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X-ray Scattering by Partially Disordered Membrane Systems

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A method of analysing the X-ray diagrams of imperfect membrane lattices with degrees of stacking disorder between 2 and 6% is presented. The method is, however, generally applicable to partially disordered lattices of centrosymmetric membranes. This can also be extended to the case of asymmetric membranes occurring in one orientation only, or in pairs, back-to-back. The procedure allows the determination of the statistical quantities characteristic of the partially disordered lattices and the separation of the diffuse scattering component. As a direct consequence, the refinement of membrane electron density projections becomes feasible and an example is presented for erythrocyte membrane lattices. A one-dimensional model of the refined electron density profile of the erythrocyte membrane is used to calculate numerically the X-ray scattering for a partially disordered lattice of N membrane units. A residual of 15% is obtained with this model and the difference between the observed and calculated values can be minimized further with the proposed refinement procedure.

Introduction

The analysis of X-ray diffraction patterns of membrane lattices has been previously carried out for well orientated membrane systems (Finean & Burge, 1963; Moody, 1963; Worthington & Blaurock, 1968; Caspar & Kirschner, 1971; Levine & Wilkins, 1971; Worthington & Liu, 1973). The degrees of disorder of these systems were less than 2% in the directions of layering of membrane lamellae and, apart from the Lorentz correction and the limited resolution obtainable, the correction of experimental X-ray intensities did not pose further problems. Subsequent model calculations by Weick (1974) and Moody (1975) allowed the estimation of the extent of membrane asymmetry from the corrected intensity profile of membrane dispersions and thus extended the analysis of X-ray data to the case of systems with no positional correlations among the membrane lamellae. Most membrane preparations give lamellar diffraction patterns characteristic of imperfect lattices with degrees of disorder higher than 2% and it therefore becomes important to employ methods developed for the analysis of X-ray data from partially disordered lattices.

An attempt is made here to apply selectively the general calculations, introduced previously for paracrystals, to the interpretation of X-ray diagrams of erythrocyte membrane lattices with degrees of stacking disorder between 2 and 6% (Baianu, 1974, 1978). The approach can be generalized to other membrane systems by taking into account the membrane asymmetry (Moody, 1975).

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One-dimensional models

The total scattering intensity for a paracrystal can be written as

$$I_{\text{tot}} = I_B + I_C, \tag{1}$$

where I_B is the Babinet component and I_C is the crystalline term (Hosemann & Bagchi, 1962; Blundell, 1970). For a lattice made of N stacked lamellae of widths X_j with uniform density in their plane and electron density profiles $R(x_j)$, the two intensity components are

$$I_B(s_1) = 2/(2\pi s_1)^2 \times \text{Re}\{1/\bar{X}[J - P_x G_x F_z/(1 - F_x F_z)]\} \tag{2}$$

$$I_C(s_1) = 2/(2\pi s_1)^2 \times \text{Re}\{P_x G_x F_z[1 - (F_x F_z)^N]/N\bar{X}(1 - F_x F_z)^2\} \tag{3}$$

with the notation:

$$F_x = \int_0^\infty fH(x) \exp(-2\pi i s_1 x)\, dx,$$

$$P_x = \int_0^\infty f^* H(x) \exp(-2\pi i s_1 x)\, dx,$$

$$F_z = \int_0^\infty h(z) \exp(-2\pi i s_1 z)\, dz,$$

$$G_x = \int_0^\infty fH(x)\,dx$$

$$f_j = \int_0 R(x_j)\exp(-2\pi i s_1 x_j)dx_j,$$

$$J = \sum_{j=1}^{N} \int_0^\infty |f_j|^2 H(x)\,dx,$$

where $H(x)$ is the distribution of the widths of lamellae, x_j, and $h(z)$ is the statistical distribution of gaps, z_j, between membrane lamellae (Blundell, 1970).

Numerical calculations of these expressions are currently carried out on a computer (Blundell, 1970) for a specific choice of an average electron density profile, $R(x)$, statistical distribution of membrane widths $H(x)$ and gap distribution $h(z)$. For a step-function model of $R(x)$ these are readily evaluated (Balyuzi & Baianu, 1973).

The electron density projection derived for erythrocyte membranes (Baianu, 1974, 1978) is here used as an example. This profile has been obtained with the method of phase selection introduced by Finean & Burge (1963), and independently by Moody (1963), to membrane structure determination. A step-function model which approximates this profile is presented in Fig. 1. The modulus of the Fourier transform of this model compares favourably with the square root of the corrected intensity profile of the erythrocyte membrane, obtained either from dispersions or from partially disordered membrane lattices (Wilkins, Blaurock & Engelman, 1971; Baianu, 1974). For a one-dimensional, disordered lattice made of N membrane units of scattering amplitudes

$$\begin{aligned} f_j = C\exp[(-2\pi i s_1 x_j)/(2\pi i s_1)]\{1 - \exp(-2\pi i s_1 x_j) \\ + P\exp(-2\pi i s_1 o x_j) - P\exp(-2\pi i s_1 p x_j) \\ + S\exp(-2\pi i s_1 q x_j) - S\exp[-2\pi i s_1(1-q)x_j]\} \end{aligned} \quad (4)$$

the following substitution can be made in (2) and (3)

$$\begin{aligned} P_x G_x = (1 - PF_p + PF_o + SF_q - SF_{1-q} - F_x) \\ \times (F_x - PF^*_{p-1} + PF^*_{o-1} + SF^*_{q-1} - SF_q - 1), \end{aligned} \quad (5)$$

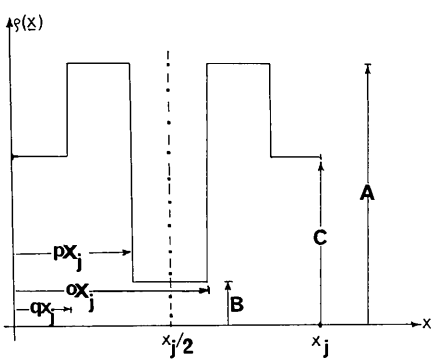

Fig. 1. Step-function model of the average electron density profile of the erythrocyte membrane.

where $P = (A - B)/C$, $S = (A - C)/C$,

$$F_p = \int_0^\infty \exp(-2\pi i s_1 p x)H(x)\,dx,$$

$$F_o = \int_0^\infty \exp(-2\pi i s_1 o x)H(x)\,dx,$$

$$F_{p-1} = \int_0^\infty \exp[-2\pi i s_1(p-1)x]H(x)\,dx,$$

$$F_{o-1} = \int_0^\infty \exp[-2\pi i s_1(o-1)x]H(x)\,dx,$$

$$F_q = \int_0^\infty \exp(-2\pi i s_1 q x)H(x)\,dx,$$

$$F_{q-1} = \int_0^\infty \exp[-2\pi i s_1(q-1)x]H(x)\,dx.$$

I_{tot} and I_B calculated in this manner with the model of Fig. 1 are presented in Fig. 2. The statistical parameters for this calculation were derived from the X-ray diagrams of erythrocyte membranes (Baianu, 1974, 1978) with the methods presented by Hosemann & Bagchi (1962). The residual obtained with the integrated intensities of Fig. 2 is approximately 15% and this provides further support for the use of the swelling method of phase selection [suggested by Bragg & Perutz (1952), and Sayre (1952)] to derive the continuous electron density profile of partially disordered membrane lattices (Baianu, 1974). The values

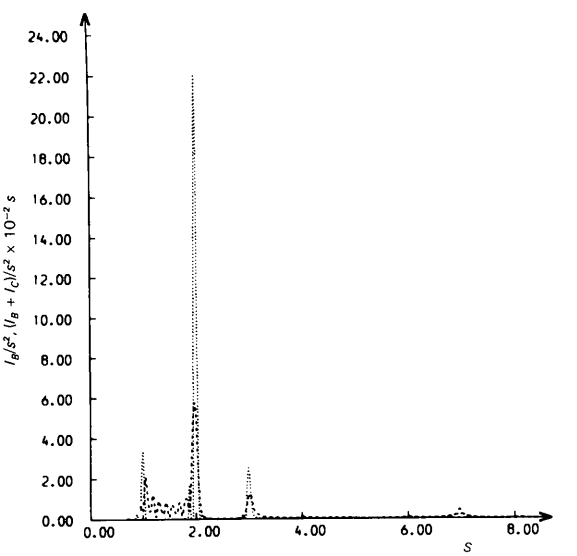

Fig. 2. Calculated scattering intensity of a one-dimensional membrane lattice of N units with profiles as in Fig. 1. ... I_B/s^2, ——— $(I_B + I_C)/s^2$, *versus* $s(k/\bar{d})$ with $p = 0{\cdot}3870$, $o = 0{\cdot}6130$, $q = 0{\cdot}1870$, $P = 0{\cdot}393$, $S = 0{\cdot}22$, $N = 8$, $\Delta x/\bar{x} = 0{\cdot}030$, $\Delta z/\bar{z} = 0{\cdot}000$, $K = 0{\cdot}000$.

estimated for the degree of disorder from the total number of observed reflections (Hosemann & Bagchi, 1962) agree with those derived from the integral widths after correcting for the small crystallite sizes.

Discussion

In view of the absence of a unique solution within the statistical approach, it is preferable to obtain a first approximation to the electron density profile by using the X-ray data uncorrected for the diffuse scattering and then to refine the approximate profile using the model calculations presented above.

The paracrystal approach is particularly useful for removing the diffuse scattering which would otherwise lead to artificial details in the reconstructed electron density profile (Baianu, 1978).

To account for membrane asymmetry (Moody, 1975) additional terms have to be included in (2)–(5). In this case, however, the refinement procedure proposed above cannot be applied unless the asymmetric membrane lamellae are stacked back-to-back as in myelin (to form centrosymmetric units), or an alternative method of phase determination is found.

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Crystallographic Pedigree of Organic Compounds

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A scheme of systematization of organic crystals is proposed on the basis of morphotropism. The organic crystal structures are classified into two main families, *Fm3m* and *Pbca*, whose prototypes are provided by methane and benzene respectively. The system of organic crystals is made up from these prototypes by chemical substitutions frequently accompanied by descent in symmetry of the crystal structures by way of the isotranslational subgroups. The *Pbca* family is predominant in organic molecular crystals. Polymorphism suggests the pedigree relation for descent or ascent in symmetry. The concept of an extended isomorphism is proposed to describe the crystallographic pedigree of organic compounds.

Introduction

Every chemical individual has its own characteristic crystalline form or sometimes forms (polymorphs). To discover the systematic relation between the chemical composition and the crystal structure is a fundamental task in organic crystal chemistry. Previous investigations in this field have dealt almost exclusively with inorganic crystals. This is because organic crystals are not uniformly distributed over all the space groups, but rather are concentrated into a small number of groups. This fact has been attributed by Nowacki (1943) to a zigzag chain structure of dipoles for which the appropriate symmetry element is either a twofold screw axis or a glide plane. Kitaigorodsky (1961) has demonstrated on the grounds of his close-packing