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Evaluation of paracrystalline lattice distortion by profile analysis using a single X-ray diffraction peak

Shigeo Yamamura[a,*], Yasunori Momose[a], Katsuhide Terada[b], Eihei Fukuoka[a]

[a]*School of Pharmaceutical Sciences, Toho University, 2-2-1 Miyama, Funabashi, Chiba 274, Japan*
[b]*Chugai Pharmaceutical Co., Ltd., 41-8, Takada 3-chome, Toshima-ku, Tokyo 171, Japan*

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Abstract

A method of analyzing the paracrystalline lattice distortion and the size of crystallites was investigated by X-ray powder diffraction. This method was based on Fourier analysis of X-ray diffraction peaks and only a single peak was required for the analysis. The observed peak profiles were well described in split pseudo-Voigt function. After correction for the instrumental broadening, the Fourier coefficients for pure diffraction peak profiles were calculated. The size of the crystallites were calculated from the initial slope of the Fourier cosine coefficients at a harmonic number of 0; then the paracrystalline lattice distortion was evaluated from the extinction curve of the Fourier cosine coefficients. Crystalline powders of griseofulvin, tolbutamide and acetazolamide were used for the model drugs. In the griseofulvin powder, the paracrystalline lattice distortion increased and the size of the crystallites decreased with the grinding time. By the single-peak method, the paracrystalline lattice distortion was underestimated while the size of crystallites was overestimated, compared with those obtained by the multiple-peak method reported previously.

Keywords: Paracrystal; Peak profile analysis; Lattice distortion; X-ray powder diffraction; Crystallite; Grinding

1. Introduction

There are several reports which have analyzed the degree of crystallinity of solid-state drugs, because the lattice disorder may affect the stability and bioavailability of drugs (Morita et al., 1985; Oguchi et al., 1989). In a preceding paper, we reported a method for determining the paracrystalline lattice distortion in crystalline pharmaceuticals by means of X-ray powder diffractometry (Fukuoka et al., 1995). By the multiple-peak method reported previously, it was necessary to produce at least three well-resolved peak profiles in different order for the analysis. Since this is not always possible, the multiple-peak method often cannot be used, because there are few pharmaceuticals which produce three different orders of peak profiles such as 100, 200 and 300.

We also reported that the paracrystalline lattice distortion in griseofulvin and dibasic calcium phosphate dihydrate powders increase with grinding, and the lattice distortion was found to affect their pharmaceutical properties (Fukuoka et al., 1995). The paracrystalline lattice distortion in

* Corresponding author

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other pharmaceuticals was considered to affect their physicochemical and pharmaceutical properties.

Because the multiple-peak method is applicable to very limited drugs, new methods, having wider applicability, are desirable to analyze the lattice distortion. These methods would help to understand the relationship between lattice distortion of solid-state drugs and their pharmaceutical properties.

There are several methods for analyzing lattice distortion using a single diffraction peak (Mignot and Rondot, 1977; Zocchi, 1980; Delhez et al., 1982; Nandi et al., 1984). Nandi et al. (1984) reported an initial slope method using Fourier cosine coefficients.

In the present report, we apply the Fourier analysis method to evaluate the paracrystalline lattice distortion in crystalline pharmaceuticals using only one X-ray diffraction peak profile (single-peak method). Changes in lattice distortion and the size of crystallites on grinding of some crystalline pharmaceuticals were investigated. The relationship between the paracrystalline lattice distortion and the molecular arrangement in the crystal was also examined.

2. Materials and methods

2.1. Materials

Griseofulvin (GRIS, Nihon Kayaku Co., Tokyo, Japan) and tolbutamide (TOL, Sigma Chemical Co., MO, USA) were used after recrystallization from acetone, and acetazolamide (AZM, Lederle Japan Co., Tokyo, Japan) was used without purification. Sample powders were used after being passed through a 63 μm sieve. The crystalline structures of GRIS (Malmros et al., 1977), TOL (Donaldson et al., 1981) and AZM (Mathew and Palenik, 1974) have been determined previously.

2.2. X-ray diffractometry (powder method)

A RAD-type diffractometer was used. The X-ray source used was a Cu-Kα line with a voltage of 35 kV and a current of 10 mA. The diffracted X-ray beam was monochromated by a bent-type graphite monochromator. Diffraction intensities were measured by a fixed-time step-scanning method and X-ray polarization was corrected using the equation reported by Yao and Jinno (1982).

2.3. Correction for instrumental broadening

Instrumental broadening based on the slit widths in a goniometer was corrected by the Fourier method (Stokes, 1948). The profile of 100 reflection from a (100)-faced aspirin crystal was used as a standard profile (instrumental broadening profile) which was free of the broadenings of the size of crystallites and lattice distortion (Fukuoka et al., 1995). The effect of diffraction of the $K_{\alpha 2}$ line is removed with the correction for the instrumental broadening. The analytical procedure for analyzing the lattice distortion is discussed below.

2.4. Determination of crystallinity

The degree of crystallinity (X_{cr}) and the lattice disorder parameter (k) were determined by the Ruland method (Ruland, 1961; Fukuoka et al., 1993). A computer program for the determination of crystallinity was developed in our laboratory.

3. Results and discussion

3.1. Determination of peak profile function of discrete Bragg reflection

It is necessary to describe quantitatively the peak profile of the discrete Bragg reflection in the profile analysis. Some analytical expressions have been used for the approximation of the observed peak profile. In the single-peak method, because the peak profiles at a low scattering angle were used, the correction for the peak asymmetry was important to extract the discrete Bragg reflection from the total scattering intensities. We tested the suitability of six functions such as modified Lorentzian (Sonnevert and Visser, 1975), Pearson

Table 1
Comparison of reliability index for the simulation of diffraction peak profile to six different functions

R-value	Modified Lorentzian (ML)	Pearson VII (P-VII)	Pseudo-Voigt (PV)	Split ML	Split P-VII	Split PV
Profiles: 100 Reflection of aspirin powder						
R_p	0.121	0.093	0.084	0.077	0.038	0.032
R_{wp}	0.169	0.151	0.127	0.111	0.085	0.080
Profile: 011 reflection of griseofulvin powder						
R_p	0.220	0.221	0.212	0.094	0.048	0.036
R_{wp}	0.275	0.274	0.262	0.120	0.065	0.059

$R_p = \Sigma|y_{oi} - y_{ci}|/\Sigma y_{oi}$; $R_{wp} = [\Sigma w_i\{y_{oi} - y_{ci}\}^2/\Sigma w_i y_{oi}]^{1/2}$. y_{oi} is the observed intensity, y_{ci} is the simulated intensity and w_i the weight of the data, $1/y_{oi}$.

VII (Immirzi, 1980) and pseudo-Voigt functions (Hecq, 1981) and their split type functions in an effort to improve the fit. Table 1 shows the results in which the observed profiles of the 100 reflection of aspirin and the 011 reflection of griseofulvin powders were simulated by six different peak profile functions. Because the split pseudo-Voigt function gave the best fit with the experimental data among the six functions, the split pseudo-Voigt function (Eq. (1)) was used for the peak profile function in the present investigation.

$$I(2\theta) = K[\eta_L \exp\{-\alpha_L^2(\Delta 2\theta)^2\} + (1-\eta_L)\{1 + \alpha_L^2(\Delta 2\theta)_2\}] \quad (2\theta < 2\theta_{pk})$$

$$I(2\theta) = K[\eta_H \exp\{-\alpha_H^2(\Delta 2\theta)^2\} + (1-\eta_H)\{1 + \alpha_H^2(\Delta 2\theta)^2\}] \quad (2\theta > 2\theta_{pk}) \tag{1}$$

where $I(2\theta)$ is the scattering intensity at 2θ, K is the diffraction intensity at the peak maximum ($2\theta_{pk}$), η is the mixing parameter of Gaussian and Lorentzian functions, α is the peak-width parameter and $\Delta 2\theta$ is the (2θ-$2\theta_{pk}$). The subscripts H and L are used for the range of (2θ-$2\theta_{pk}$) > 0 and (2θ-$2\theta_{pk}$) < 0, respectively.

Fig. 1 shows the experimental data and simulation profile of the 100 reflection of aspirin and the 011 reflection of griseofulvin. Simulation profiles were found to fit very well with observed intensities. As shown in Fig. 1 and Table 1, a split pseudo-Voigt function was good enough to describe the observed X-ray diffraction peak. Young and Wiles (1982) also pointed out that a pseudo-Voigt or Pearson VII functions were useful for the profile shape function.

3.2. Analysis of paracrystalline lattice distortion by single-peak method

Warren and Averbach reported that the pure diffraction peak profile free of instrumental broadening could be expressed by a Fourier series as Eq. (2) (Warren and Averbach, 1950):

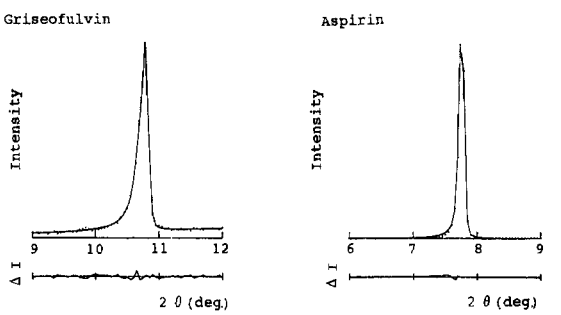

Fig. 1. Observed X-ray diffraction intensities and simulation curves of 100 reflection of aspirin and 011 reflection of griseofulvin. The split pseudo-Voigt function and linear function were used for the peak profile function and background intensities, respectively. ΔI shows the difference between the observed and calculated intensities.

$$I(h) = E(h)\Sigma\{A_n\cos(2\pi nh) + B_n\sin(2\pi nh)\}$$

$$h = 2d_0(\sin\theta - \sin\theta_{pk})/\lambda \quad (2)$$

(2)where A_n and B_n are the Fourier coefficients, n is the harmonic number, d_o is the interplanar spacing and λ is the X-ray wavelength.

The methods for the analysis of lattice distortions using a single diffraction peak (Mignot and Rondot, 1977; Zocchi, 1980; Delhez et al., 1982; Nandi et al., 1984) are based on Eq. (3) involving Stokes' (Stokes, 1948) corrected Fourier cosine coefficients, A_n (Klug and Alexander, 1974):

$$A_n = A_n^S A_n^D \quad (3)$$

where A_n^S and A_n^D are the size coefficient and the lattice disorder coefficient, respectively. After normalization of A_n^S so that $A_0^S = 1$, the size coefficients can be written for small values of n as Eq. (4), while the lattice disorder coefficients are expressed as Eq. (5) (Klug and Alexander, 1974):

$$A_n^S \approx 1 - na/M \quad (4)$$

$$A_n^D = 1 - 2\pi^2m^2 < Z_n^2 > \quad (5)$$

where a is the inverse of the period of the Fourier analysis, M is the size of the crystallites, m is the order of reflection and $< Z_n^2 >$ is the mean-square reduced displacement disorder.

Eq. (4) is generally valid only when $n \to 0$. Therefore, the single-peak method must be applied for Fourier coefficients at low harmonic number. However, these coefficients are subject to a truncation error, leading to a 'hook effect' (Warren, 1969). A method for correction of the 'hook effect' has been suggested in which the low harmonic coefficients are assumed to follow the exponential relation (Eq. (6)) (Rothman and Cohen, 1969)

$$A_n \approx \exp(-\beta n) \quad (6)$$

β is the parameter for representing the decrease of Fourier coefficients.

In the paracrystalline concept, the mean-square reduced displacement disorder ($< Z_n^2 >$ in Eq. (5)) can be expressed as Eq. (7) (Hosemann, 1972)

$$< Z_n^2 > = ng^2 \quad (7)$$

where g is the variance of the interplanar spacing within a disordered crystal (paracrystalline lattice distortion). In the present case, the expression for the disorder coefficients becomes

$$A_n^D = 1 - 2\pi^2m^2ng^2 \quad (8)$$

Nandi et al. (1984) have suggested that Eq. (6) is generally valid only for a small n. In this region, the disorder coefficients are almost unity, especially for the lowest-order peaks ($m = 1$) (Nandi et al., 1984). Thus, the size of the crystallites (M) can be evaluated from the initial slope of A_n at a harmonic number of 0 by Eq. (9), because the effect of the disorder coefficients is negligible.

$$\lim_{n \to 0} A_n \approx \exp(-na/M) \quad (9)$$

The size coefficients, being a small n, are also assumed to be an exponential expression (Eq. (10)) but this is only satisfactory when $2\pi^2m^2g^2 \ll a/M$.

$$A_n^S \approx \exp(-na/M) \quad (10)$$

Under this assumption, the size coefficient at $na = M/2$, $A_{M/2}^S$, becomes $\exp(-0.5)$. When the relation of $A_{M/2}^S = \exp(-0.5)$ is substituted in Eq. (3), the experimental coefficient at $M/2$, $A_{M/2}$ (determined by the interpolation of experimental coefficients), is expressed as Eq. (11). Thus, the paracrystalline distortion would be evaluated from Eq. (12)

$$A_{M/2} = \exp(-0.5)(1 - 2\pi^2m^2g^2) \quad (11)$$

$$g^2 = \{1 - A_{M/2}/\exp(-0.5)\}a/(\pi^2m^2M) \quad (12)$$

Fig. 2 shows a typical example of the analysis by the single-peak method. Calculated Fourier sine and cosine coefficients are plotted against a harmonic number, n. Fourier sine coefficients are the parameters representing peak asymmetry and does not relate the lattice distortion.

First step of the analysis is to determine the size of crystallites (M) from an initial slope of Fourier cosine coefficients at $n \to 0$. Next, Fourier coefficient at $M/2$ was estimated then the lattice distortion was calculated from Eq. (12).

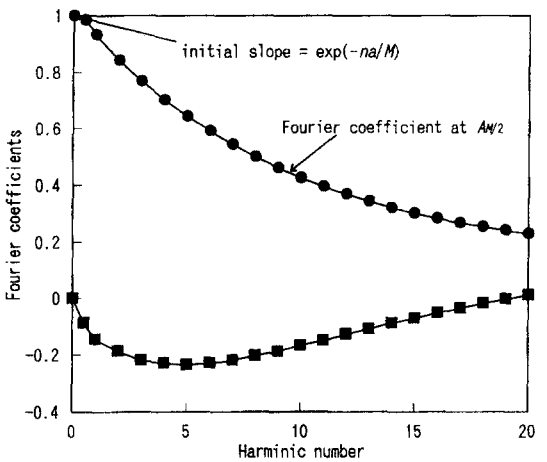

Fig. 2. Typical example of analysis by the single-peak method ●, experimental Fourier cosine coefficients; ■, experimental Fourier sine coefficients. Fourier coefficients were obtained by the Stokes' deconvolution procedure. Fourier sine coefficients are related with peak asymmetry.

3.3. Comparison of single-peak method and multiple-peak method

In a preceding paper, we reported the multiple-peak method for the determination of paracrystalline lattice distortion and the size of crystallites using at least three diffraction peaks (Fukuoka et al., 1995). To evaluate the validity of the single-peak method, M and g of intact and ground griseofulvin powders evaluated by the single-peak method were compared with those determined by the multiple-peak method.

As shown in Table 2, in the present method, the size of crystallites was underestimated and the lattice distortion was overestimated as compared with those determined by the multiple-peak method, because any decrease in A_n due to the disorder coefficients is ignored in the evaluation of M. Changes in M and g with the grinding time were similar to the results determined by the multiple-peak method (Fukuoka et al., 1995). This result suggests that the present single-peak method is valid for tracing the change in paracrystalline lattice distortion.

The single-peak method has a wider applicability to many other crystalline pharmaceuticals to evaluate the lattice distortion and the size of the crystallites than the multiple-peak method, because only one diffraction peak profile is necessary for the evaluation of M and g.

3.4. Determination of g and M of TOL and AZM powders

Fig. 3 shows the X-ray diffraction patterns of TOL and AZM powders. There were five well-resolved diffraction peak profiles in TOL (200, 110, 201, 210 and 111 reflections) and two in AZM (001 and 010 reflections), respectively.

The paracrystalline lattice distortion along five or two crystal planes could be evaluated by the single-peak method. Because other observed diffraction peaks at higher scattering angle overlapped with two or more reflections, these peak profiles could not be used for the present analysis.

Table 3 summarizes the g and M along five different crystal planes of intact and ground TOL, together with X_{cr} determined by the Ruland method. The lattice distortion, g, increased and M decreased gradually with the grinding time. The changes in g and M were independent of the crystal plane, suggesting that the TOL crystal has little directionality in the plastic or elastic properties.

X_{cr} gradually decreased and k slightly increased with the grinding time. Changes in M and g were found to correspond to the changes in X_{cr} and k, respectively, suggesting that M and g may be useful indexes of crystallinity. In the Ruland method, it is difficult to detect the small changes of degree of crystallinity and lattice distortion, because X_{cr} and k are affected by the integral upper limits and the separation of diffraction intensities from crystalline parts from total intensities. So, X_{cr} and k may have some error.

Corresponding of the results obtained by the Ruland method and profile analysis also suggests that X_{cr} and k obtained by the Ruland method would be reasonable values.

Table 4 summarizes the g and M values along (001) and (010), together with X_{cr} and k, of intact and ground AZM powders. A marked decrease in M and an increase in g along (001) were observed with the grinding time, while small changes in M and g along (010) were observed. AZM molecules

Table 3
Size of crystallites and lattice distortion along five different planes of tolbutamide powders determined by the single-peak method

Sample	Plane											
	(200)		(110)		(201)		(210)		(111)		Crystallinity	
	M (Å)[a]	g (%)[b]	M (Å)[a]	g (%)[b]	M (Å)[a]	g (%)[b]	M (Å)[a]	g (%)[b]	M (Å)[a]	g (%)[b]	X_{cr} (%)[c]	k (Å^2)[d]
Intact powder	782	1.5	625	4.2	494	4.8	615	4.4	406	5.5	87	4.5
Ground for 0.5 h	487	2.3	501	4.9	498	5.2	480	5.3	361	6.1	83	4.5
Ground for 1 h	309	3.2	373	6.3	412	6.1	363	6.5	292	7.1	74	4.7
Ground for 2 h	281	3.5	340	6.3	403	6.4	336	6.8	280	7.2	68	5.0

[a]Size of crystallites.
[b]Paracrystalline lattice distortion.
[c]Degree of crystallinity.
[d]Lattice distortion parameter. After being ground for 3 h, a part of the crystals was transformed into metastable crystal form.

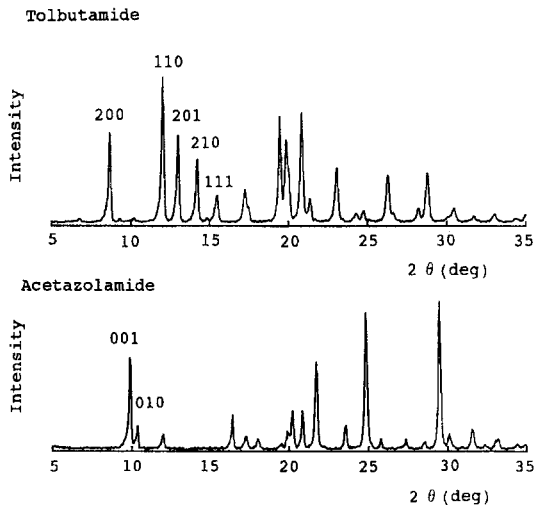

Fig. 3. X-ray diffraction patterns of intact powders of TOL and AZM. Numbers represent the reflecting plane indices.

are arranged with hydrogen bonds in the crystal as shown in Fig. 4 (Mathew and Palenik, 1974). The lengths of the hydrogen bonds along the c-axis were longer than those along the b-axis. The intermolecular force along the b-axis was considered to be stronger than that along the c-axis. A weak intermolecular interaction along (001) in the AZM crystal would explain the marked changes in M and g along (001) with grinding.

4. Conclusion

To evaluate the lattice distortion and the size of the crystallites of crystalline pharmaceuticals, we investigated the single-peak method for Fourier analysis of peak profiles of X-ray diffraction lines.

Table 2
Size of crystallite and paracrystalline lattice distortion of griseofulvin determined by the single-peak method and multiple peak method

Sample	Single-peak method		Multiple-peak method		Crystallinity	
	M (Å)[a]	g (%)[b]	M (Å)[a]	D (%)[c]	X_{cr} (%)[d]	k (Å^2)[e]
Intact powder	531	4.6	613	0.98	75	2.4
Ground for 1 h	388	5.7	568	1.15	71	2.9
Ground for 3 h	369	6.4	428	1.41	67	2.8
Ground for 5 h	336	6.6	393	1.45	67	3.1
Ground for 7 h	326	6.7	382	1.54	64	3.0

[a]Size of crystallites.
[b]Paracrystalline lattice distortion.
[c]Defined in a previous paper (Fukuoka et al., 1995).
[d]Degree of crystallinity.
[e]Lattice distortion parameter.

Table 4
Size of crystallites and paracrystalline lattice distortions along (001) and (010) of acetazolamide powders determined by the single-peak method

Sample	Plane					
	(001)		(010)		Crystallinity	
	M (Å)[a]	g (%)[b]	M (Å)[a]	g (%)[b]	X_{cr} (%)[c]	k (Å^2)[d]
Intact powder	674	3.9	542	4.5	82	2.9
Ground for 1 h	516	4.7	531	4.5	77	3.4
Ground for 2 h	464	5.1	544	4.5	77	3.4
Ground for 4 h	378	5.9	503	4.8	77	3.5
Ground for 6 h	368	5.9	506	4.9	73	3.9

[a]Size of crystallites.
[b]Paracrystalline lattice distortion.
[c]Degree of crystallinity.
[d]Lattice distortion parameter.

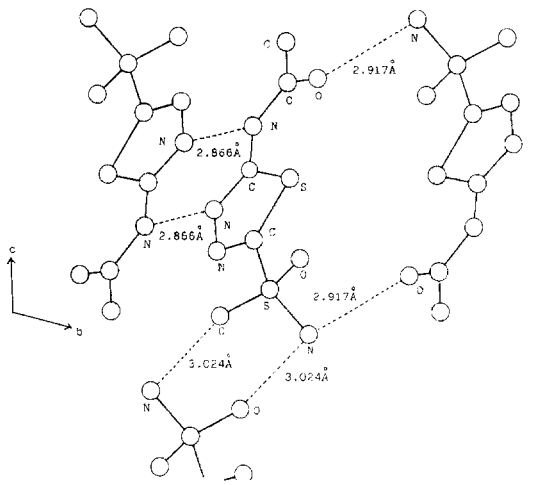

Fig. 4. Crystal structure of AZM projected on (100) plane.

The present method has wider applicability to many other crystalline pharmaceuticals than the multiple-peak method reported previously (Fukuoka et al., 1995). Furthermore, it was possible to evaluate the lattice disorder along some crystal planes. Molecular arrangement in the crystal was suggested to affect the lattice distortion along the crystal planes. We will discuss the relationship between the lattice distortion and pharmaceutical properties of crystalline drugs by the present method in subsequent reports.

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