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X-ray Attenuation Coefficients of Carbon in the Energy Range 5 to 20 keV

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Abstract

Mass attenuation coefficients for pyrolytic graphite have been determined by an X-ray energy-dispersive method with a relative experimental error of about 0·5%. Results are given for the Fe, Co, Cu and Mo $K\alpha_1$ and $K\beta_1$ lines.

1. Introduction

Discrepancies between published X-ray attenuation coefficients have caused the Commission on Crystallographic Apparatus of the International Union of Crystallography to inaugurate a project aimed at improving the techniques for the measurement of these coefficients. The discrepancies are worst for atoms of biological interest, such as hydrogen, carbon, oxygen and calcium (Creagh, 1979, 1981).

The present work describes the determination of mass attenuation coefficients for carbon using an energy-dispersive method. Particular attention has been paid to scattering contributions. As the results show it is likely that cooperative scattering processes, such as small-angle scattering and Laue–Bragg scattering, can explain some of the observed deficiencies in the available data.

The experimental results are compared with theoretical data based on relativistic photoeffect calculations by Scofield (1973), Cromer & Liberman (1970*a,b*) and Storm & Israel (1970) and incoherent and coherent scattering cross sections calculated by the author or taken from recent tabulations by Hubbell, Veigele, Briggs, Brown, Cromer & Howerton (1975) and Hubbell & Øverbø (1979).

2. Experimental procedure

The narrow-beam mass attenuation coefficient, μ/ρ, is determined by an exponential law,

$$I = I_0 \exp[-(\mu/p)x],$$

where I and I_0 are the observed intensities with and without the absorber and x is the mass per unit area of the absorber.

The experimental arrangement has been described in detail in previous work on silicon (Gerward & Thuesen, 1977; Gerward, 1981). Slit-collimated X-rays were monochromatized by a perfect silicon crystal using the symmetric 220 or 440 reflection. The diffracted X-rays were recorded by a Si(Li) detector connected to a multichannel pulse-height analyser. The absorber was placed between the X-ray source and the monochromator, with the specimen surface perpendicular to the X-ray beam. The absorber thickness was chosen to give a value of $\ln(I_0/I)$ close to 3, giving optimum attenuation conditions (Rose & Shapiro, 1948; Nordfors, 1960).

Harmonic contamination of the incident beam was completely eliminated by the energy-dispersive system. The fundamental and the harmonic reflections from the monochromator crystal were well separated in energy by the multichannel pulse-height analyser, the energy resolution being about 300 eV.

System dead times were to a large extent eliminated by using an amplifier with a pile-up rejector and recording each spectrum for a certain preset live time. Even so, the measured attenuation coefficient was found to be slightly count-rate dependent. This effect was eliminated by measuring the attenuation coefficient as a function of the count rate I_0 and extrapolating to zero count rate.

The specimens produced for the Attenuation Project have been machined from high-purity pyrolytic graphite. They have a square cross section 15 × 15 mm and a thickness in the range from 0·1 to 2·5 mm. The mass per unit area has been determined to within 0·5% by weighing and by measuring the lateral dimensions, the latter procedure giving the main contribution to the uncertainty.

3. Results and discussion

Pinhole X-ray photographs of the specimens used in the present work do not indicate any crystalline structure. Thus, it is assumed that the coherent scattering contributing to the attenuation coefficient consists of Rayleigh scattering, typical for an assembly of independent atoms.

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Calculated scattering cross sections useful for comparisons with other experimental or theoretical work are summarized in Table 1. Rayleigh-scattering cross sections and bound-electron Compton-scattering cross sections have been interpolated from the tabulation by Hubbell & Øverbø (1979) and Hubbell *et al.* (1975), respectively. Thermal-diffuse-scattering (TDS) cross sections have been calculated in the same way as in the previous work on silicon and copper (Gerward & Thuesen, 1977; Gerward, 1981, 1982) using a single Debye temperature of 420 K (Flubacher, Leadbetter & Morrison, 1960). This is a rough approximation for graphite because of the anisotropic thermal vibrations, as pointed out by Ludsteck (1971). However, it has been considered sufficient in view of the small TDS cross sections.

It is quite possible for graphite to give considerable small-angle scattering. The measured density of the present specimens is about 1·8 g cm^{-3}. This is less than the X-ray density 2·266 g cm^{-3} (*International Tables for X-ray Crystallography*, 1962), indicating the presence of voids. The measured attenuation coefficient for such a material will depend on how much of the small-angle scattering is counted as part of the transmitted beam (Warren, 1949).

Table 1. *Calculated scattering contributions to the mass attenuation coefficient of carbon in units of* cm^2 g^{-1}

CS = Compton scattering, RS = Rayleigh scattering, TDS = thermal diffuse scattering (graphite)

Line	Energy (keV)	$(\mu/\rho)_{CS}$	$(\mu/\rho)_{RS}$	$(\mu/\rho)_{TDS}$
Fe $K\alpha_1$	6·404	0·114	0·271	0·051
Co $K\alpha_1$	6·930	0·118	0·247	0·051
Fe $K\beta_1$	7·058	0·119	0·242	0·051
Co $K\beta_1$	7·649	0·123	0·221	0·052
Cu $K\alpha_1$	8·048	0·125	0·208	0·052
Cu $K\beta_1$	8·905	0·130	0·185	0·053
Mo $K\alpha_1$	17·479	0·156	0·079	0·038
Mo $K\beta_1$	19·608	0·159	0·067	0·034

In accordance with Chipman (1955) the attenuation coefficient has been measured as a function of the angular divergence of the collimating system. Fig. 1 shows clearly the existence of small-angle scattering in the angular range less than about 6′ for Cu $K\alpha_1$ radiation. Each attenuation coefficient reported in the present work has been obtained with a slit position corresponding to the minimum measured attenuation coefficient in order to eliminate any influence of small-angle scattering.

The experimental results are summarized in Table 2. The energies of the characteristic lines have been taken from *International Tables for X-ray Crystallography* (1974). It is seen that the estimated relative experimental error in μ/ρ is about 0· 5% for the Fe, Co and Cu K lines. For the Mo K lines the error is larger, about 0·75%, because the thickness of the available absorbers was less than the optimum value discussed in § 2.

Fig. 2 presents results for 6·5 to 9·5 keV in graphical form. It is seen that a power law is a good description in this range, μ/ρ being proportional to $E^{-3\cdot02}$, where E is the photon energy. For higher energies Compton scattering is more important and the magnitude of the slope in Fig. 2 will decrease.

Table 3 shows some published mass attenuation coefficients of carbon for Cu $K\alpha_1$ radiation. Also included is the value from *International Tables for X-ray Crystallography* (1974). Attenuation coefficients reported for the unresolved $K\alpha$ doublet have been

Table 2. *Mass attenuation coefficients of graphite measured in the present work*

Line	Energy (keV)	μ/ρ (cm^2 g^{-1})
Fe $K\alpha_1$	6·40384	8·731 ± 0·044
Co $K\alpha_1$	6·93032	6·810 ± 0·033
Fe $K\beta_1$	7·05798	6·481 ± 0·032
Co $K\beta_1$	7·64943	5·060 ± 0·025
Cu $K\alpha_1$	8·04778	4·344 ± 0·021
Cu $K\beta_1$	8·90529	3·221 ± 0·015
Mo $K\alpha_1$	17·47934	0·562 ± 0·004
Mo $K\beta_1$	19·6083	0·450 ± 0·003

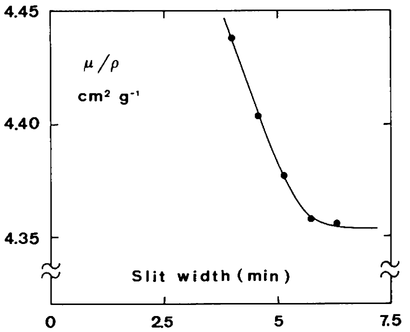

Fig. 1. Mass attenuation coefficient of graphite for Cu $K\alpha_1$ radiation as a function of the slit width expressed in minutes.

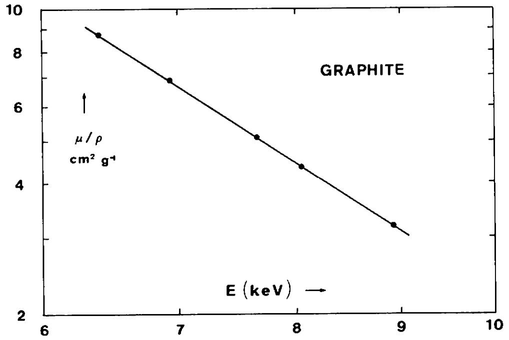

Fig. 2. Log–log plot of the experimental mass attenuation coefficients of graphite as a function of photon energy.

Table 3. *Published mass attenuation coefficients of carbon for* Cu $K\alpha_1$ *radiation*

Observer	μ/ρ (cm^2 g^{-1})	
Andrews (1938)	4·87	
Chipman (1955)	4·15	
Ergun & Tiensuu (1958)	5·06	4·49
Batterman (1958	4·30	
Bearden (1966)	4·33	
Millar & Greening (1974)	4·33	
Calvert, Killean & Mathieson (1975)	4·08	
Berry & Lawrence (1979)	4·12	
International Tables for X-ray Crystallography (1974)	4·21	
Present work	4·34	

corrected using the power law mentioned above. As mentioned in the *Introduction*, there is considerable scatter in the published values. The result of the present work is in very good agreement with those of Batterman (1958), Bearden (1966) and Millar & Greening (1974). In all these works precautions have been taken to ensure freedom from cooperative scattering effects so that the measured values of μ/ρ are characteristic of atomic absorbers.

It is probable that Andrews's (1938) large value of 4·87 cm^2 g^{-1} has been influenced by small-angle scattering because it has been obtained with equipment of high resolving power. Also, Ergun & Tiensuu (1958) report large values but in their method μ has been obtained in an indirect way by comparing scattered intensities in reflection and transmission geometry. Thus, their μ/ρ values are not necessarily comparable with the other values given in Table 3.

Chipman's low value of 4·15 cm^2 g^{-1} is more difficult to understand. It may be of importance that he has placed the absorber between the monochromator and the detector. In this way, some of the coherent and incoherent scattering may have reached the detector, giving too high a value for the transmitted intensity I.

The most recent measurements by Calvert, Killean & Mathieson (1975) and Berry & Lawrence (1979) appear to define a downward trend. It is likely, however, that the low values can be explained to a large extent by a reduction of the coherent scattering because of lattice interference. Their measurements have been performed on single-crystal graphite in a geometry avoiding the occurrence of Laue–Bragg scattering. If it can be assumed that the Laue–Bragg scattering has been avoided completely and that the crystal is perfect from an X-ray point of view, then thermal diffuse scattering (TDS) should be the only coherent scattering occurring in the specimen. The incoherent Compton scattering is practically insensitive to the structural state of the specimen and can be assumed to be the same in all investigations.

For Cu $K\alpha_1$ radiation Berry & Lawrence (1979) and Calvert *et al.* (1975) have measured 4·12 and 4·08 cm^2 g^{-1}, respectively (Table 3). In order to compare with the present work subtract 0·05 cm^2 g^{-1} due to TDS and add 0·21 cm^2 g^{-1} due to Rayleigh scattering according to Table 1. The results are 4·28 cm^2 g^{-1} and 4·24 cm^2 g^{-1}, respectively. These are still on the low side but within 1·5% of the value of 4·34 cm^2 g^{-1} obtained in the present work.

Millar & Greening (1974) and Berry & Lawrence (1979) have measured attenuation coefficients of carbon for a large number of X-ray lines allowing a more extensive comparison with the present work, as shown in Table 4. The agreement between the values of the present work and those of Millar & Greening (1974) is remarkably good. The results of Berry & Lawrence (1979) corrected for TDS and Rayleigh scattering are systematically about 1·5% lower than those of the present work, as shown by the values within square brackets.

Total mass attenuation coefficients calculated from relativistic photoelectric absorption cross sections and scattering cross sections (Table 1) are compared with

Table 4. *Comparison with recent measurements of mass attenuation coefficients of carbon*

MG = Millar & Greening (1974), BL = Berry & Lawrence (1974). The values are given in units of cm^2 g^{-1}.

Line	Energy (keV)	This work	MG* (fitted)	BL†
Fe $K\alpha_1$	6·404	8·73 (4)	8·69	
Co $K\alpha_1$	6·930	6·81 (4)	6·82	
Fe $K\beta_1$	7·058	6·48 (3)	6·45	
Co $K\beta_1$	7·649	5·06 (3)	5·05	
Cu $K\alpha_1$	8·048	4·34 (2)	4·33	4·12 (2) [4·28]
Cu $K\beta_1$	8·905	3·22 (2)	3·21	3·06 (2) [3·19]
Mo $K\alpha_1$	17·479	0·562 (4)	0·561	0·513 (4) [0·554]
Mo $K\beta_1$	19·608	0·450 (3)	0·447	0·411 (4) [0·444]

* Using cubic-fit coefficients for ln σ_{tot}/ln E curve fitting given by Millar & Greening (1974). The standard error of the interpolated values is 0·41%.

† Value within square brackets = original value minus TDS coefficient plus Rayleigh-scattering coefficient.

Table 5. *Comparison with theoretical mass attenuation coefficients of carbon calculated according to Scofield* (1973) (*Sc*), *Cromer & Liberman* (1970) (*CL*) *and Storm & Israel* (1970) (*SI*)

The values are given in units of cm^2 g^{-1}

Line	Energy (keV)	This work	Sc*	CL†	SI*
Fe $K\alpha_1$	6·404	8·73 (5)	8·67	8·51	8·80
Co $K\alpha_1$	6·930	6·81 (3)	6·81	6·70	6·82
Fe $K\beta_1$	7·058	6·48 (3)	6·45	6·34	6·44
Co $K\beta_1$	7·649	5·06 (3)	5·06	4·97	4·99
Cu $K\alpha_1$	8·048	4·34 (2)	4·34	4·27	4·26
Cu $K\beta_1$	8·905	3·22 (2)	3·22	3·17	3·16
Mo $K\alpha_1$	17·479	0·562 (4)	0·562	0·558	0·559
Mo $K\beta_1$	19·608	0·450 (3)	0·450	0·448	0·448

* Interpolated total cross section.

† Interpolated photoelectric absorption cross section plus Compton-scattering cross section plus Rayleigh-scattering cross section.

the present results in Table 5. It is somewhat surprising to notice that the values of Cromer & Liberman (1970*a*,*b*) and those of Storm & Israel (1970) differ in the low-energy range because the original calculations of both these references are based on the code of Brysk & Zerby (1968). Apart from this discrepancy the agreement between experiment and theory is good. In particular, the agreement between the present results and those based on Scofield's (1973) calculations is highly satisfactory. Photoelectric cross sections calculated from screened non-relativistic hydrogen-like eigenfunctions have been shown to be in good agreement with experimental and other theoretical data for elements of medium atomic number (Stephenson, 1975). Using the screening constants given by Stephenson (1976) for carbon it is found that the calculated photoelectric cross sections are 8 to 10% lower than those given by Scofield (1973). For example, the total attenuation coefficients for Cu $K\alpha_1$ and Mo $K\alpha_1$ obtained from hydrogen-like theory are 3·96 and 0·536 $cm^2\ g^{-1}$, respectively, which should be compared with the corresponding values in Table 5.

4. Conclusions

Mass attenuation coefficients characteristic for atomic carbon have been measured with a relative experimental error of about 0·5% using an energy-dispersive method. Theoretical attenuation coefficients derived from relativistic photoelectric cross sections are in good agreement with the experimental results.

Furthermore, it has been shown that a meaningful comparison between high-precision attenuation coefficients is possible only when the contributions from scattering processes are analysed in detail. In particular, the coherent scattering is sensitive to the structural state of the absorber and a large part of the remarkable discrepancies between the published attenuation coefficients of carbon are most probably explained by differences in coherent scattering.

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