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Optical surface roughness study of starch acetate compacts

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Abstract

Optical surface roughness of starch acetate compacts was investigated by specular reflection of a laser beam as a function of angle of incidence. The intensity data was fitted using the model of Gaussian and Lorentzian curves to solve numerical values for optical surface roughness which was observed to be order of 1 μm with the present samples. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

Surface roughness is one of the most important parameters of engineering surfaces. Therefore various theories and models have been developed for surface roughness assessment. Nowadays a standardized device for surface roughness measurement is based on the use of a diamond stylus. Using such apparatus which, however, is only for laboratory use, surface roughness parameters such as average and rms surface roughness and correlation length can be obtained. Unfortunately, the stylus records a surface profile along a very thin line, and therefore it gives only surface roughness in one dimension (typical stylus tip diameter is 5 μm and the measurement lag length is typically ranging between 1–10 mm). However, it is possible by a stylus to make stepwise scans on a rough surface. In this case an estimate for three-dimensional profile can be obtained. Nevertheless, such a measurement is usually time consuming. Stylus profilometer can not be applied for on-line measurement in a process industry such as metal industry. Another problem with a diamond stylus apparatus is that fraqile objects, such as pharmaceutical tablets, may be damaged during the inspection, and the reproducability of the measurement along the same line can be rather poor. Due to the importance of estimation of

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various surface roughness parameters other types of non-destructive measurement methods have been developed during the last few decades. Especially, optical techniques have been found useful for real time industrial measurement of surface roughness. In the metal industry measurement methods using specular reflection of laser light, triangulation based on laser profilometry (Cielo, 1988) and light scattering (Brodmann and Thurn, 1984) have been found useful. Other popular techniques for surface roughness estimation include speckle pattern metrology (Asakura, 1978), ellipsometry (Shiraishi, 1989), specular reflectance measurement with fiber optic sensor (Silvennoinen et al., 1992), specular index measurement (Peiponen et al., 1992a), grey-level histogram of optical Fourier transform pattern (Cuthbert and Huynh, 1992), and proximity sensors (Fawcett and Keltie, 1990; Gu et al., 1994). In the case of very smooth surfaces a measurement method based on total integrated scattering (TIS) (Bennett and Vögele Gourley, 1988; Bennett and Mattson, 1989) has been standardized for surface roughness estimation. It is also possible to measure simultaneously surface roughness and curvature of an object by using diffractive element sensor (DOE, Räsänen et al., 1995; Silvennoinen et al., 1996a).

In pharmaceutical technology there is an ever increasing desire to improve and optimize the function and quality of pharmaceutical tablets. One factor related to tablet quality that has great importance on drug release is the porosity of the tablet. A traditional method to get quantitative data about the porosity is based on the exploitation of a mercury porosimeter. Unfortunately, such a method is restricted for laboratory use only, in other words this technique is not suitable for porosity assessment in industrial environments on the process line. Another laboratory method, but for a qualitative picture, is the using of scanning electron microscope for surface porosity inspection. The above mentioned methods involve either mechanical disturbing or preparation of the samples, therefore the non-destructive testing is out of scope with these methods. Light interaction with powder materials including pharmaceutical powders can be also studied in laboratory using optical spectroscopy (Wilkinson et al., 1991; Olinger and Griffiths, 1993; Burger et al., 1997). Novel optical metrology of tablet quality inspection has created interest recently (Wang and Zaidi, 1991; Healy et al., 1994, 1995; Ketolainen et al., 1995; Silvennoinen et al., 1996b, 1997, 1998; Podczeck, 1998).

In this paper the optical surface roughness of pharmaceutical compacts is estimated using laser light specular reflectance technique. We wish to remark that in the case of laser profilometry, based on focusing of a laser beam, it is possible to make line scans and obtain estimate for surface texture in three dimensions. However, in specular reflection technique based on using plane wave of a laser beam, surface roughness information over a macroscopic area can be achieved immediately. Furthermore, the latter mentioned technique is much more cheaper to realize. The purpose of the present study was to get information about the surface statistics of height distribution and also get quantitative estimate for the optical surface roughness. This was realized by investigating rectangular starch acetate beams (Akazawa, 1976; Raatikainen et al., 1997). Of course, normally tablets take other shapes such as oval or concave shape. However, the height distribution of rectangular beam samples can provide information that may be useful in modeling of surface roughness for tablets of various shapes.

2. Theory

2.1. Definition of surface roughness parameters

In engineering perhaps the most often utilized surface roughness parameters (which are obtained by measuring a one-dimensional surface profile by stylus, $z = z(x)$, along x-axis) are average surface roughness R_a:

$$R_a = \frac{1}{L}\int_0^L |z(x)| \mathrm{d}x, \quad (1)$$

where L is the lag length, and root mean square (rms) surface roughness R_q:

$$R_q = \left[\frac{1}{L}\int_0^L z(x)^2 \mathrm{d}x\right]^{\frac{1}{2}}. \quad (2)$$

Both parameters can be obtained using either mechanical stylus or laser profilometer. However, it is important to know that the traditional surface roughness parameters are defined using the concept of mean-line or plane. In practise, especially for concave or convex surfaces, the values of surface roughness parameters depend strongly on the length of line scan. In other words, the macroscopic curvature can have an impact on the calculated surface roughness parameters (see e.g. Cielo, 1988).

In general, the surface profile related to the heights of the surface has to be considered as a function $z = z(x,y)$. If it is assumed that the surface has isotropic random surface roughness then Gaussian distribution of the heights $w(z)$:

$$w(z) = \frac{1}{R_q(2\pi)^{\frac{1}{2}}} \exp\left[-\left(\frac{z(x,y)}{2R_q}\right)^2\right] \tag{3}$$

centered about the plane $z = 0$, can be utilized in mathematical description of such surfaces. Dispersion of roughness distribution, not only for a Gaussian distribution, can be obtained using a Hilbert transform (Peiponen et al., 1992b).

Other widely exploited surface roughness parameters are the average slope:

$$S_a = \frac{1}{L}\int_0^L \left|\frac{dz(x)}{dx}\right| dx \tag{4}$$

and rms slope:

$$S_q = \left[\frac{1}{L}\int_0^L \left(\frac{dz(x)}{dx}\right)^2 dx\right]^{\frac{1}{2}}. \tag{5}$$

The optical surface roughness measurement device based on the ideas of Brodmann and Thurn, mentioned in the introduction, provides an empirical slope parameter for rough surface. Quite often the distribution of slope is also assumed to obey a normal distribution. However, in context of machined metal surfaces it has been shown that it is possible that the surface height distribution is Gaussian but the distribution for slope is non-Gaussian (Tanner, 1979).

Finally we write down the correlation function, C, which is defined as follows:

$$C(\tau) = \frac{1}{L}\int_0^L z(x)z(x+\tau)dx. \tag{6}$$

From Eq. (6) one can calculate the correlation length T using $C(T) = C(0)/10$. The correlation length measures the density of surface roughness in lateral direction. The estimation of correlation length can be problematic since it depends e.g. on sampling length and also whether one is dealing with Gaussian or exponential correlation function (Ogilvy and Foster, 1989).

2.2. Optical surface roughness

One widely used model for optical surface roughness, R_{opt}, is based on the classical theory (Beckmann and Spizzichino, 1963). The theory is valid under assumptions that the height distribution is Gaussian as well as the correlation function $C(\tau) = \exp[-(\tau/T)^2]$. The curvature of the surface is also assumed to be much more larger than the wavelength of the light λ. In practise it means that $T > > \lambda$.

Let us next consider the situation where two adjacent rays of a laser beam, incident on the rough surface, experience a path difference, which phenomenon can be visualized using a schematic drawing like in Fig. 1. The optical path length (r) can be calculated using the notations of Fig. 1, and according to trigonometry it is found that:

$$\Delta r = 2z \cos\theta, \tag{7}$$

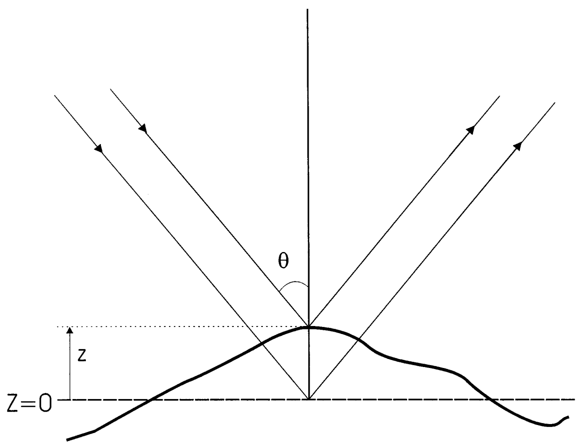

Fig. 1. A ray reflected from a surface irregularity relative to another ray that is reflected by the mean level of surface.

where z is the height or depth from the mean level ($z=0$) and θ is the angle of light incidence. Due to the reflection of rays from different levels there will appear also to be a phase shift which is as follows:

$$\Delta\phi = \frac{2\pi}{\lambda} 2z \cos\theta, \tag{8}$$

where λ is the wavelength of the laser. From Eq. (8) it can be observed that the phase shift tends to zero if the angle of incidence tends to $\pi/2$, i.e. the grazing incidence. Apparently the same effect appears when the value of surface roughness z decreases. In addition increasing the wavelength of the light causes the decreasing of the value of the phase shift. In our experiments, where the surfaces are not smooth, the technique of using large angles of incidence was applied. Therefore, the effect of increasing the angle of incidence is equivalent to decreasing of z. This, in turn, makes it possible to detect specular reflection. There always appears also diffuse reflection, which can be observed as increasing of speckle patterns (Asakura, 1978) while the angle of incidence is becoming smaller and smaller with respect to the normal of the sample surface.

The light intensity, I, detected at the far-field region can be calculated from the squared modulus of Fourier transform as follows:

$$I = \left| \int_{-\infty}^{\infty} w(z) \exp i \left(\frac{4\pi z \cos\theta}{\lambda} \right) \mathrm{d}z \right|^2, \tag{9}$$

where i is the imaginary unit. Because of the squared modulus we can not obtain probability distribution of the heights by measuring specular reflectance.

Nevertheless, we can always check if any theoretical model is appropriate for data analysis. Now in the case of Gaussian distribution w the Fourier transform gives a Gaussian function. Then according to Beckmann and Spizzichino the law for specular reflectivity is:

$$\rho = \rho_0 \exp[-(4\pi R_{\mathrm{opt}} \cos\theta/\lambda)^2], \tag{10}$$

where ρ_0 is the reflectivity from a perfectly smooth tablet surface and R_{opt} is the optical surface roughness. Unfortunately, a perfectly smooth tablet surface can never be achieved, therefore, it is assumed that the specular reflectance can be approximated from the following equation:

$$I = I_0 \exp[-(4\pi R_{\mathrm{opt}} \cos\theta/\lambda)^2], \tag{11}$$

where I is the measured specular light intensity and I_0 is the output intensity of the laser. Now the wavelength of the laser (a semiconductor laser), $\lambda = 635$ nm, is fixed in our experiments, but the angle of the laser beam incidence was changed. This was possible by using a goniometer as shown in Fig. 2(a). Fig. 2(b) shows the specular reflection and generation of speckle patterns together with corresponding one-dimensional light intensity distributions.

Specular reflection from lower and upper surfaces of the beams were measured and we considered the mean values of both measurements. This procedure is believed to cancel possible macroscopic curvature of the samples. By detecting the intensity I as a function of θ it is possible to try to solve R_{opt} from Eq. (11). A data fitting program was utilised to find optimal Gaussian curve to fit $I = I(\cos\theta)$.

3. Sample preparation

Three different starch acetate powders, each possessing different pharmaceutical properties, were manufactured by the Technical Research Centre of Finland, polymeric unit, Rajamäki, Finland. The origin of starch was barley. The pharmaceutical properties were altered by changing the degree of substitution of hydroxylic groups by acetate groups. Starch monomer possesses three hydroxylic groups and the degree of substitution (ds) describes the total acetate moiety content of the polymer. Materials here had ds values of 0.79, 2.09 and 2.5. As the ds increases, the behaviour of material alters from a rapidly dissolving tablet (ds 0–1.5) to an insoluble, matrix-like tablet (ds 1.5–3). Mechanical strength of starch acetate tablets is excellent being at the level of e.g. microcrystalline cellulose (Raatikainen et al., 1997).

Before compaction of sample beams, powders were stored for two weeks prior to compression under 33% relative humidity (RH). The rectangular beams, with cross section of $60 \times 6 \times 2$ mm

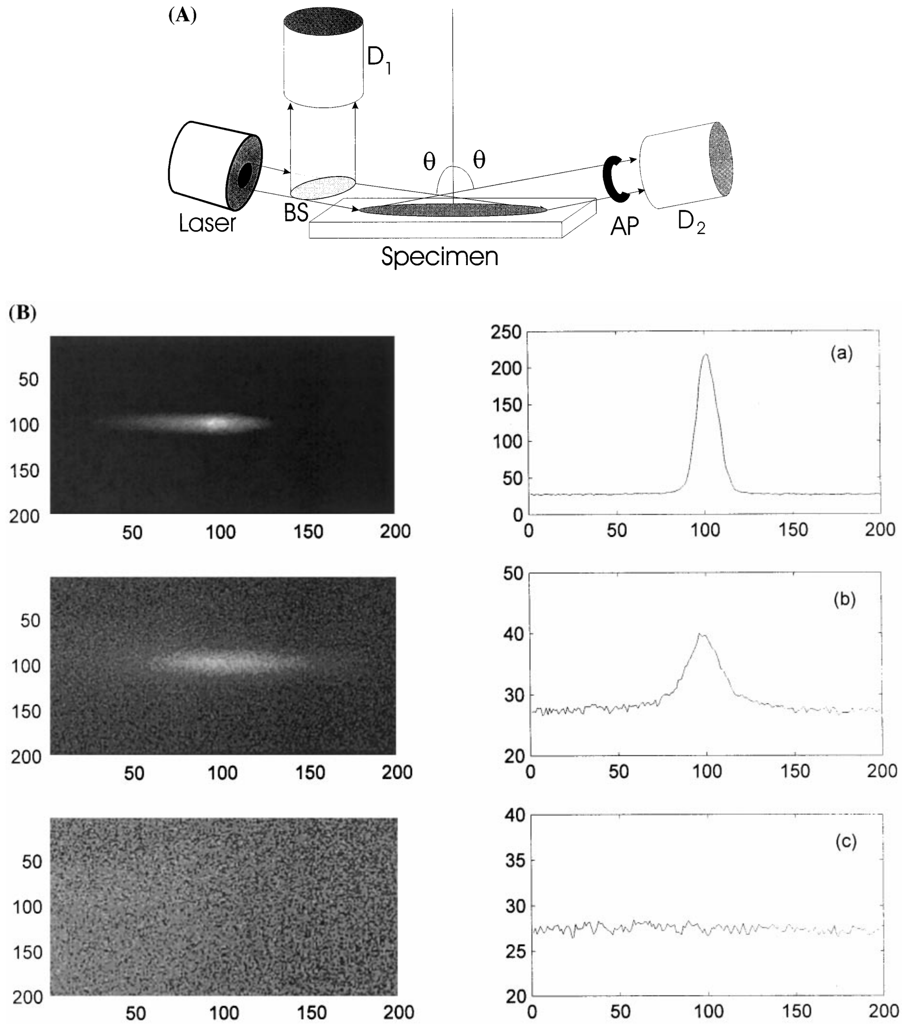

Fig. 2. (A) Schematic diagram of specular reflectance measurement using a goniometer. D_1, reference photodetector; D_2, signal photodetector; BS, beam splitter and AP, aperture. (B) Specular reflection and speckle patterns recorded by a CCD camera and the goniometer apparatus. The numbers shown on the axes are pixel numbers: (a) $\theta = 88°$; (b) $\theta = 78°$ and (c) $\theta = 60°$.

(measurement accuracy was ± 0.1 mm), were compressed using a hardened steel punch and die set. Compaction pressure was applied by manual hydraulic press (Pey Unicam, Carver Press, USA), which was operated by hand. Beams with different porosity were prepared by varying the weight of the powder in the die and compaction pressure. The maximum load was held constant for a dwell time of 10 s. The hydraulic press consists of a rough pressure gauge, however, since precise pressure was impossible to evaluate, the porosity of beams (after 24 h of recovery) were used as a descriptive character of sample. Ejection of the beam was performed by careful triaxial loosening of the die set around the compressed beam, which was to avoid structural failures possibly induced by that phase (e.g. lamination or capping).

4. Results and discussion

In Fig. 3 the results of measurements and the best Gaussian fit are shown. Evidently the signal could not be well fitted using Gaussian curve model. Therefore, we may conclude that the surface statistics of the present samples obviously is non-Gaussian. It is possible that noise due to diffuse reflection can be incident on the detector and may play a role in interpretation of the relation between the measured light intensity and the surface statistics. However, the specular component was more dominant than the diffuse one. Therefore, the role of diffuse light is believed to be minor. It was found that a Lorentzian intensity:

$$I = I_0 \frac{1}{1 + (4\pi z \cos\theta/\lambda)^2} \qquad (12)$$

gave the best fit, as shown also in Fig. 3. The Lorenzian intensity is related to exponential distribution w. This can be confirmed by a simple calculation based on Eq. (9). The optical surface roughness for the samples possessing different bulk porosity was calculated from Eq. (12), and the results are shown in Fig. 4 (where surface roughness estimates obtained by Eq. (11) are also shown). It was observed that the bulk porosity, within the present porosity interval, does not have a great effect on the values of surface roughness found from Eqs. (11) and (12). Nevertheless, it was observed that the specular reflectance could be measured for tablets with higher density at lower angles of incidence than with the tablets of lower density. Clearly the surface treatment by acetate has an impact on the surface roughness as shown in Fig. 4.

As a conclusion we state that optical surface roughness of starch acetate compacts in laboratory conditions can be investigated, using laser beam reflection technique. Gaussian and Lorenzian light intensity functions for quantitative estimation of surface roughness of the compacts give estimates of the same order. The large area compact suits better for near gracing incidence specu-

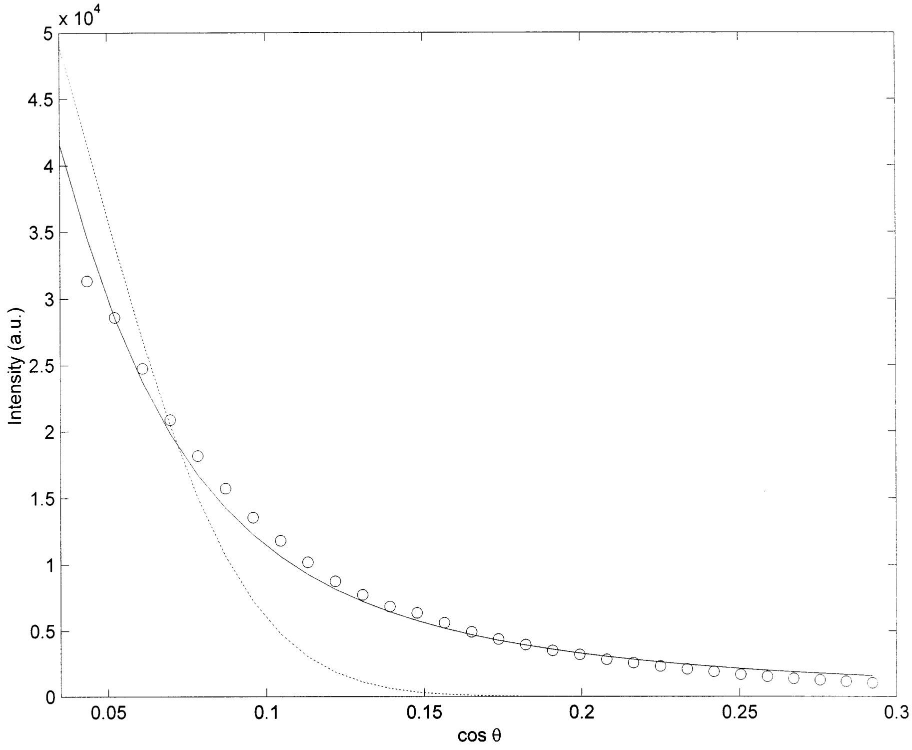

Fig. 3. Specular reflectance as a function of cos θ. Measured data (open circle), Lorentzian fitting (solid curve) and Gaussian fitting (dashed curve). The fits were obtained by a procedure where standard deviations were minimized.

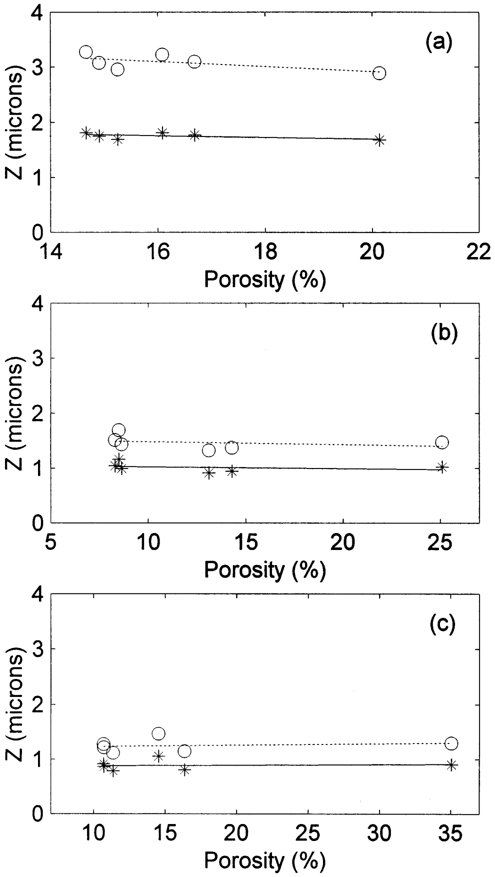

Fig. 4. Optical surface roughness of starch acetate compacts as a function of degree of acetate substitution and bulk porosity: (a) ds = 0.79; (b) ds = 2.09 and (c) ds = 2.5. Open circle, Lorentzian model and star, Gaussian model.

lar reflectance measurement than a tablet. The laser spot on the compact has a large area, approximately 1 × 25 mm. Therefore surface roughness information is obtained faster and from larger macroscopical area if we compare it with the instant area measured by a laser profilometer or a diffractive optical element based sensor. The present measurements help us to estimate appropriate statistical models for the surface roughness of tablets. Such information is needed e.g. for the calibration of the DOE sensor which can be used for simultaneous tablet surface roughness and curvature detection of concave and oval shaped tablets at normal light incidence.

We observed that degree of acetate substitution affects the surface roughness so that while the degree of acetate substition is increased the surface of the compact tends to be smoother. This is caused by the increased deformation of primary power particles as a function of degree of substitution. Moreover, the surface roughness is weakly dependent on the bulk porosity with the present samples. This indicates that the deformation of powder particles of starch acetate is also weakly dependent on compaction pressure.

Finally we remark that the present measurement technique can be applied also to inspect compacts from other type of powders.

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