
line of elements decreases with decreasing the atomic number, as shown in Table 1. Hence, the full fluorescent quantum yield for elements of coatings made of RZK-6 and RZO-4 is described by the following ratio, provided that isotope Am-241 is excited by the radiation:

$$\Delta n_{ff}^f = \omega_k \cdot n_1 \cdot \frac{\tau_1 \rho_s}{\cos \alpha} \cdot \left( 1 - e^{-\mu \rho_s \cos \alpha \cdot \frac{k}{h}} \right).$$

Taking into account that the total number of Compton-scattered quanta in elements of radiation protection coatings is determined by ratio (1), we can determine the ratio of the flux intensity of Compton scattering to the total flux of the characteristic emission excited in the elements:

$$\delta_k = \frac{\Delta n_{ff}^f (h)_{Compt}}{\Delta n_{ff}^f}.$$ (2)

**Table 1.** Energy of K-absorption edge ($E_k$), effective energy lines K – Series ($E_{eff}$), the quantum efficiency of the fluorescence yield K – lines ($\omega_k$), the probability of fluorescence excitation ($\omega$), the ratio of the flux intensity of Compton scattering to the total flux of the characteristic emission excited in the elements included in RZK-6 (rare-earth concentrate) and RZO-4 (rare-earth oxides) when used as the primary radiation isotope Am-241.

| Z | Element | $E_k$, keV | $E_{eff}$, keV | $\omega_k$, % | Absorption jump $S_k$, relative units | $\omega$, relative units | $\delta_k$, relative units |
|---|---------|------------|----------------|---------------|--------------------------------|------------------|------------------|
| 11 | Sodium Na | 1.073 | 1.043 | 0.035 | 15.2 | 0.017 | 19469 |
| 14 | Silicon Si | 1.847 | 1.744 | 0.04 | 13.2 | 0.030 | 1533 |
| 15 | Phosphorus P | 2.144 | 2.019 | 0.055 | 12.6 | 0.033 | 764 |
| 20 | Calcium Ca | 4.037 | 3.700 | 0.165 | 10.7 | 0.067 | 43.2 |
| 26 | Iron Fe | 7.112 | 6.412 | 0.32 | 9.0 | 0.12 | 4.54 |
| 39 | Yttrium Y | 17.038 | 14.970 | 0.68 | 7.0 | 0.29 | 0.231 |
| 48 | Cadmium Cd | 26.711 | 23.165 | 0.76 | 6.0 | 0.46 | 0.066 |
| 50 | Tin Sn | 29.200 | 25.258 | 0.75 | 5.9 | 0.50 | 0.059 |
| 57 | Lanthanum La | 38.925 | 33.417 | 0.69 | 5.4 | 0.68 | 0.026 |
| 58 | Cerium Ce | 40.443 | 34.177 | 0.68 | 5.3 | 0.71 | 0.024 |
| 59 | Prasodymium Pr | 41.991 | 35.910 | 0.66 | 5.3 | 0.74 | 0.021 |
| 60 | Neodymium Nd | 43.569 | 37.236 | 0.66 | 5.2 | 0.78 | 0.0196 |
| 62 | Samarium Sm | 46.834 | 39.950 | 0.64 | 5.1 | 0.86 | 0.0161 |
| 63 | Europium Eu | 48.519 | 41.355 | 0.62 | 5.0 | 0.89 | 0.0149 |
| 64 | Gadolinium Gd | 50.239 | 42.823 | 0.61 | 5.0 | 0.89 | 0.0144 |
| 65 | Terbium Tb | 51.996 | 44.279 | 0.60 | 4.9 | 0.94 | 0.0130 |
| 66 | Dysprosium Dy | 53.788 | 45.850 | 0.59 | 4.9 | 1.00 | 0.0116 |
| 67 | Holmium Ho | 55.618 | 47.308 | 0.56 | 4.8 | 1.00 | 0.0112 |
| 68 | Erbium Er | 57.486 | 48.855 | 0.55 | 4.7 | 1.00 | 0.0108 |
| 70 | Ytterbium Yb | 61.332 | 52.094 | 0.51 | 4.7 | - | - |
| 71 | Lutetium Lu | 63.316 | 53.769 | 0.51 | 4.6 | - | - |
| 82 | Lead Pb | 88.004 | 74.340 | - | 4.1 | - | - |
| 83 | Bismuth Bi | 90.526 | 76.454 | - | 4.1 | - | - |

4. Conclusion

The analysis shows that the source of Am - 241 effectively excites K - line elements to erbium ($Z = 68$). K - Ytterium element line ($Z = 70$), lutetium ($Z = 71$), lead ($Z = 82$), bismuth ($Z = 83$) are not excited. Therefore, for their account to be used L - lines, respectively: 10.50 keV; 10.87 keV; 15.85 keV; 16.39 keV. The contribution to the total flow of the scattered radiation is less than 23%,
since the yttrium (Z = 39). Thus, to control the thickness (areal density) of radiation - coatings can be effectively used fluorescent method with the isotope Am - 241 as the primary radiation source.

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