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Experimental Determination of X-ray Compton Scattering from Carbon Blacks and other Non-Crystalline Materials

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A method is described for the experimental determination of the fraction of Compton scattering present in the total X-ray scattering from non-crystalline materials. The Compton scattering from a carbon black has been measured and found to be in good agreement with that predicted by theory.

Introduction

In the X-ray examination of non-crystalline materials it is frequently desired to subtract the Compton (or incoherent) scattering from the total scattering. This is usually done by assuming that the scattering at high values of $(\sin\theta)/\lambda$ is due to incoherent and independent coherent scattering only. It is appreciated that this is often an approximation, and it would be useful if the fraction of Compton scattering actually present in the total scattering could be determined experimentally. A method for so doing has been described by Curien & Deroche (1956). In essence, this consisted of placing in the reflected X-ray beam a filter whose absorption edge lay between the wavelengths of the coherent and the incoherent scattering, thus using the difference in the absorption factors of the filter in the two cases to discriminate between the two wavelengths. It has been found (McKinstry & Short, 1960*a*), however, that the fluorescent radiation emitted by such a foil due to the coherent scattering is of a sufficiently high intensity as to invalidate the general applicability of this procedure. A somewhat similar method will be described which, however, circumvents this objection.

Method

The incoherent X-ray scattering (Compton & Allison, 1935) has a small band of wavelengths, all of which are longer than the wavelength of the primary radiation. The wavelength of the peak of this band is given by (Ross & Kirkpatrick, 1934)

$$\lambda_i = \lambda_c + (2h/mc)\sin^2\theta - D\lambda_c^2\,, \qquad (1)$$

where λ_c is the wavelength of the primary radiation, θ is the Bragg angle, and D is the 'defect shift' coefficient. Because the scattering has a longer wavelength than the coherent radiation, it will be more highly absorbed in any medium through which it passes than will the coherent radiation: if μ_c is the linear absorption coefficient for the coherent radiation, then the corresponding coefficient for the Compton scattering will be given by

$$\mu_i = \mu_c(\lambda_i/\lambda_c)^3. \qquad (2)$$

Strictly, this last expression applies only to the peak of the Compton band. It may be shown (see Appendix), however, that, for the purpose of absorption, the Compton band may be treated as having a single wavelength λ_i.

Consider now a specimen undergoing examination on an X-ray diffractometer to be scattering, at some given value of θ, coherent radiation of intensity I_c and incoherent radiation of intensity I_i. Then the observed intensity I_o will be directly proportional to $I_c + (1-\alpha)I_i$, where α is a correction due to the differential absorption of the Compton scattering in the specimen, the air, and the counter window (McKinstry & Short, 1960*a*):

$$I_o = kI_c + k(1-\alpha)I_i\,.$$

Let a foil of linear absorption coefficient μ_c and thickness t, and whose absorption edge lies below the wavelength of the coherent scattering, be placed in the primary X-ray beam; the new observed intensity will be given by

$$I_o^p = [kI_c + k(1-\alpha)I_i]\,.\exp\,[-\mu_c t]\,.$$

If this same filter is then placed in the secondary X-ray beam, the observed intensity will now be given by

$$I_o^s = kI_c\,.\exp\,[-\mu_c t] + k(1-\alpha)I_i\,.\exp\,[-\mu_i t]\,.$$

From these two equations it can be seen that the fraction of incoherent scattering is given by

$$(1-\alpha)I_i/(I_c+(1-\alpha)I_i)$$
$$= (I_o^p - I_o^s)/I_o^p\,.\{\exp\,[-\mu_c t]/(\exp\,[-\mu_c t] - \exp\,[-\mu_i t])\}\,.$$

We can, therefore, readily obtain the fraction of Compton scattering in the total scattering at any particular value of θ by measuring the intensities of scattering when a filter of accurately known absorption factor (i.e. $\exp\,[-\mu_c t]$) is placed first in the primary

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and then in the secondary X-ray beam, and using equations (1) and (2) to calculate $\exp[-\mu_i t]$.

It will be appreciated that the accuracy of the determination will improve at high values of θ and with short wavelength X-radiation, as these will tend to give the greatest values of $I_o^p - I_o^s$.

Results

The proposed method has been used to determine the fraction of incoherent scattering present in the total radiation scattered by a carbon black between 55° and 160° 2θ, using a General Electric XRD-3 proportional counter diffractometer. Essentially monochromatic copper $K\alpha$ radiation was provided by nickel–cobalt balanced filters placed in the primary X-ray beam (McKinstry & Short, 1960*b*). The absorbing foil was a 0·0007 in. nickel metal foil. Because the number $(I_o^p - I_o^s)$ is small and contains four separate intensity measurements

$$I_o^p - I_o^s = (I_o^p - I_o^s)_{\mathrm{Ni}} - (I_o^p - I_o^s)_{\mathrm{Co}}$$

it was found to be necessary to measure all the intensities with great care; each determination of $(I_o^p - I_o^s)/I_o^p$ occupied approximately one hour, and the total number of counts registered was in the region of 5×10^6.

In the region of a Bragg reflection the fraction of Compton scattering and hence the difference $I_o^p - I_o^s$ will be smaller than elsewhere, and therefore particularly sensitive to errors in the observed intensities. It may be noted that it was found to be impossible to obtain sensible values of the fraction of Compton scattering at the position of the 11 carbon peak.

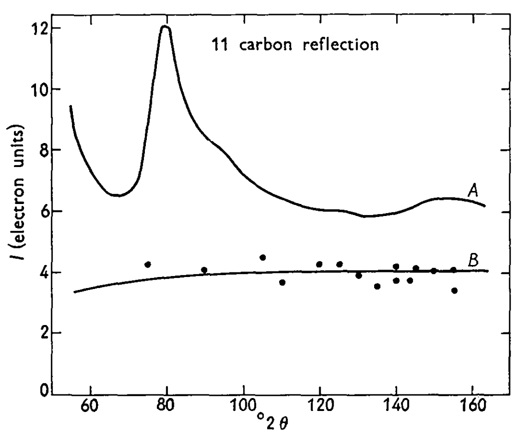

Fig. 1. Curve A shows the total scattering as a function of angle. The points are the experimentally determined intensities of Compton scattering. Curve B is the best line of the required theoretical shape passing through the experimental points.

The results obtained are shown in Fig. 1. Curve A shows the total scattering, I_o, over the range 55° to 160° 2θ, after correction for polarization $(1+\cos^2 2\theta)/2$ and absorption $(1-\exp(-2\mu t \operatorname{cosec}\theta))$. The points shown are the experimentally determined intensities of Compton scattering, given by

$$k(1-\alpha)I_i = (1-\alpha)I_i/(I_c + (1-\alpha)I_i) . I_o .$$

and curve B is the best curve *of the required theoretical shape* fitting these points. The position of this curve was determined in the following way: the experimentally determined intensities of the Compton scattering at any given value of θ should bear a constant ratio to the corresponding value of $(1-\alpha)(Z-\mathscr{F})R$, where Z is the number of electrons in the atom, $\mathscr{F}$ is the scattering function and R is the Breit–Dirac relativistic recoil factor. The experimental intensities were therefore divided by $(1-\alpha)(Z-\mathscr{F})R$, using the values of $\mathscr{F}$ evaluated by Keating & Vineyard (1956); α increased from 0·02 at 60° 2θ to 0·06 at 160° 2θ. The average of the quotients so obtained was then multiplied by the various values of $(1-\alpha)(Z-\mathscr{F})R$ to give the theoretical curve:

$$k(1-\alpha)I_i^{\text{theory}} = (1-\alpha)(Z-\mathscr{F})R \times \langle k(1-\alpha)I_i/(1-\alpha)(Z-\mathscr{F})R\rangle_{\mathrm{av.}} .$$

The total scattering could then be scaled to absolute units, and the Compton scattering subtracted as desired.

Discussion

It has not been possible, unfortunately, to compare directly the results given above with those obtainable from measurements of the scattering at high values of $(\sin\theta)/\lambda$. It has, however, been suggested by the Referee that, from a comparison with results obtained by others using the latter method, the experimental scattering should be equal to the total independent scattering at about 144° 2θ (i.e. midway between a peak and a dip on the experimental curve). On the basis of this assumption, it would be expected that the fraction of Compton scattering present at this angle would be given by

$$(1-\alpha)(Z-\mathscr{F})R/[(1-\alpha)(Z-\mathscr{F})R+f^2] = 0{\cdot}64 ,$$

where f is the coherent atomic scattering factor; experimentally this fraction is found to be 0·65. Such agreement between theory and experiment is better than might be expected, and some comments on the calculations may be of interest.

It is most important to obtain an accurate value of $\exp[-\mu_c t]$, as an error in this factor will introduce a systematic error throughout. It is also important to use as accurate as possible a value of $\exp[-\mu_i t]$, and an alternative calculation was made using the relation derived by Victoreen (1949):

$$\mu_i = C\lambda_i^3 - D\lambda_i^4 + K$$

using tabulated values of μ and λ to obtain the three constants. It was found that this made no significant difference to the results. The omission of the 'defect shift' correction, $D\lambda_c^2$, in the calculation of λ_i does, however, make an appreciable difference: the experimental fraction of Compton scattering at 144° 2θ is reduced to 0·60, compared with a theoretical value of 0·64, and the experimental peak intensity of the 11 reflection is raised to 13·6 e.u.

The Breit–Dirac relativistic recoil factor, R, was introduced for the sake of completeness; many authors prefer to omit this factor, and a separate calculation was therefore made omitting R. The results are:

	With R	Without R
Experimental fraction of Compton scattering at 144° 2θ	0·65	0·66
Theoretical fraction of Compton scattering at 144° 2θ	0·64	0·66
Experimental peak intensity of 11 reflection in electron units	12·2	13·3

APPENDIX

To place any confidence in the method described, it is necessary to show that, for the purpose of absorption, the Compton band may be considered as having a single wavelength λ_i. This may readily be demonstrated in the following manner.

The observed X-ray intensity due to the two wavelengths $\lambda_i \pm \delta\lambda$ after passing through a foil of linear absorption coefficient μ_i and thickness t will be given by

$$I_o = I_{\lambda_i-\delta\lambda}.\exp\left[-\mu_i t(\lambda_i-\delta\lambda)^3/\lambda_i^3\right] + I_{\lambda_i+\delta\lambda}.\exp\left[-\mu_i t(\lambda_i+\delta\lambda)^3/\lambda_i^3\right].$$

Since the Compton band is symmetrical (DuMond & Kirkpatrick, 1931*a*)

$$I_{\lambda_i-\delta\lambda} = I_{\lambda_i+\delta\lambda},$$

and

$$I_o \simeq I_{\lambda_i\pm\delta\lambda}.\exp\left[-\mu_i t\right].(\exp\left[3\delta\lambda\mu_i t/\lambda_i\right] + \exp\left[-3\delta\lambda\mu_i t/\lambda_i\right])$$

$$I_o \simeq I_{\lambda_i\pm\delta\lambda}.\exp\left[-\mu_i t\right].\left[2-9\mu_i^2 t^2(\delta\lambda)^2/\lambda_i^2\right].$$

And if

$$9\mu_i^2 t^2(\delta\lambda)^2/\lambda_i^2 \ll 2$$

then

$$I_o \simeq 2I_{\lambda_i\pm\delta\lambda}.\exp\left[-\mu_i t\right].$$

That is, we can consider the Compton band as having a single wavelength, λ_i, provided that the condition $9\mu_i^2 t^2(\delta\lambda)^2/\lambda_i^2 \ll 2$ is satisfied. If we consider a nickel foil 0·0007 in. thick as an absorber, then this condition reduces, for copper radiation to $\delta\lambda \ll 0{\cdot}87$ Å, and for molybdenum radiation to $\delta\lambda \ll 0{\cdot}40$ Å. DuMond & Kirkpatrick (1931*a*) have shown that the width ($2\delta\lambda_{max.}$) of the Compton band scattered by carbon when bombarded with molybdenum radiation increases from 0·02 Å at 63° 2θ to 0·03 Å at 156° 2θ. They also show (DuMond & Kirkpatrick, 1931*b*) that the width is proportional to the primary wavelength. The required condition is thus satisfied.

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