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Electron penetration and the atomic number correction in electron probe microanalysis[†]

J. PHILIBERT and R. TIXIER

Institut de Recherches de la Sidérurgie Française, 78-Saint-Germain-en-Laye, France

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Abstract. The number of ionizations on a given j shell in any electron-bombarded target has been calculated analytically by means of the Bethe formulae. A general formula for the atomic number correction has been derived in the form of an equation using parameters that are functions of target composition and accelerating voltage. This equation allows the calculation of the penetration effect to be readily carried out. The electron range, in terms of beam energy, has also been calculated. Finally, it is shown that the correction is different in the case of thin-target analysis.

1. Introduction

In electron probe microanalysis an electron beam (3–50 keV in energy) is focused to form a probe of diameter about $1\text{ }\mu\text{m}$ on the surface of an anticathode. The incident electrons ionize the atoms of this target and the x radiation excited is characteristic of the composition of the anticathode, which may either be a pure standard of element A or the specimen in which the concentration of the element A is to be determined.

The wavelength and intensity of the emitted x radiation is measured by means of a proportional counter and crystal spectrometer. Fluorescence and absorption corrections make it possible, knowing the emitted x-ray intensity, to calculate the intensity of the characteristic x rays (for instance $A\text{ K}\alpha_1$ or $L\alpha_1$) generated in the specimen and in the standard respectively.

If we call k_A the ratio of the generated intensities, the so-called ‘first approximation’ (Castaing 1960) is

$$k_A = C_A$$

where C_A is the mass concentration of A in the sample. If the atomic numbers of the elements in the sample are widely different, the first approximation must be corrected by a factor which takes into account two effects:

(i) The back-scattering of electrons by the anticathode, which depends, in addition to other factors, on the atomic numbers and the concentrations of its constituents. This effect reduces the emission, for a given characteristic wavelength, by a factor R . The back-scattering factor R has been measured or calculated by various authors (cf. Green 1962, Archard and Mulvey 1963, Thomas 1964, Bishop 1965, Brown 1965, Derian 1966, Springer 1966, and P. Duncumb 1966, private communication).

(ii) The electron penetration, in terms of beam energy, in the anticathode is also variable and it can be said that, for a given characteristic wavelength, the volume of material analysed is different in the specimen and in the standard. This effect can be characterized by a function $(1/S)$, the determination of which forms the subject of this paper.

Thus we have the following relation for the atomic number correction:

$$\frac{k}{C} = \left(\frac{R}{S}\right)_{\text{specimen}} / \left(\frac{R}{S}\right)_{\text{standard}}.$$

Experimental measurements and theoretical calculations for the R factor seem to be in good agreement, but the S factor is not so well known. Up till now the analytical

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calculation of this factor for a compound specimen has involved a number of rough approximations, though exact numerical solutions have been computed for a few particular alloys. In this paper we shall derive an analytical expression for the S factor which enables its calculation to be readily carried out in all cases..

2. Theoretical background

Let us consider a target containing elements A, B, C, ... of atomic masses A_A, A_B, A_C, \dots , atomic numbers Z_A, Z_B, Z_C, \dots , and mass concentrations C_A, C_B, C_C, \dots , such that

$$\sum_{A, B, \dots} C_A = 1.$$

Let this target be bombarded with electrons of initial energy $E_0 = eV_0$. E_j is the ionization energy of the j shell of atoms A, the element we shall study.

Let E be the mean electron energy, $Q_A^j(E)$ the ionization cross section of the A atoms for the j level, and $dE/d(\rho s)$ the rate of retardation of the electrons in the target, where ρ is the specific mass of the target and s is the length of path traversed by the electrons in the target.

The mean number of ionizations dn_j of the j shell of the A atoms produced by the electrons along the path increment ds is given (Castaing 1960, p. 320) by

$$dn_j = C_A \rho \frac{N}{A} Q_A^j(E) ds$$

and hence

$$n_j = C_A \frac{N}{A} \int_{E_0}^{E_j} \frac{Q_A^j(E)}{dE/d(\rho s)} dE \quad (1)$$

assuming that $dE/d(\rho s)$ is defined and is continuous along the path, and that dn_j may be integrated. N is Avogadro's number. Bethe (1930) and Bethe *et al.* (1938) have given the expressions below for (i) Q and (ii) $dE/d(\rho s)$.

(i) Ionization cross section

$$Q_A^j(E) = \frac{\pi e^4}{E E_j} Z_j b_j \ln \frac{4E}{B_j} \quad (2)$$

where Z_j is the number of electrons on the j shell, and $b_j = 0.35$ for the K shell and 0.25 for the L shell. $B_j = 1.65 E_j$ according to Bethe, but Worthington and Tomlin (1956) gave a better form for low excitation energy as $B_j = 4E_j$. We shall take the value $4E_j$ for electron probe microanalysis, where the electron energy is quite small (< 50 kev) (cf. Green 1962, p. 44); we then have $Q \rightarrow 0$ as $E \rightarrow E_j$.

The b_j value does not appear in the atomic number correction, as will be shown later. If $U = E/E_j$, then

$$Q_A^j(U) = \pi e^4 \frac{Z_j b_j}{E_j^2} \frac{1}{U} \ln U. \quad (3)$$

(ii) Retardation equation

$$\frac{dE}{d(\rho s)} = -\frac{2\pi e^4 N}{UE_j} \sum_{A, B, \dots} C_A \frac{Z_A}{A_A} \ln \frac{1.166 UE_j}{J_A} \quad (4)$$

where J_A is the mean ionization potential for the A atoms.

These equations presuppose that the electron energy is sufficiently high, i.e. $E > E_j$ in equation (2) and $UE_j > J_A$ for all the target atoms in equation (4).

Let

$$\sum_{A, B, \dots} C_A \frac{Z_A}{A_A} = M \quad (5)$$

and

$$\prod_{A, B, \dots} \left(\frac{1 \cdot 166 E_j}{J_A} \right)^{C_A \frac{Z_A}{A_A}} = W^M$$

or

$$\ln W = \frac{1}{M} \sum_{A, B, \dots} C_A \frac{Z_A}{A_A} \ln \frac{1 \cdot 166 E_j}{J_A} \quad (6)$$

which leads to

$$\frac{dE}{d(\rho s)} = -\frac{2\pi e^4 N}{U E_j} M \ln UW \quad (7)$$

and hence, from (1),

$$n_j = C_A \frac{N Z_j b_j}{A} \int_1^{U_0} \frac{\ln U}{M \ln UW} dU$$

where $U_0 = E_0/E_j$ (overvoltage ratio).

Let $1/S = n_j(A/N)(1/C_A)$, where $1/S$ is a function which can be used to represent the electron mass stopping effect in atomic number corrections (according to Philibert 1965, p. 14). Putting $X = UW$, we can integrate $1/S$ in terms of the logarithmic integral (li) function (cf. Nielsen 1906) to obtain

$$\frac{1}{S} = \frac{Z_j b_j}{2N} \frac{1}{M} \left((U_0 - 1) - \frac{1}{W} \ln W (\text{li } U_0 W - \text{li } W) \right). \quad (8)$$

The above equation permits the calculation of $1/S$ in any target. In a target made of a pure element, (8) reduces to an already known equation (Archard and Mulvey 1963).

It is assumed that $U_0 W > 1$. If $W \rightarrow 1$, $1/S$ remains finite, as can be seen from the expression (Nielsen 1906)

$$\text{li } x = C + \ln | \ln x | + \sum_{s=1}^{\infty} \frac{(\ln x)^s}{s s!} \quad (9)$$

where C is Euler's constant ($C = 0.577\,215\,664\,9 \dots$). When $x \rightarrow 1$, $\ln x \rightarrow 0$.

3. A practical method of calculating $1/S$

The calculation may be carried out along the following lines:

(i) For the analysis of an element with an ionization energy E_j in a target of known composition, the terms $W_A^j = 1 \cdot 166 E_j/J_A$ can first be calculated for each of the elements A, B, C, ... in the target.

We have tabulated W_A^j for all elements with Z between 3 and 92, taking the ionization energy E_j for the K and L shells from the tables of Bearden (1964) and Blochin (1957) (see figure 1). We have used $J_A = 11 \cdot 5 Z_A$, which may not be the best form for J . E_j is in units of eV.

(ii) Afterwards one can calculate

$$M = \sum_{A, B, \dots} C_A \frac{Z_A}{A_A}$$

with

$$\sum_{A, B, \dots} C_A = 1, \quad \ln W = \frac{1}{M} \sum_{A, B, \dots} C_A \frac{Z_A}{A_A} \ln W_A^j. \quad (10)$$

The assumption $U_0 W_A^j > 1$ involves a theoretical minimum value for U_0 , though in practical cases its value will always be greater. As a matter of fact a small value of W_A^j is found for the analysis of a light element in a heavy matrix; hence a low E_j and a high U_0 .

(iii) One can then calculate $1/S$ (equation (8)). In practice $Z_j b_j/2N$ need not be calculated since this term vanishes in the correction coefficient. A term $1/S'$ can then be calculated.

We have tabulated

$$\frac{M}{S'} = U_0 - 1 - \frac{1}{W} \ln W (\text{li } U_0 W - \text{li } W) \quad (11)$$

for various values of W and U_0 (see figures 2-4), such that the table of M/S' values can be used for any target. Our tables were established with a computer, using formula (9) for the logarithmic integral sub-routine. This required little computing time. Karpov and Razumovskij (1956) give a very complete table of the logarithmic integral.

(iv) After calculation of $1/S'$ (equation (11)) for the element to be analysed (ionization energy E_j) in the specimen, $1/S'$ can be calculated in the same way for the corresponding standard.

If R is the back-scattering factor (different in the standard and in the specimen) the correction is

$$\frac{k}{C} = \left(\frac{R}{S'} \right)_{\text{sample}} / \left(\frac{R}{S'} \right)_{\text{standard}}. \quad (12)$$

For this calculation the R 's have to be known from measurements or calculations (see §1 (i)).

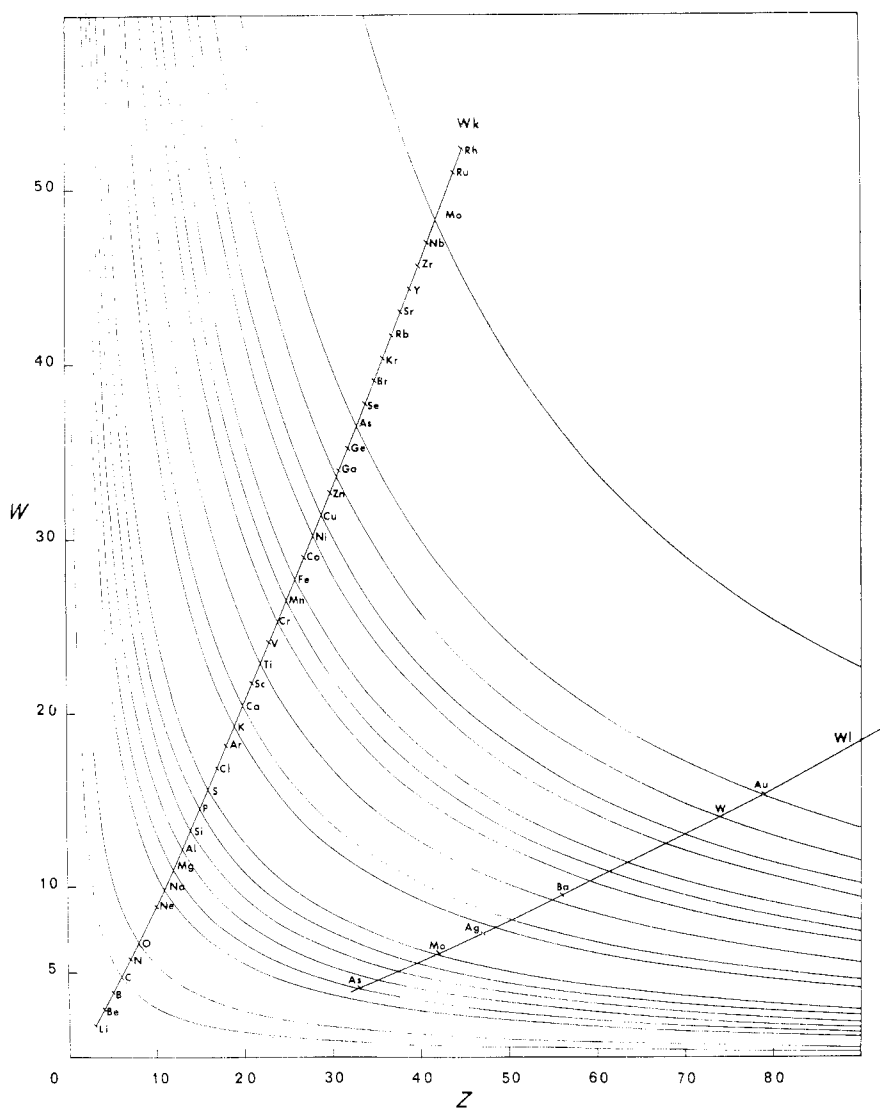


Figure 1. W plotted against Z for various excitation potential values (the W K line corresponds to the excitation of A $K\alpha$ line in pure A).

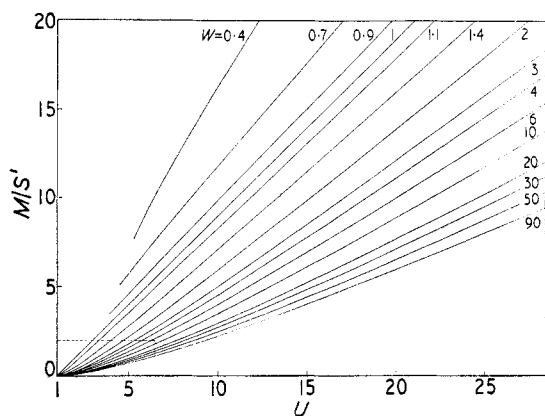


Figure 2. M/S' plotted against U ($1 < U < 30$) for various W values.

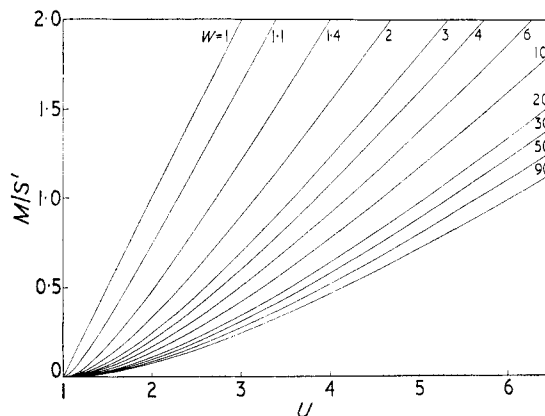


Figure 3. M/S' plotted against U ($1 < U < 6$) for various W values.

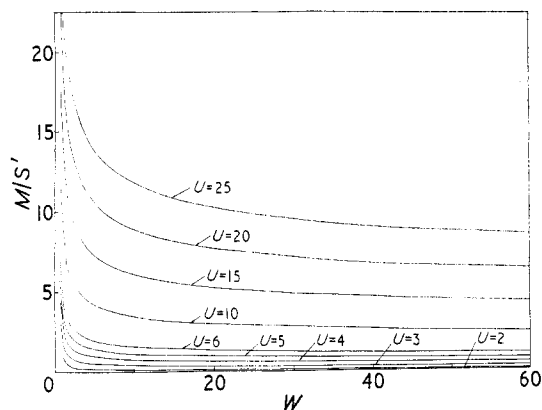


Figure 4. M/S' plotted against W for various U values.

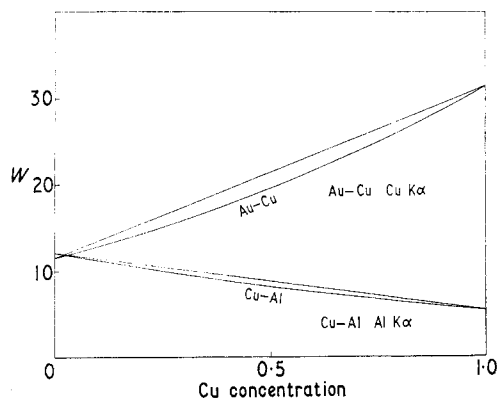


Figure 5. W plotted against Cu concentration (wt. %) in Cu-Au alloy (Cu analysed) and in Cu-Al alloy (Al analysed). A linear approximation has also been traced.

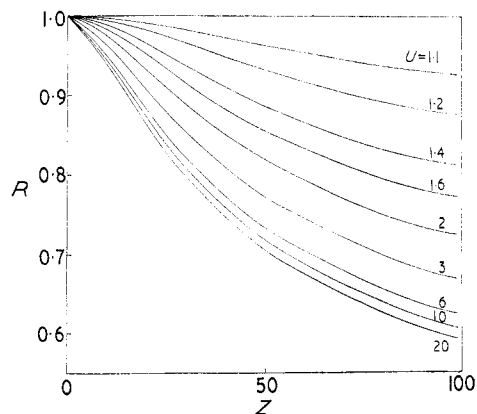


Figure 6. R plotted against Z for various U values according to Duncumb (unpublished).

4. Applications

The 'universal' curves (figures 2-4) give M/S' from U_0 and W values. W for an alloy can be calculated from W_A^j in pure specimens (figure 1), for the element analysed, according to formula (10).

For instance in copper-gold alloys where copper (Cu $K\alpha$) is analysed and in copper-aluminium alloys where aluminium (Al $K\alpha$) is analysed, variations in W have been plotted against copper concentration (figure 5).

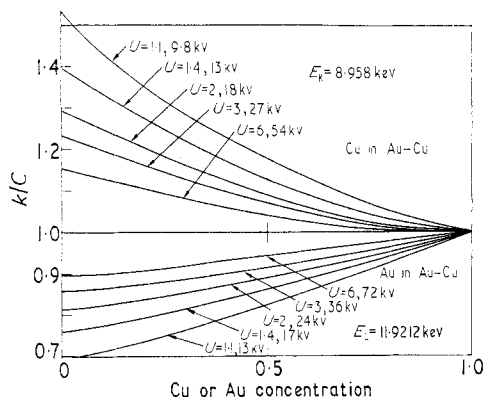


Figure 7. The atomic number correction factor plotted against concentration (wt. %), for Cu-Au system.

In a copper-gold alloy, using values for the back-scattering coefficient R computed by P. Duncumb (1966, private communication) from Bishop's measurements (figure 6) and $1/S'$ values from equation (11), we have calculated the correction coefficients for various accelerating voltages and plotted them against concentration (figure 7).

5. Comparison with other results

Thomas (1964) supposes that one may substitute an average value for the stopping power $dE/d(\rho z)$ in the range E_0 to E_f , and takes this quantity outside the integral (1). Writing $S = dE/d(\rho z)$ (different from our S), she assumes that for a specimen consisting of n elements in concentrations C_1, C_2, \dots, C_n ,

$$\frac{S}{R} = \sum_{i=1}^n C_i \left(\frac{S}{R} \right)_{A_i}.$$

R is calculated from data on the energy distribution of electrons back-scattered from pure-element targets. The values of S used by Thomas were taken from the tabulations by Nelms (1956, 1958) based on the Bethe formula for electron stopping power. The value of S chosen for estimating the correction is that corresponding to an energy half way between E_0 and E_f .

Although Nelms' tables are limited to 10 keV and are not very practical for use under normal conditions of microprobe analysis, it is interesting to compare Thomas's results with ours, since Nelms used for her tables an adjustment for the J variation law, and discussed the validity of calculations using the Bethe formulae. If we consider a 50 : 50 copper-aluminium alloy in which aluminium is analysed with $E_0 = 10$ keV, we find $(1/S)_{\text{spec.}} / (1/S)_{\text{stand.}} = 1.144$ by our method and 1.137 by Thomas's method. The discrepancy is not important in this case. Figure 7 may be compared with that computed by Archard and Mulvey (1963, p. 405). It should also be interesting to compare the k/C values obtained here with others deduced from direct calculation of x-ray production by Monte Carlo methods according to Green (1962) and Bishop (1965).

The analytical forms show that an approximation using a 'mean atomic number' in order to have the same calculation with a mixed target as with pure elements is generally physically unfounded and can lead to important relative errors (cf. Brown 1965, p. 32).

6. Electron range in terms of beam energy

With the same notation,

$$\rho s = \frac{1}{2\pi e^4 N} \int_1^{U_0} E_j^2 \frac{U dU}{M \ln UW}.$$

Putting $X = U^2 W^2$ we can integrate

$$\rho s = \frac{1}{2\pi e^4 N} \frac{E_j^2}{M W^2} (\text{li } U_0^2 W^2 - \text{li } W^2) \quad (13)$$

for the conditions $U_0 W_A^j > 1$. This range calculation, in terms of beam energy, does not allow calculation of the total penetration range. Integration between 0 and U_0 is mathematically possible, but the real difficulty arises from the fact that Bethe's formula should not be used (in this form) from $U=0$ to $U=J/E_j$.

7. Remarks

(i) The representation of the logarithmic integral by the semi-convergent series

$$\text{li } W = \frac{1}{y} \left(1 + \frac{1!}{y} + \frac{2!}{y^2} + \dots \right) \quad (y = \ln W)$$

has frequently been used. This representation is valid only if $|y| \gg 1$. In electron probe microanalysis $\ln W$ is generally less than 7 and is frequently around 1 in cases where the atomic number effect is to be investigated, as in the analysis of light elements in a heavy matrix. For this reason we have used the exact expression (equation (9)).

(ii) The validity of the present calculation is limited, especially in cases of small overvoltage, (a) by the approximation inherent in the Bethe formulae, (b) by the values of the coefficients used, and (c) particularly because straggling has been neglected by taking the mean electron energy as parameter (cf. Cosslett 1964, 1966, Brown 1965). As the atomic number effect is more important when the overvoltage is small, the present model would need to be refined, using for instance the Walske (1952, 1956) calculations to extend the validity of Bethe's formula, or adjusting the coefficient and parameter values by means of experimental measurements (P. Duncumb 1966, private communication).

8. Thin-target analysis

8.1. Theory

If we want the relative concentrations of two elements, A and B, in a thin target, i.e. a target whose thickness is very much less than the range in terms of beam energy (for instance an extraction replica or a thin foil for electron microscopy), the atomic number effect takes a different form.

For such an analysis we use a probe with constant intensity and energy on the same point of the target to measure the apparent concentration $k_A = I_A/I(A)$ of A and that of B, where I_A and $I(A)$ have their usual meanings, and we generally write the result as the ratio k_A/k_B .

If we assume that the effect of back-scattering in the specimen can be neglected (since we are calculating a ratio) in this case, and that the effects of fluorescence and absorption can be neglected or corrected, we have, with the same symbols as above, the following results.

In the specimen

$$dn_j = C_A \frac{N}{A} Q_A^j(E) d(\rho s).$$

In the bulk standard

$$n_j = R_A \frac{N}{A} \int_{E_0}^{E_{jA}} \frac{Q_A^j(E)}{dE/d(\rho s)} dE.$$

R_A is the back-scattering factor of standard A for the overvoltage ratio U_{0A} .

Introducing the Bethe equations, we have

$$dn_j = C_A \frac{N}{A} \frac{\pi e^4}{E_0 E_{jA}} Z_j b_j \ln U_{0A} d(\rho s)$$

$$n_j = R_A \frac{N}{A} \frac{Z_j b_j}{2N} \int_1^{U_{0A}} \frac{\ln U}{M_A \ln U W_A^j} dU$$

with

$$U_{0A} = \frac{E_0}{E_{jA}}, \quad M_A = \frac{Z_A}{A_A}, \quad W_A^j = \frac{1.166 E_{jA}}{J_A}.$$

Hence

$$\frac{1}{S_A'} = \int_1^{U_{0A}} \frac{\ln U}{M_A \ln U W_A^j} dU.$$

As a first approximation we can write

$$k_A = \frac{dn_j}{n_j} = C_A \frac{\pi e^4 2N}{E_0} \frac{\ln U_{0A}}{E_{jA}} \frac{d(\rho s)}{R_A/S_A'}.$$

If we put

$$P_A = \frac{\ln U_{0A}}{E_{jA} R_A/S_A'} \quad (14)$$

then

$$\frac{k_A}{k_B} = \frac{C_A}{C_B} \frac{P_A}{P_B}. \quad (15)$$

If $P_A > P_B$ the measured concentration in A as compared to that of B is greater than the real concentration.

8.2. Discussion

We have used expression (14) above to calculate the correction factor for two experimental measurements.

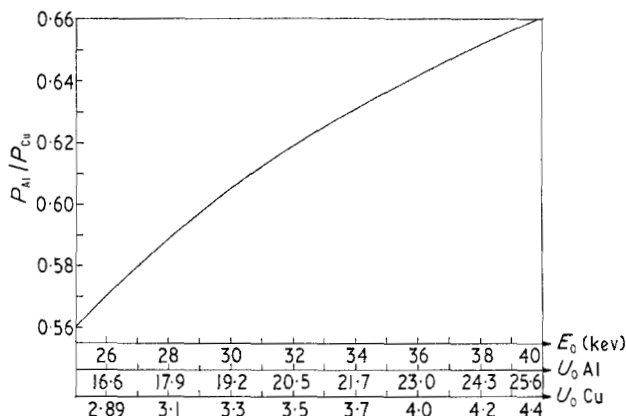


Figure 8. Thin-film analysis, atomic number effect versus voltage, for Al-Cu system.

(i) Analysis of very thin precipitates of Ni+Al in a carbon replica:

$$\frac{P_{Al}}{P_{Ni}} = 0.63 \text{ at } 30 \text{ kev, } 0.55 \text{ at } 20 \text{ kev.}$$

(ii) Analysis of low copper concentrations in a thin aluminium foil (thickness $\sim 3000 \text{ \AA}$). The results are given in figure 8.

In both cases the measured value of the aluminium concentration was less than the true value. The effect is just the opposite of that observed in a bulk target and it is far greater.

If the specimen is not thin enough or if the voltage is not sufficiently high, the effect must lie between those characteristic of a bulk target and of a thin foil. However it seems difficult to calculate the correction, or even to determine whether the factor is greater or less than unity.

8.3. Experimental

The first case studied (Campos Soares *et al.* 1968) concerns an analysis of fine inter-metallic precipitates (diam. $\sim 100 \text{ \AA}$) from a precipitation-hardening austeno-martensite stainless steel (C 0.08, Mn 0.71, Si 0.74, Cr 14.9, Ni 7, Mo 2.14, Al 1.05%). The precipitates were extracted on a carbon replica by means of a controlled potential etch of the δ ferrite.

These precipitates were suspected of being either NiAl or Ni₃Al. Within the error limits of microprobe analysis, the ratio was found to lie near Ni₂Al, but Ni₂Al is not a known compound. Corrections of the type used for massive-target cases would have been small and in the Ni₃Al direction. On the other hand, the corrections we propose for replica analyses lead to a ratio close to NiAl. The result was confirmed by means of electron and x-ray diffraction, in which only lines corresponding to NiAl were observed. The measured values were:

$$\frac{C_{Ni}}{C_{Al}} = 3.27 (\sigma = 1.0) \text{ at } 30 \text{ kv and } 4.25 (\sigma = 0.44) \text{ at } 20 \text{ kv}$$

(σ is the standard deviation).

The corrected values obtained were 1.98 at 30 kv and 2.38 at 20 kv.

The possible theoretical values are: NiAl 2.176, Ni₃Al 6.528, (Ni₂Al 4.352).

The second example is taken from the systematic experimental measurements made on thin films of an Al-4% Cu alloy†, prepared by electropolishing and checked for cleanliness and thickness in the electron microscope.

The measured value of k_{Al}/k_{Cu} varied with the thickness of the film at the point analysed. This thickness can be characterized by the apparent concentration in aluminium and we obtained the following results:

$C_{ap} \text{ Al}$	14.15	64.26	80.14	(25 kv)
k_{Al}/k_{Cu}	16.84	22.16	23.85	

The theoretical value of 13.44 calculated for an infinitely thin film must be a limit of these ratios.

In the case of the bulk target, measurements confirmed the theoretically calculated values (equation (11)).

9. Conclusion

It seems that equations (11) for bulk targets and (15) for very thin targets give quite good corrections for the atomic number effect, in spite of the physical approximations involved. In fact these approximations are not really so drastic as might at first be thought, considering that the corrections are made on the ratio between the specimen and a pure standard.

† This alloy was kindly provided by Pechiney Cie Research Centre.

It must be emphasized that in practice the values of the correction factor are very sensitive to small errors in the calculation of the denominator and the numerator of the ratio. As the logarithmic integral is involved in the calculation, the most accurate results are obtained when the formulae are calculated by a computer. We have not yet investigated whether the convergence and calculation time of the iterative methods at present used for absorption and fluorescence corrections are changed very much by introducing this correction.

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Note added in proof

Since this paper was submitted for publication Dr. P. Duncumb and Miss K. da Casa (now Mrs. Nileshwar) have informed the authors that they have also found the same atomic number effect in thin-specimen analysis. They have made corrections using a numerical method of integration based on a similar physical model.

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