References

ALKIRE, R. W. & YELON, W. B. (1981). *J. Appl. Cryst.* **14**, 362–369.
BACHMANN, R., KOHLER, H., SCHULZ, H. & WEBER, H. P. (1985). *Acta Cryst.* **A41**, 35–40.
BARTH, U. VON & PEDROZA, A. C. (1985). *Phys. Scr.* **32**, 353–358.
BECKER, P. & COPPENS, P. (1974). *Acta Cryst.* **A30**, 129–147.
BROWN, R. T. (1970). *Phys. Rev. A,* **2**, 614–620.
BUNGE, C. F. (1968). *Phys. Rev.* **168**, 92–103.
CHOU, M. Y., LAM, P. K. & COHEN, M. L. (1983). *Phys. Rev. B,* **28**, 4179–4185.
COPPENS, P., GURU ROW, T. N., LEUNG, P., STEVENS, E. D., BECKER, P. & YANG, Y. W. (1979). *Acta Cryst.* **A35**, 63–72.
COPPENS, P., LEISEROWITZ, L. & RABINOVICH, D. (1965). *Acta Cryst.* **18**, 1035–1038.
DOVESI, R., ANGONOA, G. & CAUSA, M. (1982). *Philos. Mag. B,* **45**, 601–606.
DOVESI, R., PISANI, C., RICCA, F. & ROETTI, C. (1982). *Phys. Rev. B,* **25**, 3731–3739.
DOWNS, J. W., ROSS, F. K. & GIBBS, G. V. (1985). *Acta Cryst.* **B41**, 425–431.

HANSEN, N. K., SCHNEIDER, J. R. & LARSEN, F. K. (1984). *Phys. Rev. B,* **29**, 917–926.
HELMHOLDT, R. B. (1973). Computer program *TDS1/4*. Univ. of Groningen, The Netherlands.
HELMHOLDT, R. B. & VOS, A. (1977). *Acta Cryst.* **A33**, 38–45.
International Tables for X-ray Crystallography (1974). Vol. IV. Birmingham: Kynoch Press. (Present distributor D. Reidel, Dordrecht.)
LARSEN, F. K. & HANSEN, N. K. (1984). *Acta Cryst.* **B40**, 169–179.
LARSEN, F. K., LEHMANN, M. S. & MERISALO, M. (1980). *Acta Cryst.* **A36**, 159–163.
MCMASTER, W. H., KERR DEL GRANDE, N., MALLETT, J. H. & HUBBELL, J. H. (1969). *Compilation of X-ray Cross Sections.* UCRL-50174 (§2, Rev. 1), Lawrence Radiation Laboratory, University of California at Livermore, CA, USA.
PINDOR, A. J., VOSKO, S. H. & UMRIGAR, C. J. (1986). *J. Phys. F,* **16**, 1207–1212.
SCHNEIDER, J. R., HANSEN, N. K. & KRETSCHMER, H. (1981). *Acta Cryst.* **A37**, 711–722.
SCHNEIDER, J. R. & KRETSCHMER, H. (1985). *Naturwissenschaften,* **72**, 249–259.
SMITH, J. F. & ARBOGAST, C. L. (1960). *J. Appl. Phys.* **31**, 99–102.

General Theory of the Effect of Granularity in Powder X-ray Diffraction

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Abstract

The effect of granularity in powder X-ray diffraction is reviewed, and the shortcomings of previously developed models in explaining this effect are described. A new physical model encompassing all aspects of this phenomenon is proposed. Mathematical formulae based on the model are derived, and their limit cases are discussed. Values of the theoretical intensities from the model are compared with results from a specially designed experiment.

Introduction

X-ray diffracted intensities from actual powder samples can be greatly affected compared with the ideal case if the fineness condition $d(|\bar{\mu} - \mu|) < 0.01$ is not fulfilled (Zevin & Zavyalova, 1974), where d is the mean size, μ is the absorption coefficient of the diffracting phase, and $\bar{\mu}$ is the absorption coefficient of the matrix.

It is not always possible to reduce the particle size enough to avoid granularity effects in the diffracted intensities, since many crystalline materials will not survive severe mechanical treatment. In such cases, it is more meaningful in quantitative analysis and structure determination to make mathematical corrections.

The effect of granularity has been studied from the beginnings of X-ray diffractometry. Brindley (1945) gave the first approximation to the problem. His theory, though oversimplified, successfully explained one fundamental aspect of the effect. Wilchinsky (1951) gave a second model which explained the effects of porosity and grain and particle size, although Harrison & Paskin (1964) stated that his statistical treatment of the powder was not rigorous.

These latter authors stated the question from a new point of view. They proposed to explain the effect of granularity in the angular region near 90°. More recently, Shimazu (1967) and Shimazu & Hosoya (1968) returned to Brindley's ideas, considering new aspects not taken into account by him.

None of these models explains all facets of the effect, which, in reality, is a set of effects very closely related to one another (Zevin & Zavyalova, 1974), and no further directly related work was performed in continuance of the cited works.

The effects of granularity depend on the heterogeneity of crystalline powders. An expression for the intensity from ideally homogeneous crystalline

powders was derived by Alexander & Klug (1948). However, in actual powders the following heterogeneities exist and must be taken into account: (a) heterogeneity of the diffracting phase (distribution of crystals); (b) heterogeneity of the crystalline material (presence of cavities or holes); (c) heterogeneity of composition (presence of several phases in unequal concentrations); (d) heterogeneity of the shape and size of particles; and (e) heterogeneity of the distribution of holes. We aim in this work to state a general theory which takes into account the points just listed, and successfully explains the experimental facts.

Statement of the model

Let a powder be composed of crystals of N different phases, randomly oriented and mixed. Let it be pressed like a porous block and located on the focusing plane of a diffractometer.

The incident X-ray beam can be considered to be composed of individual rays. Along the path of any one of them, it goes through or reflects from different particles and holes alternately, and does not interact with several at the same time. This is derived from the shortness of the wavelength compared with the particle size.

The model of equivalent spheres consists of the transmission of an individual ray going through the centres of a row of crystalline spheres and holes, with diameters equal to the path length of X-rays within the actual particles and holes (Fig. 1).

Let $f_0, f_1, \ldots, f_N$ be the distribution functions of the shapes and sizes of particles and holes of the mixed N phases; let $v_1, v_2, \ldots, v_N$ be the volume fractions of the different crystalline phases ($\sum v_i = 1 - v_0$); and let v_0 be the volume fraction of holes in the powder. As a first approximation we shall consider $v_0 = $ constant, but later the existence of a gradient of v_0 in the vertical direction will be assumed.

The distribution functions $f_0, \ldots, f_N$ can be found either experimentally or theoretically. For example, the distribution function of rod-like particles with thickness d and length L is, in a first approximation, $f(D) = [D \log_e (L/d)]^{-1}$, and $f(D) = k(R, d)[1 - (d/D)^2]^{1/2}$ for plate-like particles, where

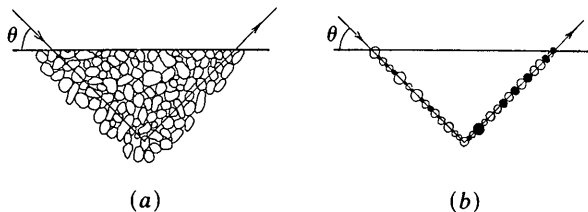(a)   (b)

Fig. 1. Path of a ray through a crystalline powder. (a) Actual particles. (b) Model of equivalent spheres. Open circles for the particles and black dots for the holes denote location and size.

R is the diameter of the plates and $k(R, d)$ is a normalizing parameter.

Derivation of the theoretical formulae

It is known (Guinier, 1956) that the diffracted intensity below the Bragg angle by a volume element dv is

$$dI = I_0 Q \, dv, \tag{1}$$

where I_0 is the incident intensity and Q is the diffracting power. When extinction effects are present, we can write the alternative formula

$$dI = I_0 Q' \, dv, \tag{2}$$

where Q' contains correction terms for primary and secondary extinction.

Supposing that a ray goes through a set of layers arranged parallel to the surface of the powder block, we can find the contribution to the intensity by each layer of particles and integrate to give the total intensity. This contribution is

$$dI_1 = I_0 Q' p_1 v_1 (1 - v_0) \tau A \, dv, \tag{3}$$

where p_1 is the probability of a particle of (diffracting) phase 1 being in the diffracting orientation, v_1 is the probability that a particle struck by the ray is of phase 1, $1 - v_0$ is the probability of finding a hole at the point of incidence, τ is the coefficient of self-absorption (Shimazu, 1967) and A is the absorption factor.

Let us now calculate the absorption factor. For this we shall develop a scheme analogous to that of Berry, Furuta & Rhodes (1969) for studying the effect in the fluorescent case.

We define the X-ray absorption path in a heterogeneous material, by analogy with the optical path, as

$$L_a = \sum_{j=1}^{2n} \mu_j D_j, \tag{4}$$

where μ_j is the linear weakening coefficient of the jth particle and D_j is the diameter of the sphere equivalent to it. The sum extends from the surface to the nth layer and again to the surface along the penetrating and outgoing paths of the rays.

Grouping absorption paths by particles of the same kind, we obtain

$$L_a = \sum_{k=1}^{N} \sum_{j=1}^{n_k} n_k \mu_k D_{k,j}, \tag{5}$$

where the index k refers to phase k.

However, taking into account our knowledge about the distribution of particle sizes, we obtain

$$L_a = \sum_{k=1}^{N} \int_{0}^{\infty} n_k \mu_k f_k D_k \, dD$$

$$= \sum_{k=1}^{N} n_k \mu_k \bar{D}_k \qquad (6)$$

where

$$\bar{D}_k = \int_{0}^{\infty} f_k D_k \, dD. \qquad (7)$$

Now let v_k be the probability (volume fraction) that a particle is of type k. From the binomial distribution we know the probability that n_k of a set of $2n$ particles are of type k, and hence we can estimate the magnitude of the absorption path as

$$L_{a_{2n}} = \sum_{k=1}^{N} \sum_{n_k=0}^{2n} {}_{2n}C_{n_k} v_k^{n_k} (1 - v_k)^{2n - n_k} n_k \mu_k \bar{D}_k. \qquad (8)$$

However, neither μ_k nor $\bar{D}_k$ depend on n_k. The second sum is the mean of n_k for the binomial distribution $2nv_k$. Hence

$$L_{a_{2n}} = \sum_{k=1}^{N} 2nv_k \mu_k \bar{D}_k, \qquad (9)$$

and the absorption factor for a ray reflecting from the $(n+1)$th layer is

$$\exp\left(-\sum_{k=1}^{N} 2nv_k \mu_k \bar{D}_k\right). \qquad (10)$$

The absorption factor in the case of a homogeneous distribution of holes is therefore

$$A = \sum_{n=1}^{\infty} \exp\left(-\sum_{k=1}^{N} 2nv_k \mu_k \bar{D}_k\right) \qquad (11)$$

$$= [\exp(2 \sum v_k \mu_k \bar{D}_k) - 1]^{-1}. \qquad (12)$$

However, according to the model, in which the ray always goes through the centres of equivalent spheres, $dv = D \, dS$, where dS is an element of area of the cross section of the beam, and D is a variable that expresses the diameter of the particles of any type. The total intensity is, from (3) and (12),

$$I_1 = \int_{S} \int_{0}^{\infty} Df \frac{I_0 Q' p_1 v_1 (1 - v_0) \tau}{\exp(2 \sum \mu_k v_k \bar{D}_k) - 1} \, dS \, dD$$

$$= \frac{I_0 Q' p_1 v_1 (1 - v_0) \tau S \bar{D}}{\exp(2 \sum v_k \mu_k \bar{D}_k) - 1} \qquad (13)$$

where

$$\bar{D} = \int_{0}^{\infty} Df \, dD, \qquad (14)$$

f is the overall size distribution and S is the total cross section of the beam.

For a better understanding of the physical sense of the correction with respect to the homogeneous case we can express (13) as

$$I_1 = I_0 Q' p_1 v_1 \tau S F_G / 2\bar{\mu} \qquad (15)$$

where

$$F_G = \frac{2\bar{\mu} \bar{D} (1 - v_0)}{\exp(2 \sum v_k \mu_k \bar{D}_k) - 1}. \qquad (16)$$

The factor F_G is the correction for the effects of granularity. It can be noticed immediately that in the limit $\bar{\mu}\bar{D} \to 0$ and $v_0 = 0$ then $F_G = 1$ and (15) remains in the form derived by Alexander & Klug (1948) for the homogeneous case, apart from the correction τ, which also approaches unity when $\mu_1 \bar{D}_1$ vanishes.

Also, it is easy to show for minute particles in a monophasic powder that

$$F_G \simeq (1 - \mu D / 2)(1 - v_0), \qquad (17)$$

which partially explains the experimental behaviour found by Cooper (1965) and Weiss (1966),

$$F_G = 1 - (\mu D / 2)(1 - v_0)^2. \qquad (18)$$

That is, our model does not explain quantitatively the effect of porosity according to Cooper and Weiss, but it rigorously describes the effect of particle size in the stated case.

On the basis of such a model an expression is derived for the α exponent in the semi-empirical equation of direct X-ray phase analysis (Leroux, Lennox & Kay, 1953):

$$I_1 = I_1^0 C_1 (\mu_1^* / \bar{\mu}^*)^\alpha \qquad (19)$$

where I_1^0 and I_1 are the intensities of a reflection of the diffracting phase (pure and mixed, respectively), C_1 is the weight fraction of the diffracting phase, and μ_1^* and $\bar{\mu}^*$ are the mass absorption coefficients for the diffracting phase and whole sample respectively.

From (15) it follows that

$$I_1 / I_1^0 = v_1 (F_G / F_G^0) \mu_1 / \bar{\mu}$$

$$= C_1 (F_G / F_G^0) \mu_1^* / \bar{\mu}^*, \qquad (20)$$

where $\mu_1^* = \mu_1 / \rho_1$ and $\bar{\mu}^* = \bar{\mu} / \rho$. Here ρ_1 and ρ are the densities of the diffracting phase and whole sample, respectively. Then, from (19) and (20), it follows that

$$\alpha = \log[(F_G / F_G^0) r] / \log r \qquad (21)$$

where $r = \mu_1^* / \bar{\mu}^*$.

Case with a gradient of holes

The development of the theory as far as relation (8) does not require an analysis of the hole distribution through the porous block. However, in an actual

powder specimen variations could occur in the hole density even if some experimental caution were exercised.

Let us suppose that in the porous block a constant positive gradient of the concentration of holes exists in the direction perpendicular to the block surface; then

$$v_0(x) = v_{min} + gx \quad \text{for } 0 \le x \le (v_{max} - v_{min})/g$$

$$v_0(x) = v_{max} \quad \text{for } x > (v_{max} - v_{min})/g. \tag{22}$$

On the other hand, we can put (8) into the form

$$L_{a_{2n}} = na \tag{23}$$

where

$$a = \sum_{k=1}^{N} 2v_k \mu_k \bar{D}_k. \tag{24}$$

The differential absorption of a sheet lying at depth x is

$$dA(x) = (1 - v_{min} - gx) \, d[\exp(-na)]$$

$$\text{for } 0 \le x \le \Delta v/g$$

$$= (1 - v_{max}) \, d[\exp(-na)] \quad \text{for } x > \Delta v/g \tag{25}$$

where

$$\Delta v = v_{max} - v_{min}. \tag{26}$$

In this case it is more convenient to approach the problem in differential form. Thus the transformation $n\bar{D} = x/\sin\theta$ shown in Fig. 2 is to be performed, where

$$\bar{D} = \sum_{k=1}^{N} v_k \bar{D}_k. \tag{27}$$

By integration of (27),

$$A = (1 - v_{min})\exp(-na)$$

$$- g \int_0^{\Delta v/g} x \, d[\exp(-xa/\sin\theta)] \quad \text{for } 0 \le x \le \Delta v/g$$

$$= (1 - v_{max}) \exp(-na) \quad \text{for } x > \Delta v/g. \tag{28}$$

It is possible to find a value $\bar{v}_0$ for which

$$A = (1 - \bar{v}_0) \exp(-na)$$

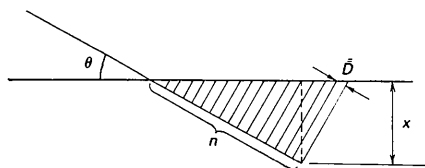$$+ g\bar{D}(\sin\theta/a) \int_0^{Z_{max}} Z \exp(-Z) \, dZ \tag{29}$$

Fig. 2. Geometrical scheme for illustrating the model of equivalent spheres with a gradient of hole concentration.

where

$$Z_{max} = (\Delta v/g)a/\bar{D} \sin\theta. \tag{30}$$

The first term of the sum in (29) leads to a result similar to (13) with $\bar{v}_0$ instead of v_0.

The integral in (29) gives the following result:

$$I_1 = \frac{I_0 Q' p_1 v_1 \tau S}{2\bar{\mu}} \left\{ F_G - \frac{2\bar{\mu}g\bar{D} \sin\theta}{a} \right.$$

$$\left. \times \left[\exp\left(-\frac{a\Delta v}{g\bar{D} \sin\theta}\right)\left(1 + \frac{a\Delta v}{g\bar{D} \sin\theta}\right) + 1 \right] \right\}. \tag{31}$$

Letting $\Delta v/g \gg \bar{D} \sin\theta/a$ in (31), we obtain

$$I_1 \simeq \frac{I_0 Q' p_1 v_1 \tau S}{2\bar{\mu}} \left[F_G - \frac{2\bar{\mu}g\bar{D} \sin\theta}{a} \right], \tag{32}$$

and letting $\Delta v/g \ll \bar{D} \sin\theta/a$, we obtain

$$I_1 \simeq \frac{I_0 Q' p_1 v_1 \tau S}{2\bar{\mu}} \left[F_G - \frac{4\bar{\mu}g\bar{D} \sin\theta}{a} \right]. \tag{33}$$

Therefore a single solution can be considered with

$$I_1 \simeq \frac{I_0 Q' p_1 v_1 \tau S}{2\bar{\mu}} \left[F_G - \frac{2K_g\bar{\mu}g\bar{D} \sin\theta}{a} \right] \tag{34}$$

where K_g is a parameter between 1 and 2 which depends on the value of Z_{max} in (30).

It should be noted in (34) that the effect of the existence of a gradient of hole concentration is revealed in the form of an additive term to the granularity factor. This term will be bigger in absolute value when g grows and when the incidence of X-rays becomes more nearly perpendicular.

Determination or even experimental setting of g could be extremely difficult. Therefore, expression (34) has a merely theoretical importance. It illustrates the effects on intensity if some experimental precautions are not taken for correcting or minimizing the cause of this behaviour.

Experimental

For a practical test of the model an experiment was designed in which the diffracted intensities were measured under rigorously controlled conditions and then compared with the theoretically calculated intensities, given the same conditions.

We prepared mixtures of pure quartz with two different grain sizes: $20 \le d \le 40 \, \mu m$ and $40 < d \le 63 \, \mu m$ with graphite in eight weight fractions between 0·1 and 0·8. The grain size of graphite was $20 \le d \le 40 \, \mu m$.

The mixes were run in a DRON 2,0 diffractometer with Bragg–Brentano focusing and a BSV-23(Cu) tube at 36 kV, 20 mA. Linear slits of 1, 8, 0·25 and 6 mm, and a Soller slit of 2·5° were used.

Selected peaks were scanned point by point over the 20-22°(2θ) and 45-47°(2θ) intervals respectively, with 40 steps of 0·05° length, each for 4 s.

The powder specimens were mounted in a cuvette of quartz and vinyl plastics with rear filling to a constant volume in order to avoid porosity gradients on the diffracting surface. The mixtures were run in a completely randomized order to avoid trends in the results. The theoretical calculations were performed on a NEC 9801 computer with a Basic program.

Results and discussion

Theoretical curves of α vs the logarithm of the absorption rate are plotted in Fig. 3 for six sets of quartz mixtures. From observation of these curves it is possible to make the following qualitative conclusions:

(a) The α exponent tends to be constant in a range around $r = 1$.

(b) The α values decrease with increase of particle size.

These two theoretically-derived behaviours have already been reported in the literature; they form the basis for the development of diffraction–absorption methods.

However, it is noticed from Fig. 3 that there is a discontinuity at $r = 1$. If the α exponent is to have a real physical sense, then curves (a), (c) and (e) should coincide with curves (b), (d) and (f) at a common point for $r = 1$. Hence, α is not a physical constant or parameter, but a simplification aid.

On the other hand, the granularity coefficient, F_G, does have a well defined physical meaning as the weakness of the diffracted intensity generated by the heterogeneity of crystalline powders. The extremum found for $\sum v_k \mu_k \bar{D}_k = 0$ in (16) is not a discontinuity, but a limit case perfectly understandable from the physical point of view, in which the diffracting
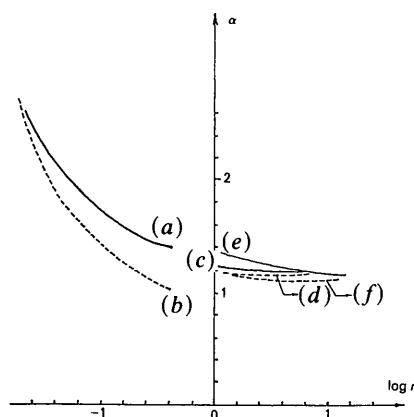
properties of the heterogeneous powders approach those of the ideal homogeneous ones when the particle sizes are diminished.

Fig. 4 illustrates the behaviour of formula (16) for quartz mixed with Fe_2O_3, BeO and graphite, showing continuity along all curves. It may be noticed, too, how the addition of low-absorbing phases enhances the diffraction intensity and *vice versa*, as is already known from the earliest works in quantitative analysis.

For a quantitative test of the proposed model, theoretical curves of $\log[(I_1/I_1^0)C_1]$ {or $\log[(F_G/F_G^0)r]$} vs $\log r$ and the corresponding experimental values were plotted.

Owing to the high dispersion of the experimental results it was not possible to distinguish the values for the intensities of the two different grain sizes and the two angular regions. We then decided to construct a single data set by taking the mean of both sets of values. Finer and more extensive experiments would be necessary to study the contribution of these effects separately.

The theoretical results for quartz–graphite mixtures are plotted in Fig. 5 for two different particle sizes of quartz. It can be seen that the differences between curves (a) and (b) are not too large, and therefore a single curve somewhere between them could be considered without much disadvantage. The experimental results for the same mixtures, obtained by the method described above, are also plotted. The coincidence of theoretical curves with experimental points is rather good for low and medium concentrations, but poor at high graphite concentrations. This lack of fit can be explained by taking into account the fact that theoretical computations from (16) were per-
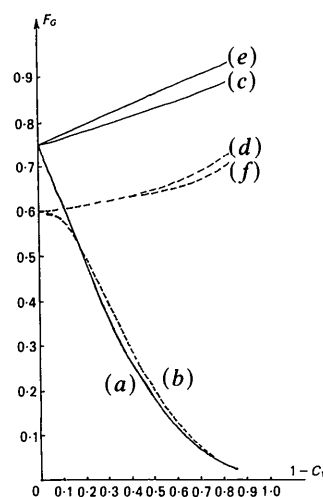

Fig. 3. Variation of microabsorption exponent with log r for (a) quartz (30 μm)-Fe_2O_3, (b) quartz (51·5 μm)-Fe_2O_3, (c) quartz (30 μm)-BeO, (d) quartz (51·5 μm)-BeO, (e) quartz (30 μm)-graphite, (f) quartz (51·5 μm)-graphite.

Fig. 4. Variation of granularity factor with $1 - C_1$ for (a) quartz (30 μm)-Fe_2O_3, (b) quartz (51·5 μm)-Fe_2O_3, (c) quartz (30 μm)-BeO, (d) quartz (51·5 μm)-BeO, (e) quartz (30 μm)-graphite and (f) quartz (51·5 μm)-graphite.

formed on the basis of particle diameters estimated by screening. This method could be rather good for quasi-isometric particles but for the layer-like graphite particles a fourfold longer diameter is to be expected by comparison with the mean particle diameter defined by (7). Therefore, at high graphite concentrations, when layer-like particles become predominant, curves (a) and (b) on Fig. 5 should be 0·2–0·3 units lower on the $\log (I_1/I_1^0 C_1)$ scale, and then would show a better fit to the experimental points. This demonstrates how the diffracte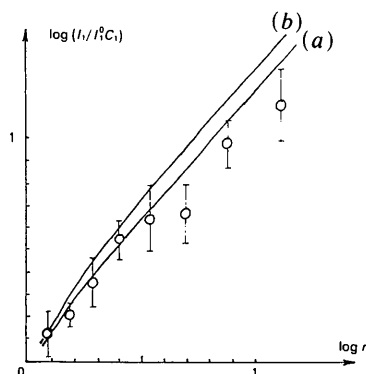d intensities are not only affected by the particle sizes but also by their shapes, and how the theory reflects their influence too.

According to the experimental results for graphite, the model is qualitatively and also quantitatively consistent within certain limits. Also, as was demonstrated in the derivation of the theoretical formulae, this model has as limiting cases the ideal case of Alexander & Klug (1948) and the empirical expressions of Cooper (1965) and Weiss (1966).

For these reasons we estimate that the stated model adequately describes the granularity effects in their most general form.

Fig. 5. Diffracted intensities for quartz, computed from the model for (a) quartz (30 μm)–graphite and (b) quartz (51·5 μm)–graphite. Open circles denote experimental points.

References

ALEXANDER, L. E. & KLUG, H. P. (1948). Anal. Chem. 20, 886–889.
BERRY, P. F., FURUTA, T. & RHODES, J. R. (1969). Adv. X-ray Anal. 12, 612–632.
BRINDLEY, G. W. (1945). Philos. Mag. 36, 347–369.
COOPER, M. J. (1965). Philos. Mag. 11, 969–975.
GUINIER, A. (1956). Théorie et Technique de la Radiocristallographie. Paris: Dunod.
HARRISON, R. J. & PASKIN, A. (1964). Acta Cryst. 17, 325–331.
LEROUX, J., LENNOX, D. H. & KAY, K. (1953). Anal. Chem. 25, 740–743.
SHIMAZU, M. (1967). Mineral. J. 5, 180–198.
SHIMAZU, M. & HOSOYA, S. (1968). Mineral. J. 5, 239–248.
WEISS, R. (1966). X-ray Determination of Electron Distributions. Amsterdam: Elsevier.
WILCHINSKY, Z. W. (1951). Acta Cryst. 4, 1–9.
ZEVIN, L. S. & ZAVYALOVA, L. L. (1974). Kolichestvennyi Rentgenografcheskii Fazovyi Analiz. Moscow: Nedra.

Use of the Polychromatic Laue Method for Short-Exposure X-ray Diffraction Data Acquisition

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Abstract

The polychromatic Laue method was developed for application in flash X-ray diffraction studies; it can also serve as a valuable tool for fast data acquisition with conventional X-ray and synchrotron sources. The present method was developed with a sealed X-ray tube (Mo anode, 45 kV, 20 mA) and a crystal of known structure for calibration of the source spectrum. The resulting experimental energy distribution of the source has been successfully approximated in the range 0·45–1·9 Å by an analytical expression based on a theoretical curve for white radiation. The problem of coincidence of several harmonics in a single diffraction spot has been partially solved by the multiple-film technique. The efficiency of the method was tested on a crystal of (2-tert-butyloxycarbonyl) - 9 - oxo - 6-oxa-1-azabicyclo[5.2.0]nonane, which was also investigated by means of a conventional four-circle X-ray diffractometer. The comparison of about 500 structure factors derived by the two methods yielded an R factor of 0·065. Data acquisition by the Laue method is about two orders of magnitude faster.

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