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SOME FACTORS IN X-RADIATION DETECTION WITH THE SCINTILLATION COUNTER

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Some Factors in X-Radiation Detection with the Scintillation Counter

by Dorothy J. Reed*

Résumé

Lorsqu'on utilise un compteur à scintillation pour détecter les lignes K des éléments dont le numéro atomique est de 57 ou plus, le sommet peut jouer un rôle important dans le choix des paramètres à utiliser.

On obtient des plateaux doubles et les courbes d'intensité vibrationnelle deviennent complexes, spécialement pour les radiations de deuxième et troisième ordre.

On a obtenu un déplacement de l'amplitude de vibration avec le taux de mesure en se servant d'un compteur à scintillation utilisant des tubes photomultiplicateurs XP 1010. Ce déplacement rend le choix du seuil de l'analyseur très critique.

Abstract

When the scintillation counter is used to detect the K radiation of elements of atomic number 57 and greater, the escape peak may play an important role in the establishment of the parameters to be used for the measurement of X-ray intensity. Double plateaus are found and pulse amplitude curves become complex, especially for second and third order radiation. A shift in pulse amplitude with counting rate has been found for a scintillation counter using XP1010 photomultiplier tubes. This shift makes the choice of a threshold for the pulse height analyzer critical.

Introduction

The purpose of this paper is to call attention to two aspects of the scintillation counter that have been widely ignored — the effect of escape peaks on pulse distributions and plateaus and the possible occurrence of a pulse shift with counting rate. The latter phenomenon may be associated with the auxiliary counting circuit.

In the Mineral Sciences Division of the Mines Branch the standard Norelco 100kV X-ray spectrograph equipped with a double detector has been used for the past eight years. The pulse height analyzer was modified upon receipt to provide an amplitude range of 150V in addition to the original 60V one.

Counter Resolution

In addition to an increase in pulse amplitude with the energy of the radiation detected, the amplitude is also increased when the amplifier gain or counter voltage is increased.(1) As the amplitude increases the distribution flattens and broadens, but the area under the curve remains the same. For the results reported in this paper an amplifier gain of 10 was used. If the gain was increased above this value, the amplitude of the pulses of a number of the heaviest elements investigated exceeded 150V.

The resolution of the scintillation counter, expressed as the width at half-height (W) over the amplitude (A), was found to improve with atomic number, *i.e.* with the energy of the radiation, for the lighter elements; while for the heavier elements it remained constant. The values obtained at four counter voltages for the Kβ radiation of a number of elements are listed in Table 1. At 950V the A of the heavier elements exceeded the amplitude limit so the values are not complete. The values for the lighter elements at all voltages are in agreement with those of Shard and Bernstein(2) who report that W/A varies as the reciprocal of the square root of the energy for energies in the 10 to 35 keV range.

TABLE 1

Resolution of Kβ Radiation at Different Counter Voltages

Element	Z	keV	800V	850V	900V	950V
Fe	26	7.06			67	63
Zn	30	9.57		56	53	52
Se	34	12.49	50	46	44	43
Sr	38	15 83	56	46	44	37
Mo	42	19.61	40	36	35	34
Sn	50	28.48	36	32	29	27
Te	52	30.99	40	30	28	27
Ba	56	36.37		27	24	
Ce	58	39.25		30	25	24
Nd	60	42.27		26	23	21
Gd	64	48.72	24	23	23	17
Er	68	55.68	25	23	21	
Hf	72	63.38	24	22	20	
Ta	73	65.21	26	23	21	
W	74	67.23	27	25	19	
Pt	78	75.76	32	28		
Hg	80	80.23	31	28		

Escape Peaks

Since the announcement of their occurrence in 1951(3) little attention has been given to the escape peaks of the scintillation counter, at least in the X-ray field.

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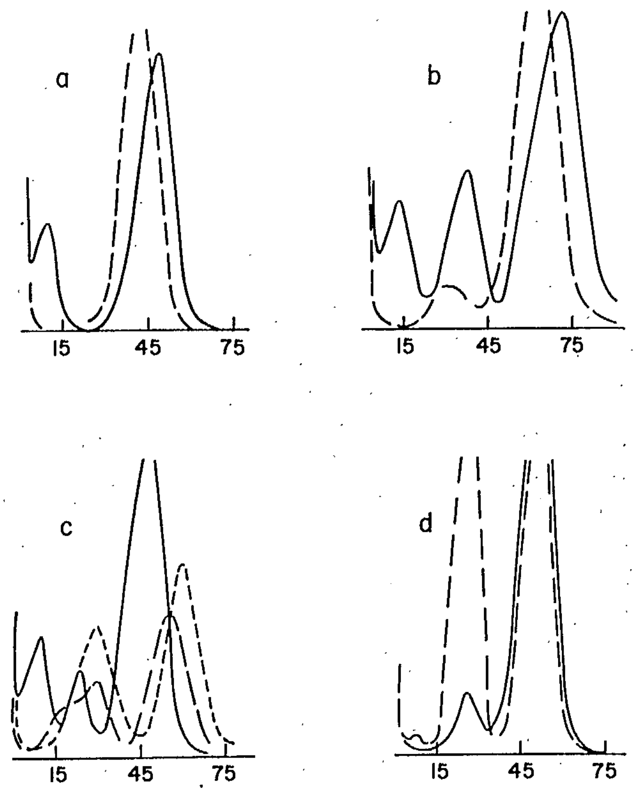

Figure 1 — Pulse Amplitude Distribution Curves [abscissae: amplitude in volts]
a) barium radiation, n=1 —— —— Kα, ———— Kβ
b) barium radiation, n=2: —— —— Kα ———— Kβ
c) rare earths, n=2: ———— NdKα, —— —— GdKβ, — — — — — ErKα
d) TaKα radiation: ———— n=1, —— —— n=2 (x4).

When the radiation striking the NaI crystal of the counter has energy in excess of that of the iodine K absorption edge, iodine atoms may be ionized in their K shell. Part of the entering radiation loses the energy required for this ionization and its residual energy appears as an escape peak in pulse amplitude distribution (PAD) curves. The keV of the iodine edge is 33.16(4) and all elements having characteristic radiation with energy greater than this produce an escape peak in addition to the photopeak in the pulse. BaKα has an energy of 32.19 and BaKβ of 36.37 keV.(5) Therefore the β radiation will produce an escape peak, but the α will not. This is confirmed in the PAD curves for these radiations presented in Figure 1a. The Kβ escape peak occurring at approximately 10V is significant.

Effect on Plateaus:

The occurrence of escape peaks results in a double plateau for the heavy elements. As the counter voltage is increased in plateau determination, the counter first records the more energetic photopeak and then, at a higher voltage, it becomes sensitive to the escape peak. A comparison of the plateaus for barium radiations, Figure 2a, shows a double plateau for the more energetic radiation. Both radiations of lanthanum, the heavier neighbour of barium, have double plateaus, Figure 2b, because both have an energy greater than that of the iodine absorption edge.

For the heavy elements plateaus can sometimes be used as a means of discrimination in place of pulse height analysis. Praesodymium could not be found in the first order in a sample because a much larger amount of neodymium masked it. It was found in the second order as a shoulder on first order ZrKβ, Figure 2d, and was apparently of considerable size. The praesodymium plateau began at 650V, well below that of zirconium. By scanning the sample over the second

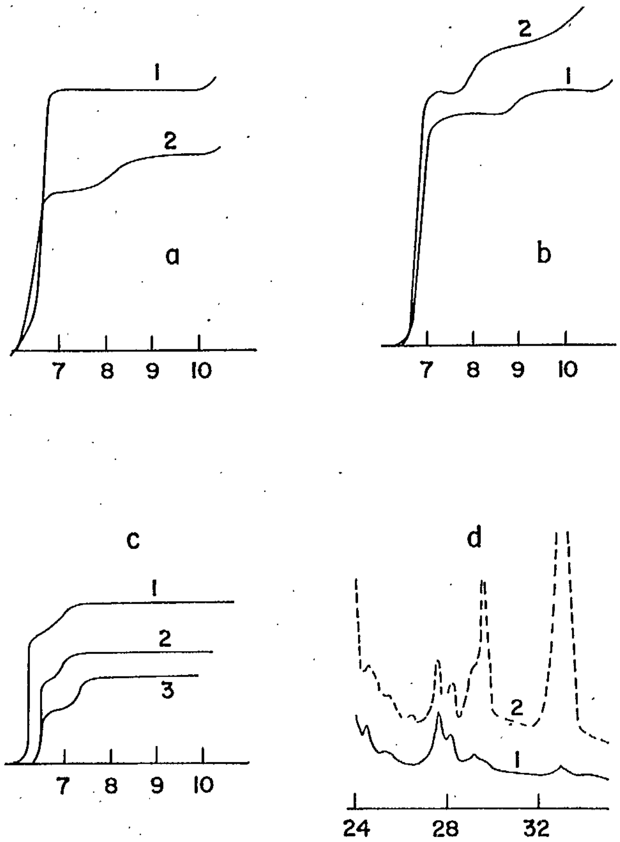

Figure 2 — Plateaus [abscissae: a) b) c) counter voltage —10^{2}V, d) degrees 2θ quartz, 2d=2.74]
a) barium radiation: 1) Kα, 2) Kβ
b) lanthanum radiation: 1) Kα, 2) Kβ (x4)
c) HfKα radiation: 1) n=1 (x0.5), 2) n=2, 3) n=3 (x2)
d) discrimination by plateau difference: 1) 650V, 2) 900 V.

order range using this counter voltage, the true relationship of the two rare earths was seen, as shown by the solid line in Figure 2d.

Pulse Amplitude Interpretation:

When the heavy elements are being determined, escape peaks are one of the factors that affect the interpretation of PADs. As BaKβ illustrated, with the first order radiation simple curves are found. However, when small amounts of a heavy element are to be determined, the use of second order lines may be necessary because the high continuum masks the first order lines. Such was the situation found in determining fractions of a percent of hafnium and tantalum in steels.(6)

The PADs of second order radiations are more complex than first order curves. With barium, Figure 1b, the second order Kα distribution shows a small secondary peak that could be mistaken for an escape peak were energy requirements ignored. This peak is that of the background radiation. The Kβ distribution makes this clear, exhibiting both a background peak at 37V and an escape peak at 15V in addition to the photopeak.

With progression to elements of higher atomic number, the escape peak, as well as the photopeak, occurs at higher energies and is closer to the background peak. Eventually the two have almost the same energy and result in a single peak almost equal in intensity to the photopeak. This is illustrated by the rare earth distributions shown in Figure 1c. NdKα in the second order has distinct escape and background peaks. In the Kα distribution from heavier gadolinium these peaks have begun to merge. In the erbium PAD the two coincide. By the time tantalum is reached, Figure 1d, the combined peak is larger than the photopeak and the background has sufficient energy to produce a small escape peak of its own at 7.5V. The second order curve in this figure has been plotted at four times its original intensity to make its photopeak almost equal in intensity to that of the first order and thus illustrate the contribution of the second order background to the combined peak by comparison with the first order escape peak. In the ores of heavy elements such as tungsten, the photopeak may be dwarfed by the background peak.

The PADs of the Kβ radiation of heavy elements often appear slightly skew because the crystal does not separate the characteristic radiations completely and the contribution of the more intense Kα broadens the Kβ pulse on its low energy side. The perpendiculars dropped from the centres of the hafnium pulses in Figure 3a show a broadening of the Kβ on its low energy side, but not of the Kα on its high energy side. That it is the Kα radiation that causes the broadening is shown by comparing the HfKβ distributions using 220 and 200 LiF crystals (Figure 3b). The better dispersion of the 220 crystal separates the radiations and the Kβ is no longer distorted.

Should it be necessary to use third order radiation of an element, more complex distributions will be found. The PADs for three orders of HfKβ are shown in Figure 3c. The first order radiation exhibits a photopeak and escape peak. The second order has a combined escape and background peak equivalent to the photopeak. The third order, which has been reproduced at twice its original size, has several points of interest. The background peak is much larger than the photopeak and, of course, no longer coincides

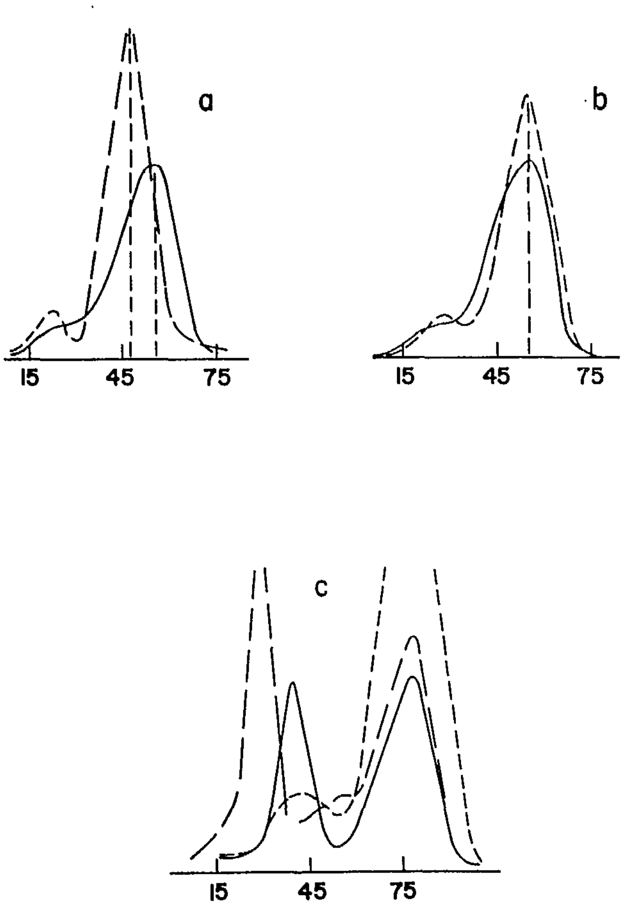

Figure 3 — Hafnium Pulse Amplitude Distributions [abscissae: amplitude in volts]

a) 200 LiF crystal: —— —— K$_\alpha$, ———— ———— Kβ

b) Kβ: ———————— 200 LiF, —— —— 220 LiF

c) Kβ — — — — — — n = 1, ———————— n=2, —— —— n=3.

with the escape peak. The small inflection in the curve might give the impression that the escape peak has shifted to higher energies. However, this is not the escape peak, but the contribution of the λ/2 component of the background. The relationship of the escape to the photopeak in the first order indicates that the former would be so reduced in the third order that it would not show.

Similar curves were obtained for the different orders of PbKα (Figure 4c). In the first order the escape peak is not separated from the photopeak. In the second order the background is intense enough and has sufficient energy to produce an escape peak of its own at 11V, well separated from the combined peak just above 45V. In the third order the λ/2 component is distinct and of the same magnitude as the photopeak, while the background peak has shifted to 30V.

To illustrate how the peaks correspond to the energies of the radiation components, the energies involved in the three orders of lead PADs have been tabulated in Table 2. The photo and background peaks and the λ/2 component with their respective escape peaks are listed. The λ/2 component of the first order cannot be excited by a 100kV generator. In the second order it is, of course, equivalent to the first order radiation. It is interesting to note that the escape peak of the photopeak should be capable of producing an escape peak of its own; but this is not evident in the curves because the escape peak itself is very small. The amplitudes of the peaks have been plotted against these energies in Figure 4b. In order of increasing energy these points represent the background escape peak, the third and second order background peaks, the photopeak escape peak, the λ/2 component and finally the photopeak.

From extrapolation of γ-ray results, it has been variously predicted that the response of amplitude to energy using a scintillation counter would not be linear below 50(7), 100(8) or 150 keV(9). The lead results showed that amplitude was linear with energy for radiation obtained from a single element. Further investigations have shown amplitude to be linear with energy over a wide range of elements when the determinations were made at a constant counting rate. Using the first order Kα radiation of twelve elements from iron (6.4 keV) to hafnium (55.4 keV), a regression line having the equation $Y=0.896226X - 2.4346$ and a correlation coefficient of $+0.9998$ was obtained. Determinations were made on four other elements ending with lead (74.21 keV), but the results obtained at angles less than 6°2θ, which were expected to diverge from linearity because at low angles the geometry of the goniometer is such that the crystal receives some undifferentiated radiation directly from the sample, diverged slightly and were not used to determine the equation. Because a 200 LiF crystal was used, the point of divergence occurred at hafnium radiation.

Plateaus of Higher Order Radiation:

Plateaus showed the presence of escape peaks

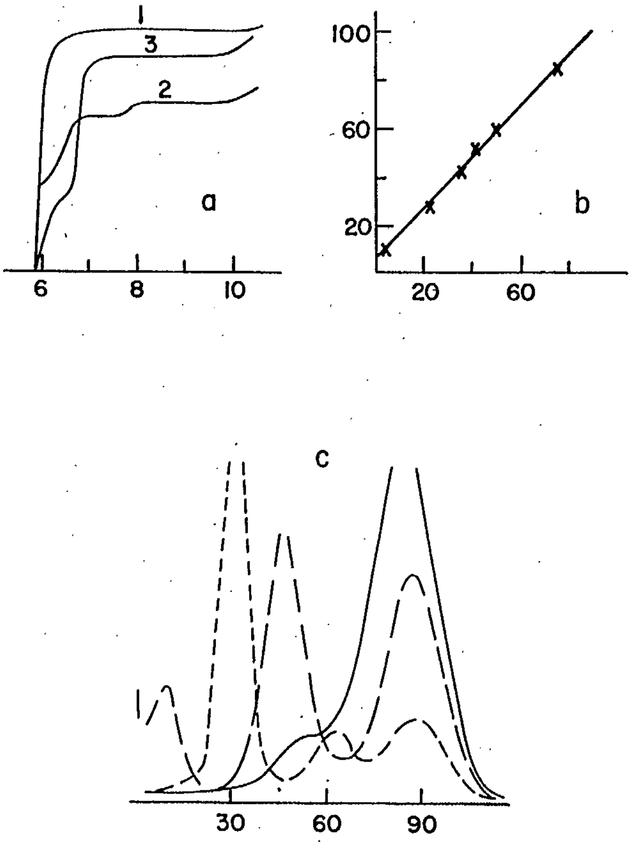

Figure 4 — PbK$_\alpha$ radiation [abscissae: a) counter voltage - 10^{2}V, b) keV, c) amplitude in volts]
a) plateaus: 1) n=1 (x0.1), 2) n=2 (x0.5), 3) n=3
b) peak amplitude vs peak energy
c) pulse amplitude distribution: ——— n=1, —— —— n=2, — — — — — — n=3.

TABLE 2

Energies for PbKα Distribution Peaks: keV

	Photo		Background		λ/2	
Order	Peak	Escape	Peak	Escape	Peak	Escape
1	74.21	41.05	74.21	41.05	(147.90)	(114.74)
2	"	"	37.10	3.99	74.21	41.05
3	"	"	24.70		49.51	16.35

in first order radiation, so it seemed probable that they would show background peaks as well if they were determined using second or third order lines. Plateaus for three orders of HfKα are shown in Figure 2c. In the first order PAD the escape peak is small; in the plateau it is just indicated. The second order distribution has a photopeak and a combined escape and background peak. The plateau has two levels. Similarly the third order plateau has two levels corresponding to the photo and background peaks with the background appearing at a higher counter voltage than in the second order because it is of lower energy.

Lead plateaus correspond to the PADs in a similar way (Figure 4a). The first order shows no escape peak because the peak is not separated from the photopeak in the distribution. The second order plateau shows the photopeaks as an inflection, then the combined background and escape peaks and the much smaller background escape peak as distinct levels. In the third order the photopeak appears as an inflection at the bottom of the curve, followed by a small $\lambda/2$ and a much larger background peak step. Again the lower energy of the third order background appears at a higher voltage than in the second order.

Pulse Shift

The shift in pulse amplitude with counting rate has been well documented for gas-filled counters(10-17) and analysts have been warned about its effect. Little has been reported about a similar shift with the scintillation counter in the X-ray field. Most of the work on scintillation counter response has used Cs^{137} and concerns γ-rays with much higher energies than those of X-rays.(18-24)

In Figure 4c the photopeak for first order lead occurs at a lower voltage than that for the other orders. In the first order the counting rate is much higher than in the second or third order under the same conditions of excitation. This pulse shift to lower amplitudes at higher counting rates has been found to be a property of our scintillation counter which utilizes an XP1010 photomultiplier. It occurs with radiations of all energies. Both a pulse shift and a resolution loss have been experienced as the results in Table 3 show. Different counter voltages were used for the three radiations listed. There is a steady decrease in A as the counting rate increases with the exception of the lowest rate for SnKβ. The resolution is better for the more energetic radiation. These results were obtained by increasing the kV to increase the counting rate, but similar results were obtained using MoKα and increasing the rate by using different concentrations of the element in high temperature alloys and a constant potential.

TABLE 3

Pulse Shift with Counting Rate

Rate	A:volts	W:volts	W/A:%	Rad'n	Counter Voltage
2,000	15.5	10.4	67.1	FeKα	925
7,760	15.4	10.5	68.2		
22,300	15.2	10.8	71.0		
43,100	15.0	11.2	75.0		
66,500	14.7	11.4	77.6		
87,000	14.4	12.0	83.3		
2,500	35.7	11.4	31.9	SnKα	850
5,800	35.5	11.4	32.1		
15,000	35.4	11.4	32.2		
38,300	34.8	11.7	33.6		
53,600	34.5	12.6	36.5		
68,400	34.2	13.5	39.5		
1,600	66.4	18.8	28.3	SnKβ	900
6,200	66.8	18.8	28.1		
12,400	66.4	19.0	28.6		
19,500	65.8	19.9	30.2		
27,500	65.4	20.3	31.0		
34,800	65.1	20.8	32.0		
54,200	64.3	21.4	33.3		
72,800	62.8	22.5	35.8		

Recent results for WKα, using first and second order to secure different counting rates, are given in Table 4. They show the expected increase in A and decrease in W/A with counter voltage and, in addition, show a decrease in A and an increase in W/A with the counting rate. With A being affected by several variables, the value of W/A as a measure of counter resolution is questionable, at least for our counter.

The pulse shift is of small magnitude in some cases and its importance may be questioned. The effects are not negligible as the tellurium counts in Table 5 show. The percentages shown for the discriminated tellurium counts are based upon the counts in the first column using a 6V threshold to eliminate noise and an infinite channel. With the 3V channel there is a loss of 21% of the counts for a rate of 55,000 and 4% for 20,000 cps. With the 9V channel these losses are 23 and 5.6%.

TABLE 4

Effect of Counting Rate on WKα Pulses

Counter Voltage	First Order			Second Order		
	Rate	A	W/A	Rate	A	W/A
750	25,000	8.2	47.6	7750	8.7	41.4
850	26,000	29.7	37.7	7950	30.6	29.6
950	26,500	85.5	31.6	7900	88.5	28.2

TABLE 5

Effect of Pulse Shift on Counts: TeKα (A: 34.5V, W: 6V)

Rate	% counted 33 – 36V	 30 – 39V
2,600	39.7	79.9
5,272	39.3	78.4
8,399	38.8	78.4
12,087	38.3	77.2
14,612	38.0	76.4
19,190	37.3	75.4
24,053	36.7	74.4
27,496	36.0	73.1
34,454	34.7	70.9
44,834	33.0	68.6
55,660	31.2	61.3

This shift may not be a true pulse shift. It may be the result of a baseline shift(25) which is due to the associated electronics rather than the counter itself. For γ-rays it appears to vary with the type of photomultiplier(18,23) and the individual tubes as well as their counting history.(19) The resulting photomultiplier gain has been found to consist of immediate and long-term factors(21) with different recovery times. Similar factors undoubtedly enter into the response of photomultipliers to X-rays. Whatever the origin of the shift, when it occurs it is a practical pulse shift as far as the analyst is concerned and must be compensated for in all non-routine work.

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