

DEPARTMENT OF ENERGY, MINES AND RESOURCES MINES BRANCH OTTAWA

DEAD TIME CORRECTION IN X-RAY SPECTROGRAPHY

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MINERAL SCIENCES DIVISION

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Paragraph 1, lines 4-6 of the text should read: "therefore, correction of counting rates for dead time has not been necessary. In the expectation, etc."

Dead Time Correction in X-Ray Spectrography

by Dorothy J. Reed and A. H. Gillieson*

Résumé

La formule usuelle de correction pour le temps mort d'un compteur s'est avérée insatisfaisante lorsque appliquée aux instruments utilisés dans les laboratoires de la Division des Sciences Minérales de la Direction des Mines. Une nouvelle formule est proposée pour établir les taux de comptage vrais: cette formule a été établie par la technique multifoil pour les longueurs d'ondes variant de CrK α jusqu'à BaK α . Les résultats obtenus par la technique single foil avec deux sources s'accordent assez bien avec ceux obtenus par la technique multifoil, lorsque la nouvelle formule est utilisée pour établir les taux de comptage vrais.

Abstract

The customary dead time correction applied to X-ray counting rates has been found unsatisfactory for the counting equipment used in the Mineral Sciences Division of the Mines Branch

Using a range of wavelengths from CrK_{α} to BaK_{α} and foils of several materials, an amended formulation for calculating dead time and the true counting rate has been developed by the multifoil technique. Results obtained by a single foil and a two source method agree well with the multifoil ones using the amended formula.

In the Mineral Sciences Division of the Mines Branch, samples are usually analyzed by X-ray spectrography using a full range of primary or synthetic standards; therefore, correction of counting losses. The accepted formulae for the sary. In the expectation of undertaking investigations where the true counting rate is required, dead time has recently been studied in our laboratory.

This paper presents our findings using a Norelco 100 Kv constant potential spectrograph equipped with a tungsten tube and a double detector. Of the detector, only the scintillation counter, comprising in part an XP1010 photomultiplier tube, was energized. A Norelco tube type circuit panel was used to register the counts.

Symbols

The following symbols will be used in this paper:

N — true counts

n — observed counts

- r dead time
- p point of deflection
- m number of foils: 0, 1, 2, - m.
- I_{0,1-m} number of counts transmitted by the indicated number of foils.

Background of the Investigation

A common practice for the determination of dead time is to insert successive foils in the path of radiation^(1,2,3) and plot log I_m against m. By extrapolation of the linear portion of the resulting curve, N for I_0 is determined and compared with the corresponding n. The difference between these two numbers is due to dead time counting losses. The accepted formulae for the determination of N and r from the observed counts, n, are: N = n/(1 - nr) and r = (N - n)/Nn. These were established for the Geiger counter using the probability of counts arriving at the counter during its non-responsive or dead time.^(4,6,5)

Although the formulae were derived for the Geiger counter, they have been applied, apparently with little question, to the scintillation and proportional counters, with their much higher counting rates. In 1942, Beers⁽⁷⁾ divided Geigers into two types — those which could not be reexcited during their cycle and those capable of re-excitation - and gave different dead time formulae for the two types. Earlier Skinner⁽⁸⁾ from a mathematical viewpoint considered two types of discharge in Geiger tubes with a different distribution of recovery time for each. Other mathematical derivations of the efficiency of Geiger counters in terms of resolving times have been made.(1,10,11) Of these, Ruark and Brammer considered the contribution of counter, amplifier and recorder to the total efficiency. Blackman and Michiels evaluated efficiency formulae derived by a number of authors.

In 1942, Korff⁽⁴⁾ showed the occurrence of additional pulses within the recovery time of a proportional counter. It was in 1960 that Short⁽¹²⁾ pointed out that if the Geiger correction were used for the proportional counter r increased steadily with n. He presented curves of the correction to be made to n, but offered no new formula. Our work with the scintillation counter has

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confirmed Short's findings as to the variability of r when the Geiger formula is used. Failure of the Geiger formula has been reported by Wittry also.⁽¹³⁾

The first part of this paper deals with our experience with the cumulative multiple foil method and the development of a new correction formula. The latter section reports our experience with two other methods and compares our results using them with those obtained by the multiple foil technique.

Cumulative Foil Method

This multifoil technique was tested using radiation varying from $CrK\alpha$ to $BaK\alpha$ dispersed by a LiF crystal. For such a range of energy, foils of different materials were necessary. Aluminum of 1, 2 and 5 mil and brass of 1 mil thickness were used. The foils were usually placed in front of the crystal because sufficient space was available between the collimator and the crystal, but they were also positioned after the crystal, between it and the detector collimator, in several instances to ensure that foil position had no effect on absorption. Io in excess of 10^5 cps was normally used, but for CrK α this could not be reached because of the low fluorescent yield of the element. In one instance, $SnK\alpha$ Io counts were lowered to see if there was an effect on results. In each series of measurements



Figure 1 Typical Absorption Curves using 0.001" Al Foils.

Io was determined every three or four foils and found to be constant within the counting error. An I_m of approximately 10³ cps was desired, but in some cases the number of foils required to achieve it was too large to be accommodated in the space between the collimator and the crystal.

In all cases experimental curves similar to those in Figure 1 were obtained when $\log I_m$ was plotted against m. The number of foils required before the response became linear depended upon the absorption coefficient of the foil material for the radiation being used and on the foil thickness, as well as I₀. For FeK α using 1 mil aluminum, log I₅ to log I₁₃ gave a straight line while log I₀ to log I₅ deviated from the line with the deviation proportional to m. For MoK α with 1 mil brass, log I₀ to I₂ deviated.

To obtain N, the ratios I_m/I_{m+1} were calculated. At the smaller values of n these ratios were constant, within the limits of counting statistics; at the larger values, they varied. The values giving a constant ratio were used to determine the regression Y = a + bX, where X represented m and Y log I_m . Using this equation, which was that of the extrapolated line and represented the counts that would have been recorded were there no dead time, the corrected log Im's for the foils were calculated and from these the values for N were obtained. Calculation of r from the calculated N's and the observed n's by the Geiger formula gave results typified by those shown in Table 1 for Fe and Sn radiation. In all cases tested, r increased with increasing n.

If the absorption coefficient of the foil used is large for the radiation in question, the point of deflection and the log I_m 's to be used in the calculation of the regression may be observed from the curve. When the absorption coefficient is not large, the point of deflection is not readily as-

TABLE 1

Variation in r with n Using the Geiger Formula

	FeKa				SnKα					
m	N	n	r		N	n	r			
0	152,050	126,460	1.33		158,560	131,960	1.27			
1	100,100	90,620	1.04		111,870	98,080	1.26			
2	67,300	63,210	0.96		78,920	72.260	1.17			
3	44,770	43,140	0.84		55,680	52,680	1.02			
4	29,785	29,290	0.51		38,285	37,930	0.91			
· 5	19,860	18,720			27.715	27,000	010 1			
6	13,185	13,182			19,550	19,230				
7	8,770	8,773			13.800	13.620				
8	5,845	5,850			9,730	9.670				
9	3,880	3,874			6,865	6,900				

n — observed counts

N - counts calculated by linear regression

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certained and the ratios should be used to determine it.

New Formulation

A close look at the curves in Figure 1 and the results in Table 1 shows that below approximately 20,000 cps N and n are equivalent, allowing for the statistical counting error, and no dead time correction is necessary. Correction need be applied only to values of n above the points of deflection or to n-p counts. Substitution of n-p for n in the exponential of the formula for the probability of n counts being recorded for N arriving at the counter gives:

$$n/N = \exp - (n-p)r \tag{1}$$

the solution of this empirical equation yields:

$$N = \frac{n}{1 - (n - p)r}$$
(2)

and

$$\mathbf{r} = \frac{1 - n/N}{n - p} \tag{3}$$

The values of r calculated from the N's and n's above the point of deflection were constant within experimental error.

p may be calculated by assuming a constant r and equating pairs of the right-hand sides of equation (3) using the values of N and n for I_0 with those for I_1 , I_2 , etc., and for I_1 with I_2 , I_3 , etc. This pairing may be carried out for all values of I where the difference between N and n is greater than that due to counting error. For a good estimation of p, several values of I_m above p are necessary because, calculated by equating r, it varies significantly due to counting errors. The mean value calculated from the six combinations of four I_m 's is a good estimate.

Table 2 presents values of r calculated by the Geiger and the amended formula using a variety of radiations and foils. The deflection column in the table shows the calculated p and the m of the foil at which deflection is first evident. The variation in r with n using the Geiger formula is evident as is its constancy with the new one. The mean value of p is 18,800 cps and of r 1.56 μ sec. The low values of p with brass have not been accounted for. There is one high r value — I₃ for Fe, which could be a possible 3 δ count.

The formula using p implies that the first p counts recorded do not affect the passage of the following n-p counts through the detection system. This is, of course, not correct for random particle arrival, though the use of p may be sound mathematically. It is desirable to obtain the correction as a function of n. Then $n/N = \exp - rf(n)$. With two unknowns it is not possible to determine the value of f(n) and r separately from a single equation, but a correction combining the two may be calculated using the approximation:

$$f(n)r = (N - n)/N$$
 (4)

If the calculated values of the combined factor are plotted against the corresponding n, a linear relationship is found. Using those values of n and N that were used for the calculation of results presented in Table 2, f(n)r was found to equal $1.5268 \ge 10^{-6}n = 0.033167$ using linear regres-

TABLE 2

Calculated Dead Times

						r = (1 - n/N)/(n - p)										
	1	1	1			,	Dead Time			L	Deflecti	on	Dead time			
R	Foil	Т	Io	Im	m	CV	Io	I ₁	I ₂	I 3	p	m	Io	I ₁	I ₂	I ₃
Cr Fe	A1 A1	1	81,000 126,000	600 800	8 13	1100 1050	1.38 1.33	$\begin{array}{c} 1.21\\ 1.04 \end{array}$	0.88 0.96	0.84	10,000 25,000	2 3	1.58 1.66	1.57 1.44	1.53 1.59	2.01
Zn Mo	Al Al Brass	2 5 1	106,000 128,000 134,000	500 3500 500	$ \begin{array}{c} 10 \\ 21 \\ 6 \end{array} $	950 900 850	1.28 1.29 1.47	1.02 1.25 1.39	$\begin{array}{c} 0.68\\ 1.16\end{array}$	1.10	25,000 26,000 5,000	2 9 1	$1.68 \\ 1.62 \\ 1.53$	$1.61 \\ 1.64 \\ 1.52$	$1.74 \\ 1.60$	1.63
Sn	Brass Brass Brass*		71,000	1100	12 15	900 850	1.29 1.27	1.22 1.26	$1.08 \\ 1.17 \\ 0.00$	$\begin{array}{c} 1.01 \\ 1.02 \end{array}$	8,600 10,000	35	1.48	1.48	$1.41 \\ 1.42 \\ 1.42$	$1.48 \\ 1.47$
Ba	Brass Brass	1 1	106,000 127,000	1200 1800	26 25	900 900 850	$1.04 \\ 1.32 \\ 1.20$	1.28 1.14	$1.25 \\ 1.08$	$1.15 \\ 1.01$	29,000 21,000 28,000	6 9	1.51 1.64 1.54	1.51 1.67 1.54	1.48 1.70 1.54	$1.68 \\ 1.54$

*Quartz crystal

 $R - K\alpha$ radiation used

T — foil thickness: 0.001 in. CV — counter voltage

TABLE 3

Comparison of Formulas for the Determination of Z

	Ge	iger	Ame	Ratio of N's Caigar/		
n	N	0/ /0	N	%	Amended	
25,000 50,000 75,000 100,000 125,000	26,008 54,200 84,866 118,343 155,039	104.0 108.4 113.2 118.3 124.0	25,126 52,256 81,641 113,573 148,400	100.5 104.5 108.8 113.6 118.7	$1.035 \\ 1.037 \\ 1.040 \\ 1.042 \\ 1.045$	

sion. When f(n)r = 0, and hence N = n, the value of n from this equation is 21,723. Thus, for practical purposes, counts of less than 22,000 obtained on our equipment need not be corrected.

The zero value of f(n)r should be a measure of p and, although it is greater than the mean p in Table 2, it is of the same magnitude as six of the values. It is probably a better estimate of the point of deflection than the mean of the individual determinations because the linear regressions has minimized the individual errors. Using this factor $N = n/(1.033167 - 1.5268 \times 10^{-o}n)$. In Table 3 are listed the N's calculated in this manner and by the Geiger formula for various n's and the amount by which the Geiger formula would overcorrect counts taken on our equipment.

Single Foil Method

For diffractometer corrections, Short suggested the use of a single foil with changes in ma and slit width to vary the intensity. For determinations with the 100 Kv spectrograph, I_0 was varied by changes in Kv or ma or both and counts were taken on the filtered and unfiltered K α beams of a number of metals.

Because it was necessary to have sufficient results to establish the ratios of the two intensities before the value of I_0 was affected by r and because N for I_0 could only be calculated using this ratio from values of I_1 that were likewise unaffected, the choice of foil material for each radiation was critical if even a small number of estimates of r were to be made. It was discovered that it should absorb at least half the intensity of the unfiltered beam. In some cases the 'single' foil used was of two or three thicknesses.

To fulfil the intensity requirements, increments in the power applied were also critical when pure metals were used as the radiation source. Therefore, changes in Kv or ma or both were made.

N was calculated from n using the ratio of I_0 to I_1 determined from a number of paired counts in which all values of I_0 were small enough to be un-

TABLE 4

Single Foil Results for r

R	Geiger Formula	(n – p)a	(n-p)b	1r
Cr Fe Zn Mo Sn	$\begin{array}{c} 0.68 - 1.19 \\ 1.10 - 1.31 \\ 0.80 - 1.18 \\ 0.73 - 1.19 \\ 0.92 - 1.25 \end{array}$	$ \begin{array}{r} 1.62 \\ 1.50 \\ 1.51 \\ 1.38 \\ 1.40 \\ \end{array} $	$ \begin{array}{r} 1.32 \\ 1.57 \\ 1.50 \\ 1.51 \\ 1.56 \\ \end{array} $	1.53 1.62 1.71 1.64 1.71

1

affected by r. The n's used for these calculations were the L's corresponding to the L's too large to be used for the determination of the ratio but which were of themselves too small to be affected by r. The value of r was calculated from these values by the Geiger and the amended formulae. The results are listed in Table 4. Each figure in the last three columns is the mean of four results calculated from four L's. The four results upon which these means were based had an average spread of 0.1. The high and low results from the Geiger formula are given.

The results in the (n-p)a column were calculated using the p calculated from the single foil results by again equating r, those in the (n-p)bcolumn using the mean p from Table 2. The results in the last column made use of the p corresponding to f(n)r = 0. The mean of the results in the last three columns is 1.57μ sec. For four of the five elements the results in the last column are the highest. The mean of the (n-p)a column, in which the results were determined without reference to the multiple foil results, is 1.58 and is in good agreement with the mean of 1.56μ sec. of Table 2.

Two Source Method

The single foil technique was investigated because it was less cumbersome than the multifoil one. The Beers' two source approach was attractive because it involved no change in normal practice.

Beers' two radioactive sources were replaced by two strips of metal foil each positioned to cover approximately half of the irradiation area of the sample holders. Foil A was placed in the holder and counted, foil B was added and the two counted, then A was removed and B counted alone. Care was necessary in inserting B and removing A to ensure that the position of the other foil was not changed. The counting rate on the combined foils, AB, was less than the sum of the counting rates for the two separate foils, A + B. The loss measured the dead time. The shortened Beers' formula "" was used:

$$\mathbf{r} = (\mathbf{A} + \mathbf{B} - \mathbf{AB})/2\mathbf{A}\mathbf{xB}.$$

TABLE 5

Values of r Obtained by Different Methods

	R	Single Foil	Two Source	Cumulative Foil
-	CrKa	1.62		1.56
	FeKα	1.50		1.56
	NiKa		1.80	
	CuKa		1.80	
	ZnKα	1.51		1.68
	PtLα		1.61	
	PbLα		1.70	_
	ΜοΚα	1.38	1.64	1.59
	AgKα		1.72	
	SnKα	1.40		1.47
	BaKα			1.61

The error in positioning the foils in the sample holder and the holder over the X-ray beam resulted in large variations in the values of r. However, the ease of replication made it possible to obtain the mean of a number of values quickly. Values of AB in excess of 50,000 cps gave less varied results because the counting error was less significant at these rates.

The results, means of from six to fourteen determinations of r, are compared in Table 5 with the results of the foil techniques. The mean of all results is 1.60 µ sec. and their standard deviation is \pm 0.12. Comparison with Table 2 shows that even the highest r calculated by the Geiger formula is significantly less than those of the single foil or two source methods.

Conclusions

The choice of a method for the determination of dead time depends on the materials available and the ease of manipulation of the foils in the spectrograph. Any method which uses the ratio of two measurements may be used providing that sufficient data to establish the ratio may be accumulated before r affects one of the measurements.

The two source method is advantageous where foil insertion may be difficult.

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