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Local and Average Gloss From Flat-Faced Sodium Chloride Tablets

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ABSTRACT

The purpose of this study was to detect local gloss and surface structure changes of sodium chloride tablets. The changes in surface structure were reflected by gloss variation, which was measured using a diffractive optical element-based glossmeter (DOG). By scanning a surface area, we constructed a 2-dimensional gloss map that characterized the tablet's surface structure. The gloss variation results were compared with scanning electron microscopy (SEM) images and average surface roughness values that were measured by conventional diamond stylus profilometry. The profilometry data showed a decrease in tablet surface roughness as a function of compression force. In general, a smoother surface contributes to higher average gloss values. The average gloss values for this material, in contrast, showed a decrease as a function of the compression force. The sequence of particle fragmentation and deformation together with crack formation in sodium chloride particles resulted in a loss of gloss for single sodium chloride particles at the tablet surfaces, which could be detected by the DOG. These results were supported by the SEM images. The results show that detailed information regarding tablets' surface structure changes can be obtained by detection of local gloss variation and average gloss.

KEYWORDS: gloss, tablet, surface structure, sodium chloride.

INTRODUCTION

In the production of pharmaceutical tablets, powder materials are compressed to produce tablets with predefined properties. The surface properties of tablets, such as interparticle bondings, surface roughness, and pore size, are important factors that are related to drug release, adhesion of coating materials to regular/irregular surfaces, and possible crack formation during and after tabletting. Surface properties are mainly controlled by the initial particle properties, the deformation properties of the material to be compressed, the compression load, and the speed of the punches during tablet compaction. Surface roughness of the porous tablet indicates the surface quality of the tablet and can even predict the success of tablet production as a whole.

Laser profilometry has been used in the assessment of the microsurface roughness of porous media, such as paper[1] and tablets.[2,3] Using a laser profilometer, one can scan a macroscopic area and calculate various parameters such as average surface roughness from spatial topographical data.

Gloss, like surface roughness, is a useful surface quality parameter and is obtained using the specular reflectance of incident light.[4] Gloss depends on not only surface roughness but also the refractive index of the surface. The refractive index of a surface is influenced by the surface material and its density.

Pharmaceutical researchers have already shown an interest in the gloss of coated tablets.[5,6] Unfortunately, the measurement of tablet gloss, such as in the study by Rowe[5] and in industrial gloss inspection of various products, requires flat-faced surfaces. Universal gloss standards are defined for only flat surfaces. Hence, commercial glossmeters have been developed for only flat surfaces and provide information on the average gloss across a relatively large macroscopic area. Such devices integrate gloss information from the object surface. Recently, the problem of measuring local and average gloss from complex shaped objects has been solved using a diffractive optical element-based glossmeter (DOG).[7,8] The DOG provides information about local gloss and average gloss of curved and flat samples by scanning whole objects. An excellent correlation between the DOG signal and the commercial glossmeter was found for flat samples.[7,8] Basically, the DOG is similar to the device used for inspecting the surface quality of the flat and concave punches used in tabletting machines.[9,10] However, the present study differs from the studies by Hyvärinen et al,[9,10] since the DOG signal is directly related to gloss instead of to the intensity distribution projections of a light pattern. In addition, a 2-dimensional map of gloss variation is a new feature that can be obtained from the surface of a tablet.

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The DOG differs further from commercial glossmeters since the measurement is conducted at normal light incidence. Therefore, light polarization is not an important issue. The DOG involves the use of coherent laser radiation, the receptor aperture angle is comparable to that of the human eye, and vertical movement of the object is to some extent (a few millimeters) permitted. The sensitivity of the DOG to small gloss variations is better than with conventional glossmeters that use oblique light incidence. The DOG is also sensitive to the local refractive index changes of the medium. It has been shown in the case of paper[11] that the measurement of surface roughness is not sufficient to obtain a full picture of surface quality; the variation of spatial refractive index (ie, information on gloss by the DOG) is needed. In addition, glossy and dull surfaces can be measured with the DOG solely by means of 1-angle geometry, namely the normal incidence of laser radiation.

The main aim of this paper was to study the local gloss variations and average gloss of flat-faced sodium chloride tablets by means of a DOG. The reason for choosing only sodium chloride powder as a test material for this study was to examine local gloss changes of relatively glossy tablets.

MATERIALS AND METHODS

Materials

The material used in this study was sodium chloride (glidant-free, Akzo Nobel, Hengelo, The Netherlands). The coarse sodium chloride was milled (Moulinex, Birmingham, UK) and sieved with an Alpine Air Jet Sieve (Alpine, Augsburg, Germany) to obtain a particle size fraction between 106 and 212 μm. Pictures of single sodium chloride particles were made by a light microscope (Olympus BX-50, Olympus, Tokyo, Japan) connected to a digital camera (Sensicam, PCO, Kelheim, Germany).

Methods

Tablet Preparation

Cylindrical flat-facet tablets (500 mg, diameter 13 mm) were compressed with a compaction simulator (PCS-1, Puuman Ltd, Kuopio, Finland) at an average compaction speed of 4 mm $second^{-1}$. The upper punch displacement was a sine wave with different amplitudes to create different compression forces. The lower punch was stationary during the compression of the sodium chloride tablets. The die was prelubricated with magnesium stearate prior to each compaction to ensure a continuous force transport throughout the entire powder bed. The resulting compression forces varied between 7 and 35 kN. The average ejection time was 2 seconds. After a relaxation time of 24 hours, the tablet dimensions were measured with a micrometer (Digitrix, NSK, Osaka, Japan) and weighed with an analytical balance (A200S, Sartorius, Goettingen, Germany). The tablets were stored in a desiccator for 5 months prior to the measurements to allow for possible structural relaxation.

Gloss Measurement by DOG

An essential part of the DOG is a diffractive optical element (DOE). The DOE employed was a binary amplitude element, calculated using the Rayleigh-Sommerfeld diffraction integral.[12] The element was produced by electron beam lithography. The imaging properties of the DOE have been reported by Silvennoinen et al,[13,14] and therefore only a brief description of the DOG is given here. A schematic diagram of the DOG is shown in Figure 1. The DOE produces a 4 × 4 light spot matrix at its focal plane (f =100 mm). The light source is a low-power HeNe laser with a wavelength of 632.8 nm. A light spot matrix recorded with the DOG is shown in Figure 2a. The distance between neighboring light spots is approximately 125 μm. The DOE used in this study is a computer-generated hologram. It is sensitive to both the amplitude and the phase of the scattered field. The diffracted light pattern is incidented directly on the chip of the charge-coupled device (CCD) camera; there is no object lens in the camera.

The analysis of gloss is based on the calculation of the total intensity I of the detected DOE image. Total intensity is defined as follows[8,9]:

$$I = \frac{1}{nm} \sum_{i=1,\, j=1}^{n,m} I_{i,\, j}, \qquad (1)$$

when n and m are the dimensions of the region in which the total intensity I was measured, and $I_{i,j}$ is the image

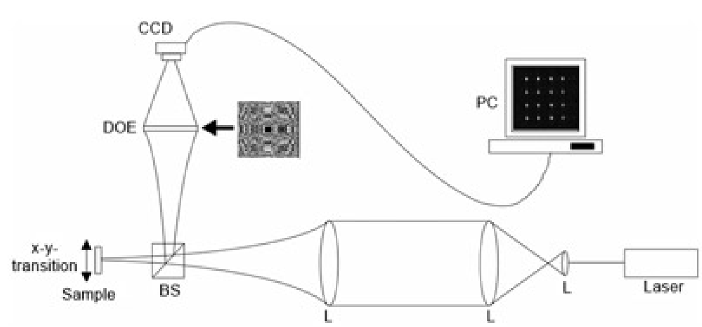

Figure 1. Schematic diagram of diffractive optical element-based glossmeter. CCD indicates charge-coupled device; DOE, diffractive optimal element; PC, personal computer; BS, beam splitter; L, lens.

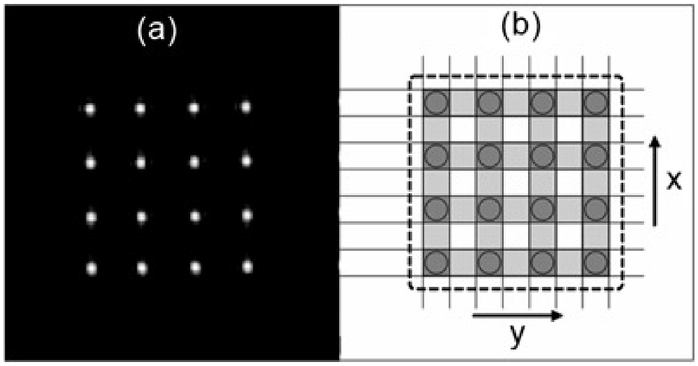

Figure 2. (a) Image pattern of 4 × 4 light spot matrix produced by the diffractive optical element. (b) Dashed lines mark the area, which was used for calculating the total intensity of the image pattern.

intensity observed by the $(i;j)th$ element of the CCD camera array. The gloss G, detected by the DOG, is defined as the ratio

$$G = 100 \times \frac{I}{I_{ref}}. \tag{2}$$

The reference surface I_{ref} is normally chosen according to a gloss standard, but in this study a high–optical quality mirror was used. In Figure 2b, the region in which the total intensity I was measured is indicated by a dashed line. The focal point diameter of the laser beam was 30 µm. Thus, the lateral resolution of the measurement is also 30 µm.

Stylus Profilometry

The surface roughness of 6 tablets for each compression was measured mechanically by a conventional diamond stylus profilometer (Mitutoyo, Surftest 201, Kawasaki, Japan). The radius of the profilometer's measuring tip was 5 µm, the tip pressure was 4mN, and the measuring speed was 0.5 mm/s. The measured length was 4 mm with a resolution of 10 µm.

Scanning Electron Microscopy Imaging

Scanning electron microscopy (SEM) images were taken from tablets using Field Emission Scanning Electron Microscope Hitachi S-4800 (Honshu, Japan) with 1 and 5 kV acceleration voltages. In SEM imaging, the use of voltage in the sample stage (deceleration voltage) improved the images' quality by preventing the charging of sample surfaces with low accelerating voltages. With this method, imaging of insulating material was performed without coating the samples with a conducting material—that is, gold.

RESULTS AND DISCUSSION

Tablet Surface Roughness

The tablet surface roughness profiles from 6 sodium chloride tablets for each compression force were measured by a diamond stylus profilometer. In Figure 3 the average surface roughness (R_a) is presented as a function of compression force. The average surface roughness is defined as an integral of the absolute value of the roughness profile measured over an evaluation length:

$$R_a = \frac{1}{L}\int_0^L |f(x)|dx, \tag{3}$$

where L is the lag length and $f(x)$ is the surface profile. Figure 3 shows that average surface roughness decreases as a function of compression force. The tablets that were compacted with low compression forces demonstrate high average surface roughness, while higher compression forces equalize the tablet surfaces to a larger extent.

Gloss of Single Sodium Chloride Particles

The surface structure of single sodium chloride particles was initially analyzed by light microscopy. A microscopy image of a single sodium chloride particle is presented in Figure 4 as illustration. Sodium chloride particle surfaces have some flat, smooth, and shiny parts; other parts are more rough and irregular.

Figure 5 presents the gloss measurements scanned from a 1.5 mm × 1.5 mm area containing single sodium chloride particles. This sample was prepared by spreading the sodium chloride particles on a mat black gloss standard ($G = 0$). It is preferable for the sodium chloride particles to lie on the surface of the gloss standard as a single particle

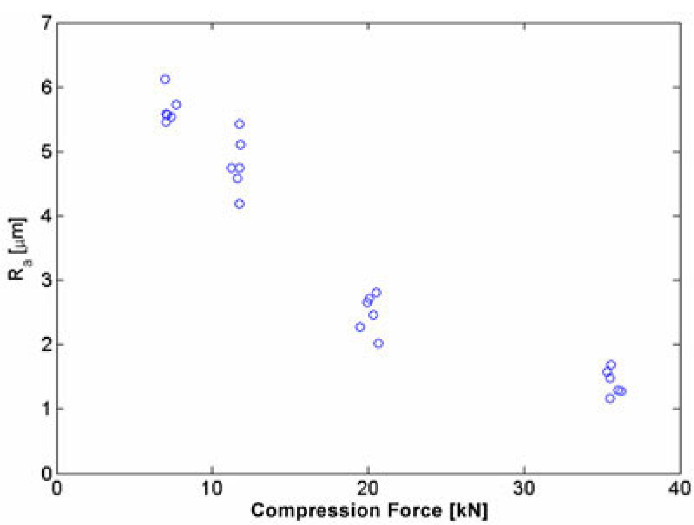

Figure 3. Average surface roughness R_a measured from sodium chloride tablets as a function of compression force.

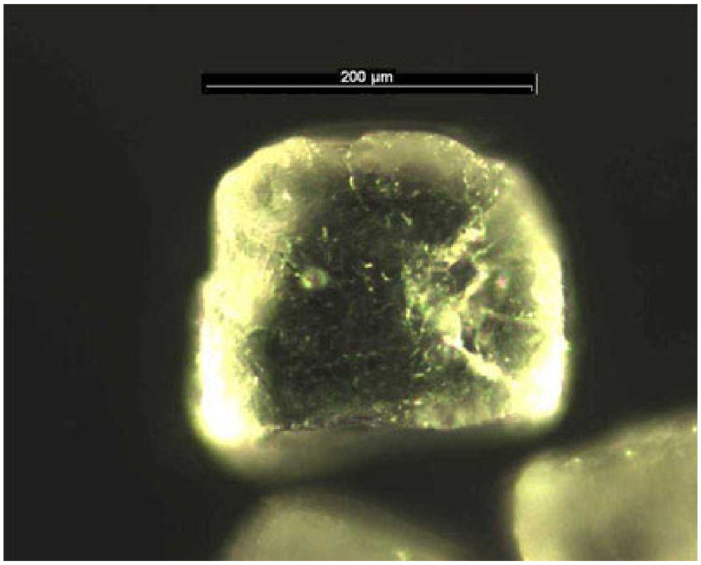

Figure 4. Microscope image of a single sodium chloride particle.

layer. Because of the rectangular particle shape, most sodium chloride particles rest on a long side. Some of the particles may lie on a tilted angle; in this situation, the light reflected from the surface of the sample extends beyond the detection angle of the glossmeter. In Figure 5 the contrast in gloss between particles and background can be distinguished. Although most particles have a gloss between 0.6 and 0.8, some single measurement points show higher gloss values of 1.4 to 1.7. A single measurement point equals the focal point of the laser beam incidence on the particle surface and, as mentioned earlier, is 30 μm in diameter. This means that the focal point diameter of the laser beam is smaller than the initial particle size of 106 to 212 μm. When the laser beam is reflected by the flat, relatively smooth and shiny surface of a sodium chloride particle (Figure 4), a gloss of approximately 1.7 G can be detected. Therefore, it can be postulated that the specular gloss of the present sodium chloride particles is about 1.7 G. The refractive index of sodium chloride is 1.54 at the 633-nm wavelength.[15] The gloss value at normal light incident from a perfect sodium chloride crystal mirror facet can be theoretically calculated using Fresnel's equation for light reflection. The theoretical gloss of such sodium chloride crystals would be 4.5 G. Lower values are measured from the crystals because of the minor surface roughness of single sodium chloride crystals, which causes light scattering.

Gloss of Sodium Chloride Tablet Surfaces

The gloss variation of tablets compacted with different compression forces was examined with the DOG. During the gloss measurement, an area of 3 mm × 3 mm was scanned for each tablet. The average gloss data of 6 individual tablet surfaces for each compression force are presented in Figure 6. These data were obtained by calculating the average gloss from the scanned 3 mm × 3 mm gloss map. A larger variation in gloss between similar tablet surfaces for lower compression forces is observed in Figure 6. Figure 6 also shows that the average gloss of the tablets decreases almost linearly as a function of the

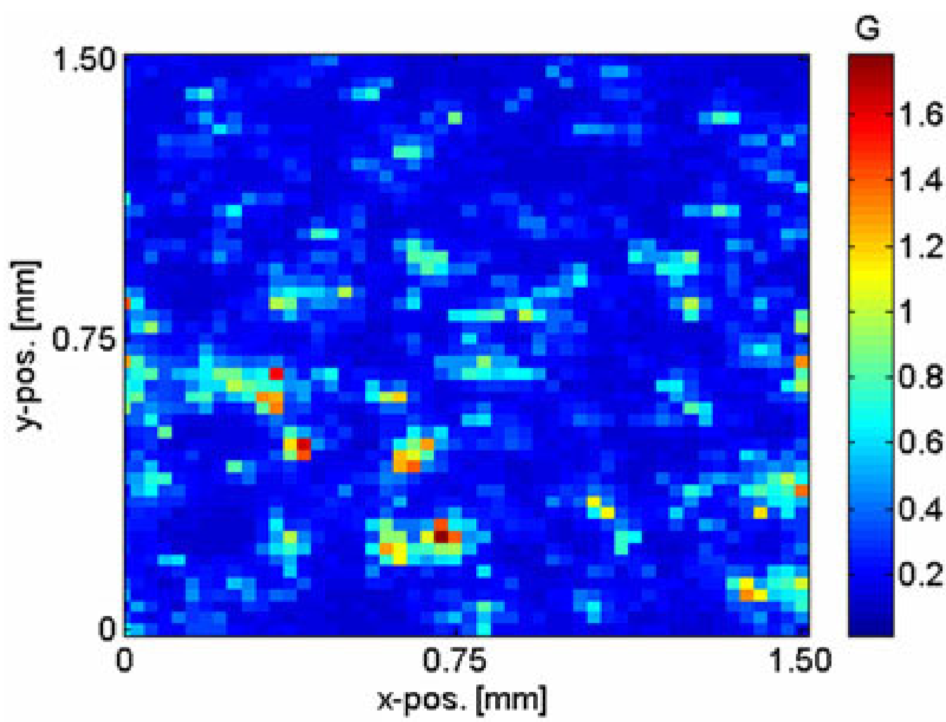

Figure 5. Two-dimensional gloss map of 1.5 × 1.5 mm^2 area of single sodium chloride particles lying in mat black commercial glossmeter gloss standard (G = 0).

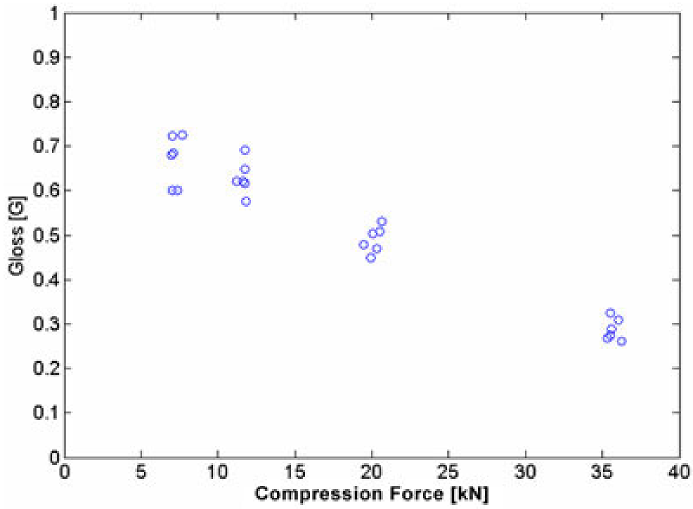

Figure 6. Average gloss as a function of compression force measured from sodium chloride tablets.

compression force. Assuming that there is no change in refractive index, this trend in average gloss value suggests that the surface roughness of tablets increases when the compression force rises. This is in contradiction to the surface roughness results measured by stylus profilometer (Figure 3). It is generally assumed that a smoother surface area will yield higher gloss values.

When the data of Figures 3 and 6 are combined, it can be deduced that at low compression forces the surface of sodium chloride tablets is clearly more glossy and rougher than that of high-compression-force–compressed tablets. It is interesting that samples with higher surface roughness can be glossier than smooth samples.

Information regarding local gloss variations is given by the 2-dimensional gloss maps. Two typical 2-dimensional gloss maps recorded for sodium chloride tablets (compression forces 11 kN and 35 kN) are shown in Figure 7a and 7b. Figures 7a and 7b show local gloss variations caused by mirror facets and the surface roughness of the tablet surfaces. Comparing the 2-dimensional gloss maps presented in Figures 7a and 7b, the variation in gloss is highest for the lower compression force and vice versa. In Figure 7a, several high gloss values of 1.4 to 1.7 are observed on the tablet surface, indicating smooth and shiny sodium chloride surfaces like those seen in single sodium chloride particles (Figure 5). These high gloss values are not detected at high compression forces (Figure 7b).

SEM Images of Sodium Chloride Tablet Surfaces

To explain and support given observations of changes in tablet surface roughness and gloss variation of sodium chloride tablets, SEM images were taken from the tablet surfaces. As noted above, the SEM images of tablet surfaces were taken without any coating layer. A large advantage of

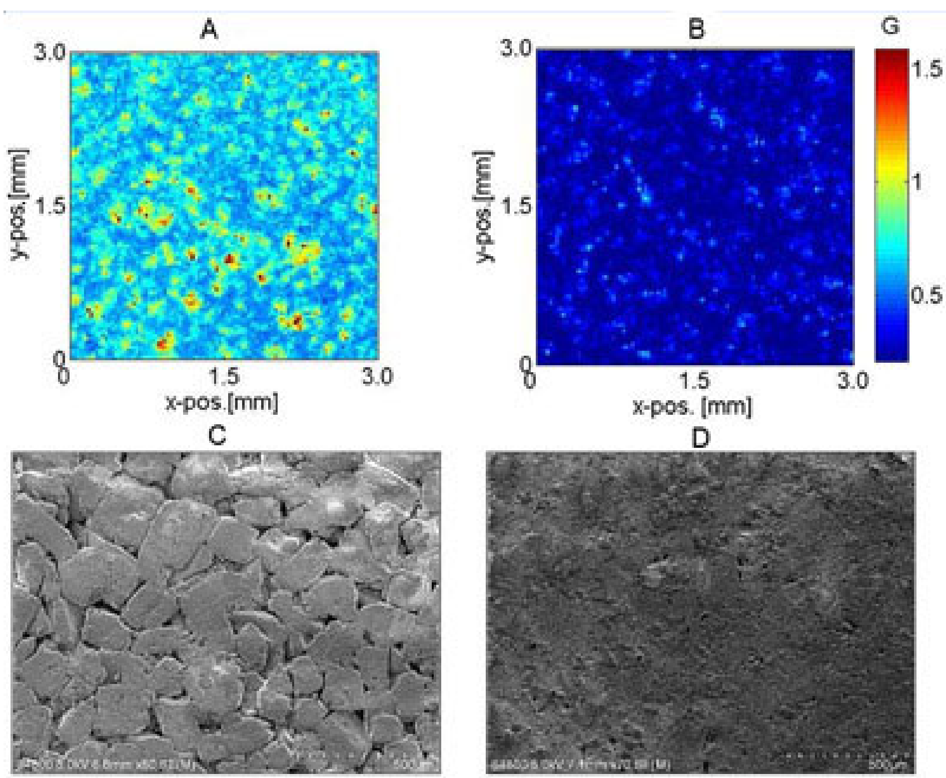

Figure 7. Two-dimensional gloss maps with local gloss variation from sodium chloride tablets compressed with compression forces of (a) 11 kN and (b) 35 kN. SEM images from surfaces of tablets compressed with corresponding compression forces of (c) 11 kN and (d) 35 kN.

this technique over the standard coating technique is that microscopic structure changes will not be obscured by the coating layer. Figures 7c and 7d contain SEM images of sodium chloride tablets compressed with the same forces as in Figures 7a and 7b (11 kN and 35 kN, respectively). When compression forces are low (Figure 7c), sodium chloride particles partially fragment or maintain their original shapes and predominantly deform at the particle corners.[16] Several sodium chloride particles preserve their smooth surfaces and related mirror facets (Figure 7a). Nevertheless, the tablet surface is rough at a macroscopic scale because of the random packing of relative large and unaltered sodium chloride particles (106-212 μm) and large pores between the particles as measured by stylus profilometer (Figure 3). At higher compression forces (Figure 7d), particles fragment and deform completely and cracks occur in the sodium chloride crystal structure.[16,17] The mirror property of single particles becomes worse, which leads to a reduction in gloss (Figure 7b). The tablet surface roughness at a macroscopic scale is flattened out at higher compression forces, while the highly deformed sodium chloride particles and cracked crystal structures maintain less gloss.

CONCLUSIONS

The DOG allows one to detect tablets' local and average gloss, which simultaneously depend on the surface roughness and variation of refractive index of the tablet surface. Two-dimensional gloss maps help us to visualize tablets' overall surface structure changes on a microscopic level. Along with conventional stylus profilometry, the DOG is useful device, particularly for obtaining information regarding surface structure changes of tablets and of other products such as coatings and films.

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