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THE SURFACE STRUCTURE OF COATED PAPER AND THE FORMATION OF GLOSS

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Abstract

The factors which are responsible for the surface reflection of coated paper at specular angles have been explored by means of models of specular reflection and of coating roughness. The model of specular reflection used is based upon the addition of the roughnesses which arise from independent sources, i.e. the roughness caused by the base paper on the one hand, and by the coating pigments on the other. Coating surface roughness is approached theoretically by the incorporation in a model of the influences of particle shape and size, size distribution and the hypothetical statistics of particle position at the surface.

The theoretical predictions have been tested with the aid of experimental data, on specular reflection, Hunter gloss, profilometric roughness, and particle size. The samples studied were different types of coated paper, blade-coated on a pilot scale.

Introduction

It can be considered that the requirement for paper gloss originates in the first place from the requirement for print gloss. Frequently, the two are closely related, as a consequence of the influence exercised by the paper roughness upon the roughness of the top surface of the print. On printed half-tones, paper gloss as such contributes to the resulting gloss. In this study, the relations between surface roughness and surface reflection, of which gloss is a parameter, are explored with the aim of acquiring more understanding of gloss formation on coated paper.

The problem of the relations between surface roughness and the distribution of surface reflection is of interest in diverse fields of technology. The various applications differ with respect to

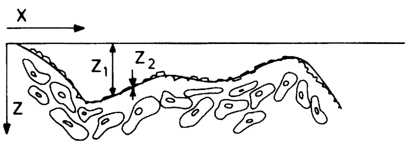
- the measuring conditions of surface reflection,
- the type of surface roughness, random or deterministic, and
- the magnitude of roughness in relation to the wavelength of the illuminating radiation.

As a rule, paper gloss is measured with white, unpolarised light, at a fairly large angle of incidence, measured relative to the surface normal. Detection occurs in the specular direction, while the angle of acceptance varies in accordance with the method of gloss measurement.

In regard to the roughness of coated paper, it arises from the roughness of the base paper, and from the roughness of the coating colour: it is largely statistically random. The contributions of these two components are governed by the coating process and the coat weight, which are themselves characterised by inherent length scales, or their inverse, frequency scales, as determined by the size of the constituents, viz. paper fibres and coating pigments.

In previous studies(1-5), coated paper gloss has been correlated with roughness in accordance with a wave optical approach(6-8). This is consistent with the assumption that paper roughness occurs in the length scale of the wavelength of light, and is small in comparison with this. Such an assumption is reasonable with a view to the roughness which originates from coating pigments. However, it cannot apply to the roughness which arises from the base paper, as the fibre size is some two orders of magnitude above that of the wavelength of light. Surface reflection from the base paper profile which is still evident at the coated surface is thus likely to obey the laws of geometrical optics(9). This suggests that if the formation of gloss on coated paper is to be accounted for, there is a need for an approach which combines the wave-optical and geometrical-optical starting points.

Methods



$Z = Z_1 + Z_2$

Z ROUGHNESS PROFILE

Z_1 ROUGHNESS OF THE BASE PAPER IN THE COATED SURFACE

Z_2 ROUGHNESS OF THE COATING LAYER

$\sigma^2 = E\left[(Z-\bar{Z})^2\right]$

σ ROOT MEAN SQUARE ROUGHNESS

E STATISTICAL AVERAGE

$A(r) = E\left[Z(x+r)\,Z(x)\right]$

A(r) CORRELATION FUNCTION

$a = \overline{A(r)}$

a AUTOCORRELATION LENGTH

$P(f) = F\left[A(r)\right]$

f SPATIAL FREQUENCY

P(f) POWER SPECTRUM

$f^2 P(f)$ ROUGHNESS SLOPE

F FOURIER TRANSFORM

Fig 1—Definition of terms

Firstly, the angular distribution of surface reflection is determined by the angular distribution of roughness, i.e. roughness slopes. The more deeply sloped a surface is, the lower also will be its specular reflection. If the roughness profile is regarded as a signal, the rate of its variation is related to the product of its amplitude and frequency. The amplitude of a random signal is given by its root mean square value (σ-value), while the power spectrum (P(f) in Fig. 1) expresses the distribution of σ at different frequencies. Consequently, the product $f^2P(f)$ can be used as a measure of the distribution of roughness slopes at different frequencies. This measure is presented in Fig. 2 in respect of the roughness arising on coated paper from the base paper, and of that arising from coating colour pigments, using typical values for the roughness parameters, and a Gaussian power spectrum of roughness. For the present argument, the exact form of the power spectrum is not critical. It is evident from Fig. 2 that the two sources of roughness fall into different frequency ranges and that the profile caused by pigment particles is steeper.

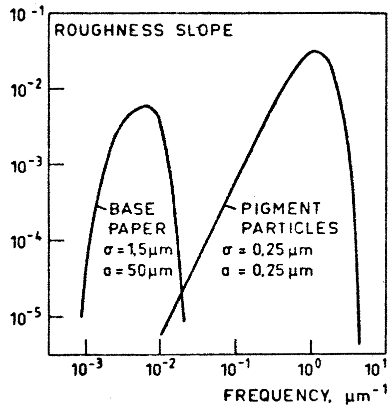


Fig 2—Distribution of surface slopes at different frequencies. Gaussian power spectra are estimated.

As the base paper and the coating colour make independent contributions to roughness, the ultimate roughness and its power spectrum are formed as sums of the two components (Figure 1). The influence on the specular reflection exerted by roughness, is characterised by different frequency ranges (Fig. 2), and arises in a more complex way. The reflection is directed in the specular angle either on the condition that both the microscopic and the optical profile direct it in this angle or that the sum profile does so. Mathematically this can be expressed as follows:

$$I = I_{ref}\left[\int_{-\Delta e/2}^{\Delta e/2} p_m(s_m)ds_m \cdot \int_{-\Delta e/2}^{\Delta e/2} p_o(s_o)ds_o + \int_{\substack{-\infty \\ |s_m| > \Delta e/2}}^{+\infty}\left[p_m(s_m)\cdot\int_{-\Delta e/2-s_m}^{\Delta e/2-s_m} p_o(s_o)ds_o\right] ds_m\right] \quad (1)$$

in which $p_m(s_m)$ and $p_o(s_o)$ denote the distributions of the microscopic and the optical roughness slopes s_m and s_o, while Δe is the angle corresponding to the angular spread of the specular reflection of the smooth reference. Provided that the distributions of roughness slope of the microscopic and optical profiles differ sufficiently, the second additive term in Equation (1) becomes small and can be discarded. This allows the specular reflection to be expressed as a product of the fractions A_m and A_o of the respective specular reflections.

$$I = I_{ref}\, A_m A_o \qquad (2)$$

To enable testing of the hypothesis formulated in Equation (2) by the aid of experimental data, some dependence of A_m on measurable profile parameters must be assumed. A likely form of the relationship between A_m and the surface slope s_m is exponential.

$$A_m = \exp\,(-ks_m^2) \qquad (3)$$

Equation (3) is equivalent to the expression for the specular reflection (A_o) from an optically rough (σ_o) surface[6,8].

$$A_o = \exp\,(-(4\pi\sigma_o \cos\psi/\lambda)^2) \qquad (4)$$

in which λ is the wavelength of light, and ψ the angle of incidence. The surface slope s_m is approximated in the following by means of the ratio of the σ_m-roughness and a characteristic depression width $\bar{b}$. The combination of (2), (3) and (4) thus produces:

$$I = I_{ref}\cdot \exp\left[-\,k(\sigma_m/b)^2 + (4\pi\sigma_o\cos\psi/\lambda)^2\right] \qquad (5)$$

Provided a satisfactory fit is obtained with the model of Equation (5), estimates for σ_o are determinable by the aid of experimental data in respect of specular reflection.

It can be assumed that the Equation (5) is applicable only when the acceptance angle of the receptor is so small that the detection is confined to the specular reflection in the macroscopic plane. When the acceptance angle $\Delta\omega$ is increased, the following applies:

$$I = I_{ref}\cdot\left[A_m + \int_{\Delta\omega} dA_m\right]\left[f_o + \int_{\Delta\omega} df_o\right] = \left[A_m + \Delta A_m\right]\left[f_o + \Delta f_o\right] \qquad (6)$$

In other words, the contribution of light other than that specularly reflected rises with an increase in the angle of acceptance of the receptor. It is evident that the additive terms in (6), as compared with those in (2), are more markedly dependent on the fine structure of the profile than are the specular terms[8]. The relative significance of the additive terms controls the way in which a relation exists between the results of gloss measurements made with a wide angle of acceptance, and the specular reflection of the sample.

With a view to exploration of the origins of coating surface roughness, as this is displayed in optical roughness (Eq. (4)), simple surface roughness models have been formulated. A coating itself contributes to surface roughness through particle shape, particle size, the size distribution, and the effects of particle orientation and packing. These effects have been included stepwise in the models introduced in Fig 3. Orientation effects are not formulated explicitly but can be taken into account by appropriate assumptions of particle shape. Pigment particles constitute the building blocks of the models. When no other type of roughness is present, particle shape and size, and their distributions, limit the smoothness achievable. The ultimate level of smoothness is dependent upon the manner in which the particles are aligned in the surface layer (cf. Fig. 3). Additional roughness is induced by deviation from tight packing.

In the following, surface roughness, as it arises from a coating containing two types of pigments, has been calculated in accordance with the additivity rule:

$$\sigma^2 = x.\sigma_1^2 + (1 - x).\sigma_2^2 \qquad (7)$$

in which x and (1 - x) refer to the weight fractions of the two pigments, and σ_1 and σ_2 to the roughness caused by each alone.

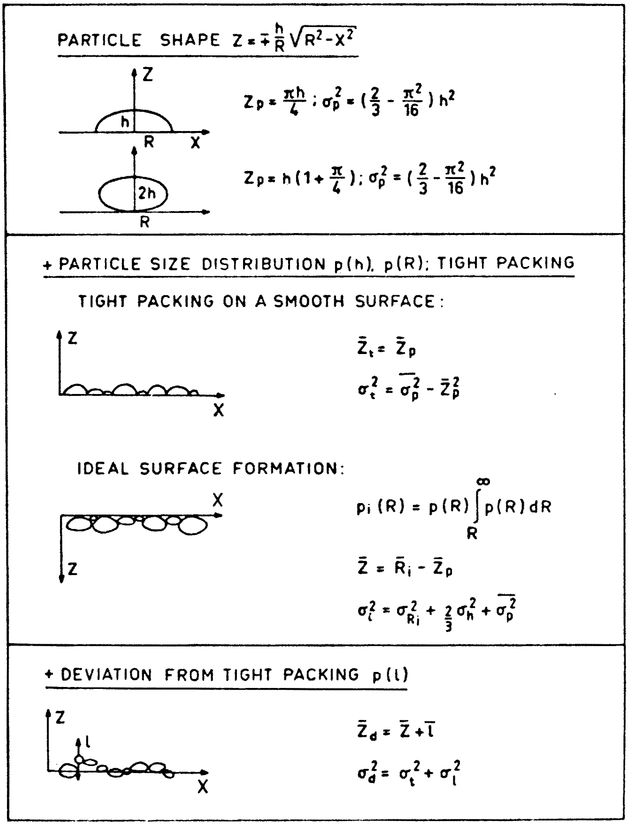


Fig 3—Surface roughness models.

Experimental

The surface reflection measurements were made with a Zeiss goniophotometer[10], and a Hunter glossmeter[11]. The angles of incidence and detection in all measurements were 75°. In the goniophotometer, the receptor field angles are variable in four steps, from 0.25 x 0.25° to 2 x 2°. Figure 4 shows the dependence of detected reflection of a black glass plate as a function of the receptor field angle. It is observable that the angular spread of the measurement is of such a magnitude that field angles of 0.5 x 0.5° are needed if all the specular reflection is to be detected. The term specular reflection is employed below in the discussion of measurements made on the Zeiss goniophotometer with a field angle combination of 0.5 x 0.5° in the illumination and the detection head.

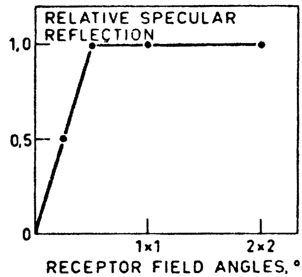


Fig 4—Influence of the receptor field angles on measured reflection in the specular direction. Black glass plate.

The measurements with the Hunter glossmeter were made in conformity with TAPPI Standard T 480[11]. The receptor aperture used in the measurement is circular, with a diameter corresponding to a field angle of 11.4°.

For the profilometric measurements of surface roughness, the apparatus described elsewhere[12] was used. Roughness in the frequency range of 7.5×10^{-4} to 5×10^{-2} μm^{-1} contributes to measured values of σ_m, and the width distribution of roughness. A characteristic width b is determined as the mean of the width distribution in the range of widths of 20 to 200 µm. The choice of the upper limit is dictated by the spectrum curves, such as those presented in Figure 2.

Particle size analysis was made on a Sedigraph apparatus. Table 1 lists the surface roughness data predicted by means of the models of Figure 3 for the pigments used.

Pigment	Roughness caused by Particle size σ_p µm	Particle size distn. σ_t µm	Distribution of particle position σ_d µm	Ideal surface σ_i µm
Clay 1 80% < 2 µm	0.05	0.19	0.21	0.16
Clay 2 50% < 2 µm	0.19	0.73	0.81	0.65
Talc 50% < 2 µm	0.19	0.66	0.74	0.56
$CaCO_3$ 1 90% < 2 µm: 40% < 0.5 µm	0.10	0.23	0.29	0.24
$CaCO_3$ 2 90% < 2 µm: 30% < 0.5 µm	0.11	0.25	0.31	0.23

Table 1
Surface roughness predicted by means of the models of Figure 3. Particle size and position statistics are included.

The series of samples of coated paper to be discussed represent LWC offset and gravure and woodfree offset paper grades. The samples were blade-coated on a pilot scale, on typical base-paper grades at different coat weights. The main variable in a coating is the pigment combination, and the greatest emphasis in this study was given to coatings containing clay and calcium carbonate.

Results and discussion

The surface reflection behaviour of coated paper at different coat weights can vary within the limits set by the base paper at light coat weights, and by the coating at heavy weights.

Typical curves are illustrated in Figures 5 and 6.

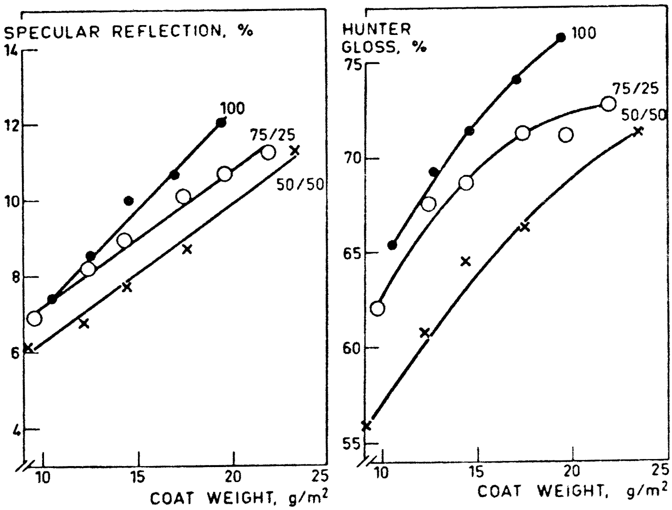


Fig 5—Specular reflection and Hunter gloss as a function of coat weight. Woodfree offset paper. Pigment combination expressed as the ratio of clay (1) and calcium carbonate (1).

The curves of gloss, plotted against coat weight are characterised by initial rises with increasing coat weight, followed by a tendency to level out. The rise is attributable to filling of the roughness volume of the base paper by the coating, while the emergence of a saturation level points to the effect of the coating itself. The curves of specular reflection differ from those of gloss plotted against coat weight, in that the slope of the curves normally remains constant up to higher coat weights. This indicates that the measurement of specular reflection is more critical to roughness in the microscopic scale than is the gloss measurement. However, if the specular reflection also approaches saturation, it can be inferred that the coating markedly restricts the development of gloss (Figure 6). In this case, the two types of curve have a similar shape.

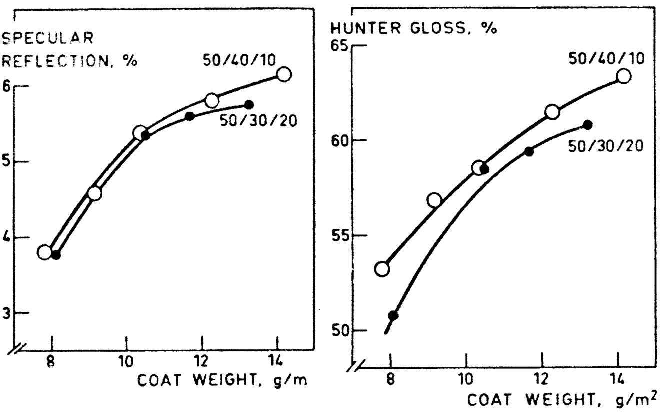


Fig 6—Specular reflection and Hunter gloss as a function of coat weight. LWC offset paper. Pigment combination expressed as the ratio of clay (1), talc and calcium carbonate (1).

The difference in the level of the specular reflection or gloss between different coatings at given coat weights, discernible in Figure 5, to some extent occurs also in the curves of the profilometric σ_m-roughness (Figures 7 and 8). As far as concerns the profilometric roughness, the cause may lie in the way different coatings fill the roughness volume of the base paper, or in influences exerted by the coating colour pigments. The fact that the characteristic depression width appears to be independent of the type of pigment (see Figures 7 and 8), allows the first hypothesis to be dismissed. The influence of the pigment particles can be assessed by application of the formula[13]:

$$\sigma_L^2 = \sigma^2/N \; ; \quad N = L/R \tag{8}$$

in which σ refers to the actual mean square roughness, and σ_L to its value as determined with a resolution of length L (20 µm),

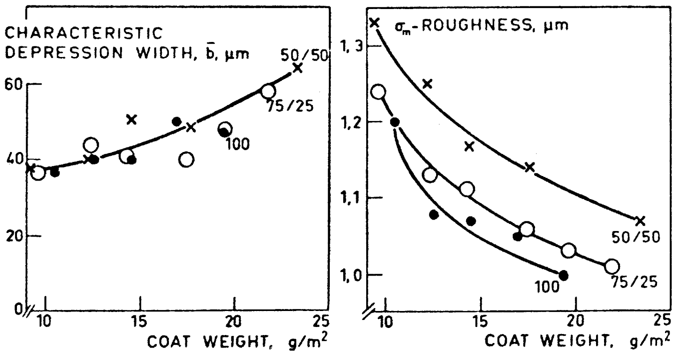


Fig 7—Profile σ_m -roughness and characteristic depression width as a function of coat weight. Samples of Fig 5.

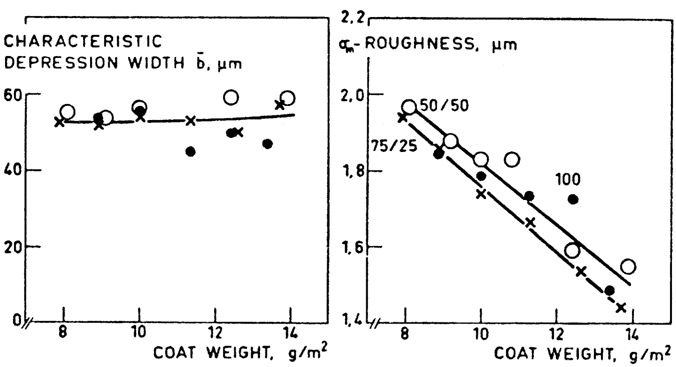


Fig 8—Profile σ_m -roughness and characteristic depression width as a function of coat weight. LWC offset paper. Pigment combination expressed as the ratio of clay (1) and calcium carbonate (1).

while $\bar{R}$ denotes the mean particle size. The data in Table 1 provide the possibility of confirming particle influence: coarse particles ($\bar{R} > 0.25$ µm) on a smooth surface ($\sigma_m < 1.5$ µm) act by increasing the profilometric σ_m-roughness. At most, the magnitude of the influence is 0.1 µm. As regards the characteristic depression width, a consequence of the flat power spectrum of pigment particles within the frequency range of the profilometric measurements, is that it is unaffected.

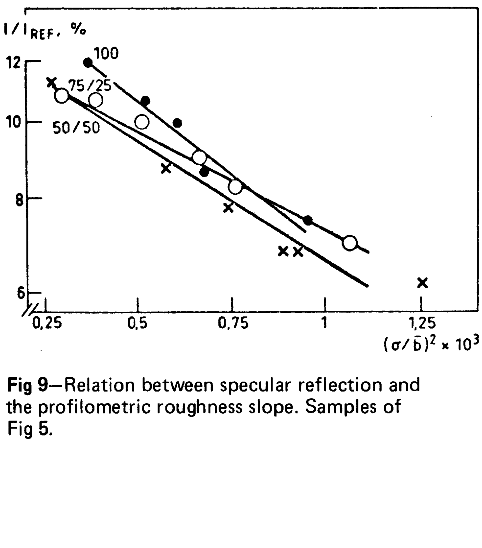


Fig 9—Relation between specular reflection and the profilometric roughness slope. Samples of Fig 5.

According to Equation (5), in a semi-logarithmic presentation the specular reflection and the square of the slope of profilometric roughness $(\sigma_m/b)^2$ should be linearly related. The data shown in Figures 9 to 11 support this hypothesis. The slopes of the lines express the quantitative role of the microscopic roughness. Fitting of regression lines to typical measuring data shows that on LWC type papers, this alone causes the specular reflection to be halved in comparison with the smooth reference. On woodfree paper, the diminution is of the order of 30 %.

The positions of the lines relative to the vertical axis characterises the optical coating roughness; from this the roughness parameter σ_o is determinable (assuming the wavelength of light to be 0.55 µm). Its magnitude rises with an increase of the fraction of calcium carbonate in the coating (Figs. 9 to 11), particularly above 25 % relative to clay.

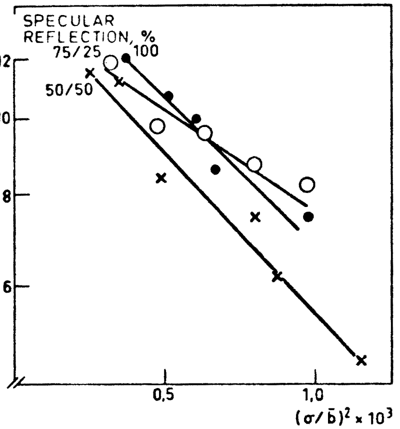


Fig 10—Relation between specular reflection and the profilometric roughness slope. Woodfree offset paper. The pigment combination in the coating colours given as the ratio of clay (1) and calcium carbonate (11).

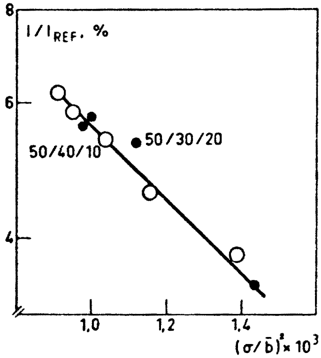


Fig 11—Relation between specular reflection and the profilometric roughness slope. Samples of Fig 6.

Figure 12 illustrates how σ_0 is related to the theoretical prediction of coating roughness. The theoretical prediction (cf. Figure 3) includes the statistics of particle size and particle position relative to the macroscopic plane; it has been assumed that the freedom of the particles to choose position is random within one particle diameter. As the calculations have been made with equivalent spherical diameters, the influence of particle shape is omitted. The orders of magnitude of the two measures of coating roughness are the same, despite the occurrences of systematic differences. The relatively lower optical roughness, observed with the increase in the fraction of calcium carbonate, can be tentatively explained by the contribution of these components to the profilometric roughness. On the other hand the optical roughness, in absolute terms higher than the theoretical roughness, which occurs in predominantly

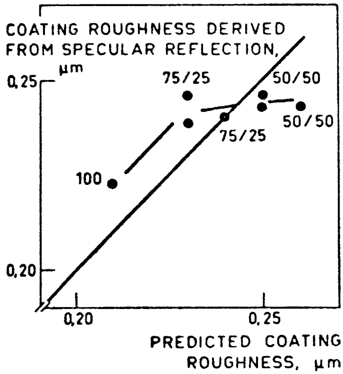


Fig 12—Relation between theoretically-predicted coating roughness (Fig 3) and roughness obtained from the specular reflection measurement by means of Equation (5). LWC and woodfree offset paper.

clay-containing coatings must be attributed to deficiencies in the theoretical model. Obviously the particles have a greater freedom with respect to their position than is expressed by one particle diameter. On the whole, however, the coating roughness can be predicted with reasonable quantitative accuracy with particle size and simple position statistics, omitting particle shape effects. This situation was found to hold for clay and calcium carbonate coatings but not for those containing talc. The extreme platelike nature of this pigment provides an explanation for this.

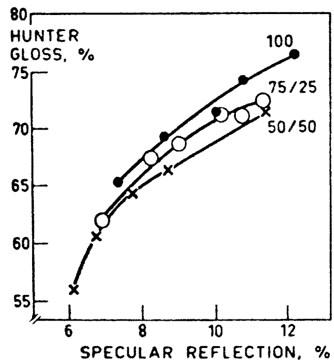


Fig 13—Relation between specular reflection and Hunter gloss. Samples of Fig 5.

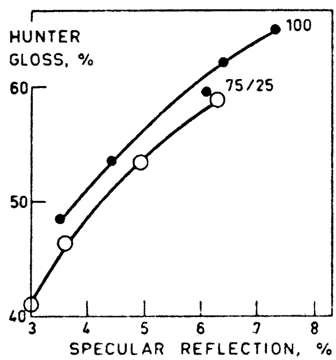


Fig 14—Relation between specular reflection and Hunter gloss. LWC gravure paper. The pigment combination given as the ratio of two grades of clay (I and II).

Figs. 13 and 14 compare Hunter gloss and specular reflection, and it can be seen that there is not a unique relationship between them on a given base paper. The relationship is governed by the pigment combination in the coating. Increasing the content of calcium carbonate or coarse clay leads to a lower Hunter gloss at a given level of specular reflection. It should be noted that the values of Hunter gloss far exceed those of the specular reflection, as a proof of the signifance of the additive terms in Equation (6). The integrating nature of the Hunter measurement gives grounds for the deduction that the gloss is more sensitive to the fine structure of the coating than is the specular reflection.

The fine structure can be interpreted in terms of particle shape, since the roughness influences originating from the base paper and particle size statistics have already been considered in connection with the specular reflection.

For the elimination of influences other than that of particle shape, Hunter gloss has been related to theoretical estimates of particle roughness at given levels of specular reflection (Fig. 15). In computation of the roughness arising from the pigment shape, it has been assumed that clay particles are plate-like with an aspect ratio of 1:10, and that calcium carbonate particles are spherical. The calculations have been made in accordance with the model illustrated at the top of Figure 3. For pigment mixtures, the additivity rule of Equation (7) has been assumed.

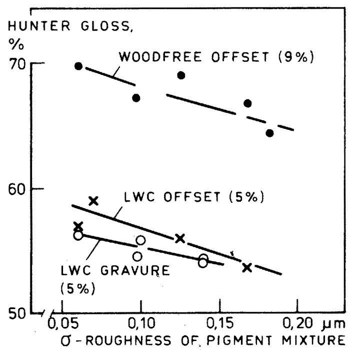


Fig 15—Relation between Hunter gloss and pigment roughness arising from pigment shape. Figures in parentheses refer to the level of specular reflection.

In Figure 15, a clear tendency for gloss to diminish with increasing pigment roughness is discernible, indicating that the additive term Δf_o in Equation (6) is less when the particles are rough, as a consequence of shape effects. Differences of about 5 Hunter units occur between all-clay and 50/50 clay/calcium carbonate coatings. The fact that coatings on different base papers fall on different levels of Hunter gloss is attributable to variations induced by the base-paper in the term ΔA_m of Equation (6).

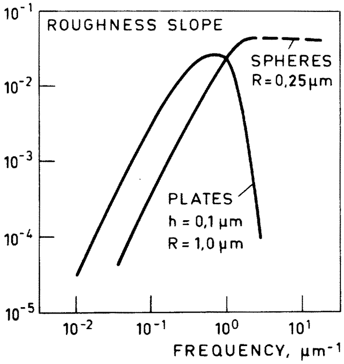


Fig 16—Distribution of surface slope at different frequencies for platelike particles (clay) and for spheres (calcium carbonate).

The calcium carbonate pigments utilised in this study are rougher than clay pigments, not only by reason of particle shape, but also because of particle size. A practical question is whether a decrease in particle size can result in a significant reduction in particle roughness, as this influences gloss. Roughness slope distributions for plate-like particles and for spheres calculated from their power spectra(14) are shown in Fig. 16 for particles of the same equivalent spherical diameter. The curves reveal a significantly higher spectral content of roughness slope with calcium carbonate at frequencies corresponding to the region of diffraction of light. It thus becomes evident that calcium carbonate coatings would be bound to scatter more light than are clay coatings, even if the particle sizes were comparable.

The spectral representation of roughness slopes can be further employed to summarise the results obtained in this study (see Figure 17). A measurement of specular reflection functions as an integrator up to a given very low level of roughness slope, marked by a line in Figure 17. The most marked variations between samples of coated paper up to this level originate in the base paper roughness, and the size of the coating pigment particles. However, a wide angle gloss measurement, such as the

TAPPI method, integrates over the roughness slopes up to a markedly higher level. Within this region, variations in particle shape become significant, whereas those originating in the base paper diminish.

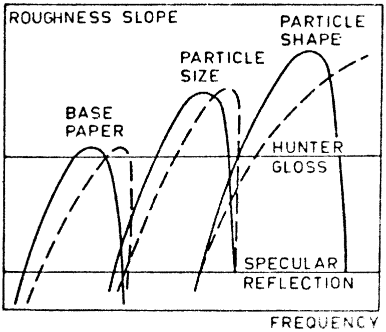


Fig 17—Schematic representation of the formation of gloss on coated paper.

Conclusions

The results obtained have shown that on coated paper, the specular reflection is influenced independently by the microscopic roughness profile for which the base paper is largely responsible, and the profile induced by the coating, viz. the overall specular reflection is formed as a product of the respective fractions of specular reflection. This mechanism indicates the path leading to good paper gloss as far as papermaking is concerned. It should be noted, however, that the role of the coating process was not investigated.

The influence of the composition of the pigments in the coating upon the specular reflection in all-clay and clay/calcium carbonate coatings can be accounted for by a roughness model, which incorporates the influences of the particle size distribution and simple statistics of particle position at the surface. The model thus offers a means of predicting the level of coating gloss on a given base paper. The fact that particle shape need not be taken into consideration demonstrates that the coating process does not effectively utilise the pigment properties.

Conversely, the values provided by a gloss measurement, using a fairly wide angle of acceptance, are influenced by the particle shape. This situation raises the question of the most appropriate way of measuring gloss when this is either an end-use property of coated paper, or a means of achieving print gloss.

Symbols used

a	autocorrelation length (μm)
$\bar{b}$	characteristic depression width of the microscopic profile (μm)
d	denotes a roughness model in which deviation from tight packing is allowed
f	spatial frequency (μm^{-1})
h	particle thickness (μm)
i	denotes ideal surface formation from pigment particles
l	variable for deviation from tight packing
m	denotes microscopic roughness
o	denotes optical roughness
p	distribution of a variable
s	roughness slope
t	denotes a roughness model with tight packing of particles
x	position co-ordinate
	weight fraction
z	roughness co-ordinate
A	fraction of specular reflection
I	intensity of reflection
I_{ref}	intensity of reflection from a smooth surface
L	resolution of profilometric roughness measurement (μm)

P	power spectrum
R	particle radius (μm)
λ	wavelength of light (μm)
Δe	acceptance angle in measurement of specular reflection
ψ	angle of incidence
$\Delta\omega$	acceptance angle of detection
σ	root mean square roughness (μm)

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Transcription of Discussion

Discussions following paper 6.2 by Dr. P. Oittinen

Dr. J. Colley

Could you please briefly describe your microscopic roughness measurements?

Dr. P. Oittinen

We make profilometric measurements in the wavelength range from 20 µm to 1 mm, with a stylus.

Dr. J. Colley

If your stylus is too big it will not give you a true picture of the roughness. What is its tip diameter?

Dr. P. Oittinen, The Finnish PPRI

About 5 µm at the tip.

Dr. E. Böhmer, The Norwegian PPRI

On your Figure 5 you plot coat weight against specular reflection for different pigment compositions. I agree that pigment composition has an effect on surface roughness, but how do you make allowance for differences of binder composition in the surface layers, which will also influence specular reflection?

Dr. P. Oittinen

We have not studied the influence of the binder in depth so far, but it appears that its main effect is to influence the particle positions at the surface.

Prof. J. Silvy, Ecole Francaise de Papeterie

Thank you very much Dr. Oittinen for your excellent description of the very difficult subject of the appearance of gloss of coated paper. Did you do any visual ranking of the appearance of the gloss in parallel with your physical measurements?

Dr. P. Oittinen

No.

Dr. H. Corte

Have you tried relating your reflectance measurements to the visual perception of gloss, as seen by a number of observers? Eight years ago I demonstrated that a combination of two perfectly matt surfaces could appear very glossy. I just wonder if there is any correlation between your instrumental measurements and people's perception of gloss?

Dr. P. Oittinen

We haven't investigated this. But, I am confident, judging by the results given by other workers, that instrumentally measured reflectance and peoples' perception of gloss are very closely related.