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Density and Interaction Data for Lutetium-Yttrium Ortho-Aluminate-Based Scintillation Crystals for Gamma-ray Spectroscopy and Imaging

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Abstract

The density values of (Lu_pY_{1-p}) AP scintillators based on the results of a literature survey have been found strongly positively linearly correlated to the value of the parameter p, that represents the ratio of lutetium vs. yttrium oxides content by weight. Two cases of serious lack of information have been investigated to search reliable data. Further studies are in progress aimed at inferring possible unknown compositions by using the data published by researchers or manufacturers or from scintillators databases.

Keywords: Lutetium-yttrium ortho-aluminate, Monte Carlo simulation, gamma-ray spectroscopy, radionuclide medical imaging, SPECT, PET

INTRODUCTION

In general, the design and the evaluation of a detector for gamma-ray spectroscopy and/or imaging require a knowledge of the basic specifications of scintillation materials better than that obtainable from the present status of technical documentations from manufacturers. For Monte Carlo simulations, accurate information about the scintillator characteristics is of prime importance.

For the (Lu-Y) based scintillators quite all the manufacturers show a certain reluctance to give complete specifications, as hereafter illustrated:

a. The Saint-Gobain Crystals (Fr) is the only one that, for the LYSO scintillators, gives the composition as $Lu_{1.8}Y_{0.2}SiO_5$ crystals, the 7.1 g/cm^3 density, and the attenuation length for 511 keV of 1.2 cm values (Saint-Gobain Crystals, 2021)
b. The Crytur company (Cz) specifies for the scintillation crystal so-called CRY019 some basic properties like density of 7.1 g/cm^3 and a "radiation length for 511 keV" of 2.1 cm, but does not specify information about their composition [1]
c. The Dynasyl company (Uk, before: Hilger-Crystals) gives information in a concise way announcing: "Lutetium Yttrium Silicate (LYSO): A non-hydroscopic scintillator that is both bright and fast" [2]
d. The Advatech company (Uk) also provides for LYSO:Ce the density value of 7.15 g/cm^3 as well as the radiation length value of 1.1 cm, while in [3] the composition is not specified.

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e. The Kinheng-Crystal company (Cn) gives for LuYAP crystals only the density value of 7.44 g/cm³ [4]
f. The Filar Optomaterials company (It) only cites Lu(Y)AP and L(Y)SO scintillators in their products catalog, without giving more technical information [5].

As the reader can realize, an estimation of the value of a missing quantity among i) the density, ii) the elemental composition, and iii) the radiation length can be deduced for LuYAP crystals, referred to, in the present work, by using the values of the two remaining quantities. It is worth noting that the contents of doping elements is not considered for the interactions calculations for LuYAP:Ce compounds because the Ce content is in the range of a few units ‰ in weight that cannot affect not so much the results. It equally worthy that this is not a general rule, because the content of dopant elements can range from 0 to some units %, like in the case of BGO, and $LaBr_3$: Ce because, respectively, the first one is an intrinsic scintillator, and the second one may reach the 5 % in weight of dopant content.

Similar studies can be carried out for other multi-component scintillators like those based on (Lu-Y) SO or (Lu-Gd) SO mixtures, whose results would allow to verify the agreement between the quantities declared by the manufacturers.

METHODS

Density Values

A survey of (Lu_pY_{1-p}) AlO_3: Ce density values of scintillation crystals from the literature has been carried out. Among the huge quantity of papers on the LuYAP: Ce, only the ones cited hereafter have been found reporting on results including density measurements of samples. It is worth noting that *p* represents the values of the ratios of lutetium to yttrium contents in the oxide's mixtures $Lu_2O_3 + Y_2O_3$ used for growing the considered crystals [6]. The results of the survey density values are synthesized in chronological order in Table 1.

Table 1. Summary of (Lu_pY_{1-p})AP:Ce values from the literature survey [7–13].

Reference	*p* value	Compound id.	Meas. Density (g/cm^3)	Manufacturer	Calc. Density (g/cm^3)
(Moses, 1995)	1	LuAP	8.34	ISC (1)	8.27
(Chval, 2000)	0.1	Lu1	5.73	Crytur, Cz	5.65
	0.2	Lu2	5.92		5.94
	0.3	Lu3	6.19		6.23
	0	YAP	5.36		5.36
(Petrosyan, 2000)	0.65	Lu6.5	7.3±0.1	IPR (2)	7.25
(Weber, 2003)	0.8	Lu8	7.7±0.1	BTP (3)	7.69
(Weber, 2004)	0.7	Lu7	7.4	not specified (4)	7.40
(Annenkov, 2004)	0.7	Lu7	7.2±0.1	BTP (3), INP (5)	7.40
(Balcerzyk, 2004)	0.7	Lu7	7.1	PM (6)	7.40
(Szupryczynski, 2005)	0.7	Lu7	7.34	BTP (3)	7.40

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Photon Interaction Data

The calculations of photon interaction data for the selected (Lu_pY_{1-p}) AlO_3 compounds of interest, have been performed at NIST website (US National Institute of Standards, 2021), by using the XCOM software and the XGAM web-database. XGAM includes the photon cross sections for scattering, photoelectric absorption, and pair production, at energies from 1 keV to 100 GeV, where the energy-range from 10 keV to 1 MeV of interest for SPECT and PET imaging is included.

Seven runs have been performed, corresponding to the compounds described by the given chemical formulas. The work has been planned as 6 steps of 0.2 *p* -spaced values for covering the interval from 0 to 1, plus a seventh intermediate step for $p = 0.7$ that is the most popular lutetium compound on the market. The results of calculations are shown and are reported in numerical mode in the Appendix (Tables A1–A7). Table 2 shows the values of elemental fractions by weight computed by the XCOM software per each *p* value. These data were used by the software as input data for the calculations of the mass attenuation coefficients (in cm^2/g) for the given grid of energy values [14]. At the end of each step XCOM produced a list of the mass attenuation coefficient values related to each kind of interaction for a gamma-ray traveling in such a crystal calculated by using the XGAM database. It is worth noting that, in the energy-range of interest for gamma-ray medical imaging, the scattering and the photoelectric absorption exhaust the chances of interactions. Therefore, the trends of photoelectric absorption mass attenuation coefficient, and the photo-fraction data are presented in the following, to describe the behavior of such a kind of crystal. For a thorough examination of gamma-ray interactions the reader can consult [15]. Results of interaction data per each kind of crystal are reported in the Appendix.

RESULTS AND DISCUSSION

Density

Calculated values have been obtained where the slope and intercept values have been obtained by a linear fitting of experimental data, with a R^2 value of 0.9956. The maximum deviation between measured and calculated values resulted of 0.3 g/cm^3 for the density value reported in Figure 1 [13].

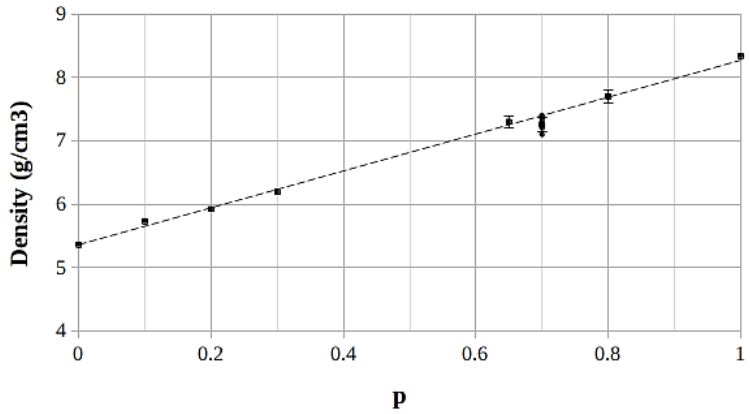

Figure 1. Plot of density values as a function of the ratio *p* of lutetium vs. yttrium contents in the oxides mixtures used for growing the crystals. Measured values are from the references belonging to the survey of literature above described (see Table 1).

The results agree very well with the linear expression of Eq.(1), fitting the experimental data, where the independent variable is *p*, the slope is represented by the difference between the maximum and the minimum density values, and by the intercept value:

$$Density_{(p)} = 2.91 * p + 5.36, (g/cm^3) \quad (1)$$

that is:

$$Density_{(p)} = (Density_{(1)} - Density_{(0)}) * p + Density_{(0)}, \quad (2)$$

or:

$$Density_{(p)} = Density_{(1)} - Density_{(0)} * (1 - p). \quad (3)$$

It is worthy that the Eq.(3) is suitable for more general use, like in the cases of the mixed scintillators like (Lu-Y)SO, or (Lu-Gd)SO.

Photoelectric_Absorption_Coefficients

The values of elemental fractions by weight per *p* values, and for the considered compounds, from XCOM preliminary calculations performed at NIST website are reported in Table 2.

Table 2. Values of elemental fractions by weight per *p* values for the considered compounds from XCOM calculations performed at NIST website (US National Institute of Standards, 2021).

***p* value**	**Compound id.**	**Chemical Formula**	**Oxygen (*Z* = 8)**	**Aluminum (*Z* = 13)**	**Yttrium (*Z* = 39)**	**Lutetium (*Z* = 71)**
0.	YAP	$YAlO_3$	0.292876	0.164636	0.542487	0.
0.2	Lu2	$(Lu_2Y_8)Al_{10}O_{30}$	0,265040	0.148989	0.392742	0.193229
0.4	Lu4	$(Lu_4Y_6)Al_{10}O_{30}$	0.242036	0.136057	0.268990	0.352916
0.6	Lu6	$(Lu_6Y_4)Al_{10}O_{30}$	0.222706	0.125191	0.165005	0.487097
0.7	Lu7	$(Lu_7Y_3)Al_{10}O_{30}$	0.214155	0.120384	0.119002	0.546459
0.8	Lu8	$(Lu_8Y_2)Al_{10}O_{30}$	0.206236	0.115933	0.076401	0.601430
1.	LuAP	$LuAlO_3$	0.192034	0.107949	0.	0.700017

The results of the *Mass_Photoelectric_Absorption_Coefficient* values (in cm^2/g) are reported in graphical way in Figure 2 as a function of the photon energy, where the curves related to the considered crystals are referred to by the legend, in the same order, from the top to the bottom. The numerical results from calculations are reported in the Appendix.

Since:

$$Linear\_Attenuation\_Coefficients = Mass\_Attenuation\_Coefficients^* Density, \quad (4)$$

The trends of the *Linear_Photoelectric_Absorption_Coefficient* values have been obtained by using the Eq. (4), are shown in Figure 3. It is worth noting that the curves seem better spaced, with respect to those of Figure 2, due to the density values which increase, passing from the YAP (*p*= 0) to the LuAP (*p* = 1), more than the respective values of *Mass_Photoelectric_Coefficients* in the given energy range.

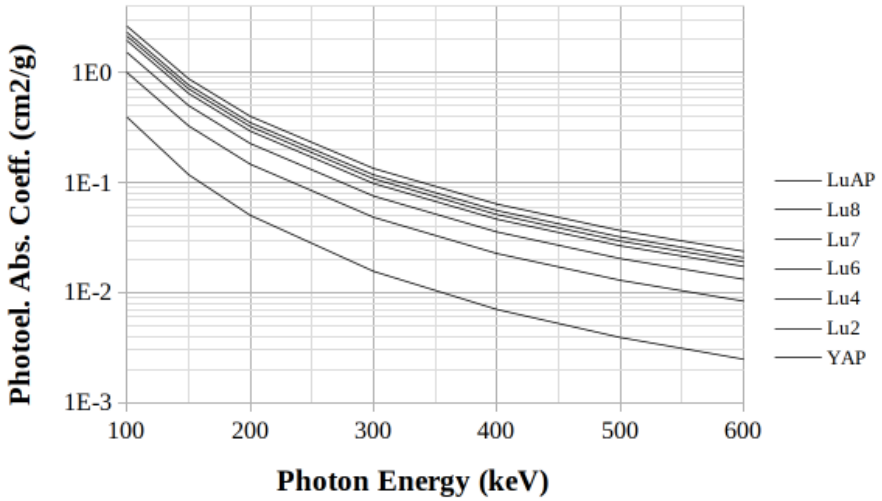

Figure 2. Semilog plot of *Mass_Photoelectric_Absorption_Coefficient* values (in cm^2/g) as a function of the photon energy calculated by the XCOM software for the compounds reported in the legend, which refers to the curves in the same order, from the top to the bottom. The corresponding values of *p* (i.e. the ratios of lutetium vs. yttrium contents in the oxides mixtures used for growing the crystals) are reported in Table 2.

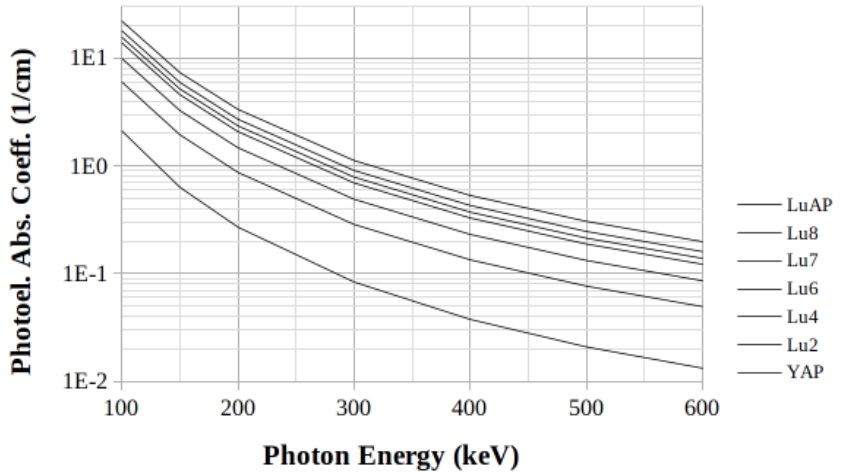

Figure 3. Semilog plot of *Linear_Photoelectric_Absorption_Coefficient* trends (in cm^{-1}) as a function of the photon energy calculated for the compounds reported in the legend, which refer to the curves, in the same order from the top to the bottom. The corresponding values of *p* (i.e. the ratios of lutetium vs. yttrium contents in the oxides mixtures used for growing the crystals) are reported in Table 2.

In the notation of Eq. (4) the dependence of terms on the photon energy, as well as that of crystal composition are omitted for ease of reading.

The *Mean_Free_Path*, whose value is defined as the average path covered in the crystal before a photoelectric or scattering interaction occurs, can be calculated as:

$$Mean\_Free\_Path = 1 / Total\_Linear\_Attenuation\_Coefficient \quad (5)$$

The trends of *Mean_Free_Path_Coefficient* values have been obtained from *Total_Linear_Attenuation* values by using the Eq. (5). The curves are shown in Figure 4, where they are referred to by the legend items, in the same order, from the top to the bottom. The numerical results from calculations are reported in the Appendix.

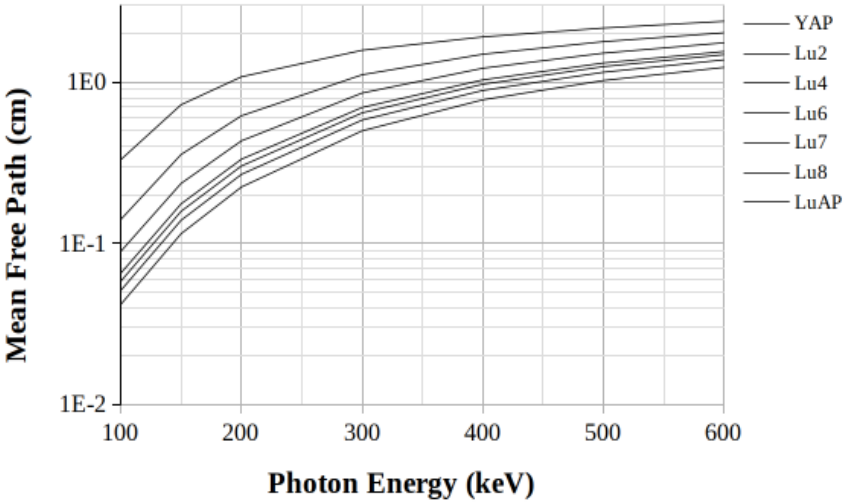

Figure 4. Semilog plot of *Mean_Free_Path* trends (in cm) as a function of the photon energy, calculated by using the Eq.(5) for the compounds reported in the legend, which refer to the curves in the same order, from the top to the bottom. The corresponding values of *p* (i.e. the ratios of lutetium vs. yttrium contents in the oxides mixtures used for growing the crystals) are reported in Table 2.

The *Photo_Fraction* values, have been calculated as

$$Photo\_Fraction = (\ Mass\_Photoelectric\_Coefficient\ ) / (\ Total\_Mass\_Coefficient). \quad (6)$$

The trends of *Photo_Fraction* values have been obtained from *Total_Linear_Attenuation-Coefficient* values by using Eq.(6). The curves are shown in Figure 5, where they are referred to by the legend items, from the top to the bottom.

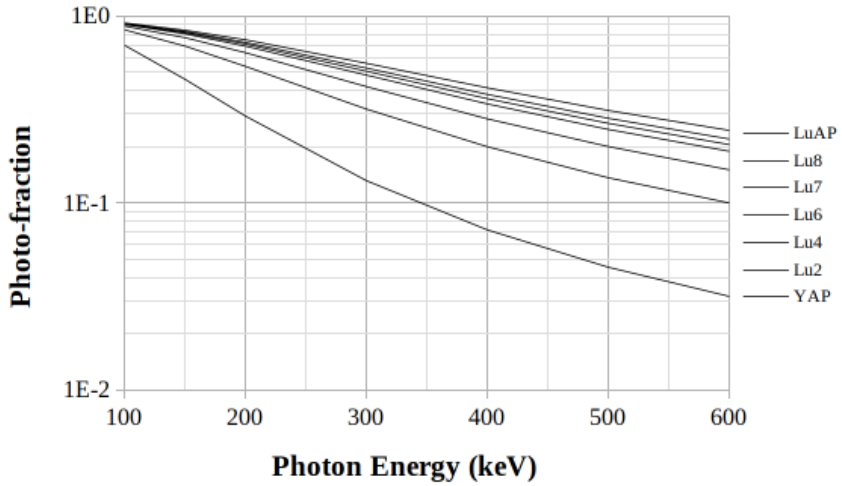

Figure 5. Semilog plot of *Photo-Fraction* values as a function of the photon energy ones, calculated by using the Eq.(6) for the compounds reported in the legend, which refer to the curves, in the order from the top to the bottom. The corresponding values of *p* (i.e. the ratios of lutetium vs. yttrium contents in the oxides mixtures used for growing the crystals) are reported in Table 2.

CONCLUSION

The density values of (Lu_pY_{1-p}) AP scintillators from the survey of literature have been found strongly positively correlated to *p*, that is the value of the ratio of lutetium vs. yttrium contents in the oxides mixture $Lu_2O_3 + Y_2O_3$ used for growing the crystals (see Eqs (1-3)). It is worthy that the Eqs (1-3) are suitable for more general use, like in the cases of the mixed scintillators like (Lu-Y)SO, or (Lu-Gd)SO.

Two cases among those regarding the scintillation crystals characteristics listed in the Introduction may be discussed in light of what we have formulated so far.

In the case of the CRY019 scintillator from Crytur, whose composition is unknown for users, it was only possible to exclude that it is a LuYAP compound. In fact, the value of $p = 0.60$ obtained from Eq (1) hypothesizing such a composition is not comparable with the value of $p = 0.03$ inferred from the trend of mean-free-path for the given 2.1 cm value. In other words, the CRY019 density value is too high for having a mean-free-path value of 2.1 cm, or vice-versa.

In the other case of LuYAP from Kinheng-Crystal, the result from Eq.(1) is $p = 0.71$, that corresponds to a composition of $(Lu_{0.71}Y_{0.29})$AP.

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APPENDIX

Results of interaction data calculations per scintillation crystal in the photon energy range from 100 keV to 1 MeV, calculated at NIST website (US National Institute of Standards, 2021) by using the XCOM software and the XGAM web-database are presented hereafter. XGAM includes the photon cross sections for scattering, photoelectric absorption, and pair production, at energies from 1 keV to 100 GeV. Results are reported hereafter in Tables A1–A7.

Table A1. Values of photons mass interactions coefficients for the YAP scintillation crystal from XCOM calculations performed at NIST website (US National Institute of Standards, 2021).

Photon Energy (keV)	Coherent Scatter. (cm^2/g)	Incoher. Scatter. (cm^2/g)	Photoel. Absorb. (cm^2/g)	Total (cm^2/g)	Photo Fraction	Photoel. Absorb. (1/cm)	Mean Free Path (cm)
100	4.127E-02	1.300E-01	3.931E-01	5.643E-01	6.966E-01	2.107E+00	3.306E-01
150	1.981E-02	1.198E-01	1.180E-01	2.576E-01	4.581E-01	6.325E-01	7.243E-01
200	1.156E-02	1.110E-01	5.036E-02	1.729E-01	2.913E-01	2.699E-01	1.079E+00
300	5.322E-03	9.751E-02	1.558E-02	1.184E-01	1.316E-01	8.351E-02	1.576E+00
400	3.046E-03	8.777E-02	7.023E-03	9.784E-02	7.178E-02	3.764E-02	1.907E+00
500	1.968E-03	8.032E-02	3.905E-03	8.619E-02	4.531E-02	2.093E-02	2.165E+00
600	1.375E-03	7.441E-02	2.479E-03	7.827E-02	3.167E-02	1.329E-02	2.384E+00
800	7.783E-04	6.548E-02	1.272E-03	6.753E-02	1.884E-02	6.818E-03	2.763E+00
1000	4.996E-04	5.893E-02	7.927E-04	6.022E-02	1.316E-02	4.249E-03	3.098E+00

Table A2. Values of photons mass interactions coefficients for the Lu2 scintillation crystal from XCOM calculations performed at NIST website (US National Institute of Standards, 2021).

Photon Energy (keV)	Coherent Scatter. (cm^2/g)	Incoher. Scatter. (cm^2/g)	Photoel. Absorb. (cm^2/g)	Total (cm^2/g)	Photo Fraction	Photoel. Absorb. (1/cm)	Mean Free Path (cm)
100	6.351E-02	1.260E-01	1.012E+00	1.201E+00	8.426E-01	5.991E+00	1.406E-01
150	3.074E-02	1.166E-01	3.275E-01	4.748E-01	6.898E-01	1.939E+00	3.558E-01
200	1.814E-02	1.082E-01	1.468E-01	2.732E-01	5.373E-01	8.691E-01	6.183E-01
300	8.473E-03	9.529E-02	4.833E-02	1.521E-01	3.178E-01	2.861E-01	1.111E+00
400	4.887E-03	8.587E-02	2.267E-02	1.134E-01	1.999E-01	1.342E-01	1.490E+00
500	3.173E-03	7.864E-02	1.293E-02	9.475E-02	1.365E-01	7.655E-02	1.783E+00
600	2.224E-03	7.290E-02	8.354E-03	8.348E-02	1.001E-01	4.946E-02	2.023E+00
800	1.265E-03	6.418E-02	4.367E-03	6.981E-02	6.256E-02	2.585E-02	2.420E+00
1000	8.145E-04	5.777E-02	2.737E-03	6.132E-02	4.463E-02	1.620E-02	2.755E+00

Table A3. Values of photons mass interactions coefficients for the Lu4 scintillation crystal from XCOM calculations performed at NIST website (US National Institute of Standards, 2021).

Photon Energy (keV)	Coherent Scatter. (cm^2/g)	Incoher. Scatter. (cm^2/g)	Photoel. Absorb. (cm^2/g)	Total (cm^2/g)	Photo Fraction	Photoel. Absorb. (1/cm)	Mean Free Path (cm)
100	8.189E-02	1.227E-01	1.523E+00	1.727E+00	8.819E-01	9.893E+00	8.914E-02
150	3.978E-02	1.139E-01	5.006E-01	6.543E-01	7.651E-01	3.252E+00	2.353E-01
200	2.357E-02	1.060E-01	2.265E-01	3.560E-01	6.362E-01	1.471E+00	4.324E-01
300	1.108E-02	9.346E-02	7.540E-02	1.799E-01	4.191E-01	4.898E-01	8.557E-01
400	6.408E-03	8.430E-02	3.559E-02	1.263E-01	2.818E-01	2.312E-01	1.219E+00
500	4.168E-03	7.726E-02	2.040E-02	1.018E-01	2.004E-01	1.325E-01	1.512E+00
600	2.926E-03	7.165E-02	1.321E-02	8.779E-02	1.505E-01	8.581E-02	1.754E+00
800	1.667E-03	6.311E-02	6.925E-03	7.170E-02	9.658E-02	4.498E-02	2.147E+00
1000	1.075E-03	5.681E-02	4.344E-03	6.223E-02	6.981E-02	2.822E-02	2.474E+00

Table A4. Values of photons mass interactions coefficients for the Lu6 scintillation crystal from XCOM calculations performed at NIST website (US National Institute of Standards, 2021).

Photon Energy (keV)	Coherent Scatter. (cm^2/g)	Incoher. Scatter. (cm^2/g)	Photoel. Absorb. (cm^2/g)	Total (cm^2/g)	Photo Fraction	Photoel. Absorb. (1/cm)	Mean Free Path (cm)
100	9.733E-02	1.199E-01	1.952E+00	2.169E+00	9.000E-01	1.379E+01	6.526E-02
150	4.737E-02	1.117E-01	6.461E-01	8.052E-01	8.024E-01	4.565E+00	1.758E-01
200	2.813E-02	1.040E-01	2.935E-01	4.256E-01	6.896E-01	2.074E+00	3.326E-01
300	1.327E-02	9.192E-02	9.814E-02	2.033E-01	4.827E-01	6.934E-01	6.962E-01
400	7.687E-03	8.299E-02	4.645E-02	1.371E-01	3.388E-01	3.282E-01	1.032E+00
500	5.004E-03	7.610E-02	2.667E-02	1.078E-01	2.474E-01	1.884E-01	1.313E+00
600	3.516E-03	7.060E-02	1.729E-02	9.141E-02	1.891E-01	1.222E-01	1.548E+00
800	2.005E-03	6.220E-02	9.074E-03	7.328E-02	1.238E-01	6.411E-02	1.932E+00
1000	1.293E-03	5.601E-02	5.694E-03	6.300E-02	9.038E-02	4.023E-02	2.247E+00

Table A5. Values of photons mass interactions coefficients for the Lu7 scintillation crystal from XCOM calculations performed at NIST website (US National Institute of Standards, 2021).

Photon Energy (keV)	Coherent Scatter. (cm^2/g)	Incoher. Scatter. (cm^2/g)	Photoel. Absorb. (cm^2/g)	Total (cm^2/g)	Photo Fraction	Photoel. Absorb. (1/cm)	Mean Free Path (cm)
100	1.042E-01	1.186E-01	2.142E+00	2.365E+00	9.057E-01	1.555E+01	5.824E-02
150	5.073E-02	1.107E-01	7.105E-01	8.719E-01	8.149E-01	5.158E+00	1.580E-01
200	3.015E-02	1.032E-01	3.231E-01	4.564E-01	7.079E-01	2.346E+00	3.018E-01
300	1.423E-02	9.124E-02	1.082E-01	2.137E-01	5.063E-01	7.855E-01	6.446E-01
400	8.252E-03	8.240E-02	5.126E-02	1.419E-01	3.612E-01	3.721E-01	9.707E-01
500	5.374E-03	7.558E-02	2.944E-02	1.104E-01	2.667E-01	2.137E-01	1.248E+00
600	3.777E-03	7.014E-02	1.909E-02	9.301E-02	2.052E-01	1.386E-01	1.481E+00
800	2.155E-03	6.180E-02	1.002E-02	7.398E-02	1.354E-01	7.275E-02	1.862E+00
1000	1.390E-03	5.566E-02	6.291E-03	6.334E-02	9.932E-02	4.567E-02	2.175E+00

Table A6. Values of photons mass interactions coefficients for the Lu8 scintillation crystal from XCOM calculations performed at NIST website (US National Institute of Standards, 2021).

Photon Energy (keV)	Coherent Scatter. (cm^2/g)	Incoher. Scatter. (cm^2/g)	Photoel. Absorb. (cm^2/g)	Total (cm^2/g)	Photo Fraction	Photoel. Absorb. (1/cm)	Mean Free Path (cm)
100	1.105E-01	1.175E-01	2.318E+00	2.546E+00	9.104E-01	1.785E+01	5.101E-02
150	5.384E-02	1.098E-01	7.701E-01	9.337E-01	8.248E-01	5.930E+00	1.391E-01
200	3.202E-02	1.024E-01	3.505E-01	4.850E-01	7.227E-01	2.699E+00	2.678E-01
300	1.513E-02	9.061E-02	1.175E-01	2.233E-01	5.262E-01	9.048E-01	5.816E-01
400	8.776E-03	8.186E-02	5.571E-02	1.463E-01	3.808E-01	4.290E-01	8.877E-01
500	5.717E-03	7.511E-02	3.201E-02	1.128E-01	2.838E-01	2.465E-01	1.151E+00
600	4.019E-03	6.971E-02	2.077E-02	9.449E-02	2.198E-01	1.599E-01	1.374E+00
800	2.293E-03	6.143E-02	1.090E-02	7.463E-02	1.461E-01	8.393E-02	1.740E+00
1000	1.480E-03	5.533E-02	6.844E-03	6.365E-02	1.075E-01	5.270E-02	2.040E+00

Table A7. Values of photons mass interactions coefficients for the LuAP scintillation crystal from XCOM calculations performed at NIST website (US National Institute of Standards, 2021).

Photon Energy (keV)	Coherent Scatter. (cm^2/g)	Incoher. Scatter. (cm^2/g)	Photoel. Absorb. (cm^2/g)	Total (cm^2/g)	Photo Fraction	Photoel. Absorb. (1/cm)	Mean Free Path (cm)
100	1.218E-01	1.154E-01	2.634E+00	2.871E+00	9.175E-01	2.197E+01	4.176E-02
150	5.942E-02	1.081E-01	8.770E-01	1.045E+00	8.392E-01	7.314E+00	1.147E-01
200	3.537E-02	1.010E-01	3.998E-01	5.361E-01	7.458E-01	3.334E+00	2.237E-01
300	1.674E-02	8.947E-02	1.342E-01	2.404E-01	5.582E-01	1.119E+00	4.988E-01
400	9.715E-03	8.089E-02	6.369E-02	1.543E-01	4.128E-01	5.312E-01	7.771E-01
500	6.331E-03	7.425E-02	3.662E-02	1.172E-01	3.125E-01	3.054E-01	1.023E+00
600	4.452E-03	6.893E-02	2.376E-02	9.715E-02	2.446E-01	1.982E-01	1.234E+00
800	2.542E-03	6.077E-02	1.248E-02	7.580E-02	1.646E-01	1.041E-01	1.582E+00
1000	1.640E-03	5.474E-02	7.837E-03	6.422E-02	1.220E-01	6.536E-02	1.867E+00

