Preferred citation: V.G.W. Harrison. Optical properties of paper. In **The Formation and Structure of Paper**, *Trans. of the IInd Fund. Res. Symp. Oxford, 1961*, (F. Bolam, ed.), pp 467–485, FRC, Manchester, 2018. DOI: 10.15376/frc.1961.1.467.

OPTICAL PROPERTIES OF PAPER

V. G. W. HARRISON

PRINTING, PACKAGING & ALLIED TRADES RESEARCH ASSOCIATION, LEATHERHEAD

Synopsis

Optical properties of paper are those that govern its visual appearance—mainly, colour, opacity and gloss. These depend on the fact that paper consists of a network of doubly refractive transparent fibres immersed in air as medium and, in some cases, the optical properties can be expressed in terms of the absorption and scattering coefficients of the fibres and loading materials.

Opacity is the property most thoroughly understood and there is a British Standard for its measurement. The colorimetry of near-white papers still gives rise to difficulties, particularly with those containing optical bleaches and recent work on the assessment of sheets containing appreciable amounts of fluorescent materials is reviewed. The measurement of gloss is the most difficult to perform and our knowledge here is still far from complete. Little-known research on gloss done during the past fifteen years at PATRA *and by a group of Japanese workers is summarised.*

Les propriétés optiques du papier

L'apparence à l'œil nu du papier est réglée par les propriétés optiques de celui-ci—en particulier par sa couleur, son opacité et son brillant. Ces propriétés dependent du fait que le papier est composé d'un réseau de fibres transparentes et bi-réfringeantes entourées d'air. Dans certains cas, ces propriétés optiques peuvent être exprimées en termes des coefficients d'absorption et de diffusion des fibres et des matières de charge.

De ces propriétés, l'opacité est celle jusqu'ici la mieux interprêtée. Il existe un 'British Standard' pour son évaluation. La colorimétrie des papiers presque blancs reste sujette à des difficultés, surtout quand il contiennent des adjuvants optiques. Une revue est presentée des études récentes sur l'évaluation des papiers contenant des matières *fluorescentes. De toutes les propriétés optiques, le brillant est la plus difficile à évaluer.*

On présente les résultats de recherches peu connues, effectuées en partie au Japon et en partie à PATRA *pendant les quinze dernières années.*

Optische Eigenschaften des Papiers

Unter den optischen Eigenschaften von Papier versteht man solche, die die visuelle Erscheinung bestimmen—hauptsächlich Farbe, Opazität und Glanz. Dies ist eine Folge davon, dass das Papier aus einem Netzwerk von doppelt gebrochenen, transparenten, in Luft eingetauchten Fasern besteht, so dass die optischen Eigenschaften zum Teil durch Absorption und Brechungskoeffizient von Fasern und Füllstoffen charakterisiert werden können.

Am besten bekannt ist bisher die Opazität, für die es eine britische Standardmessmethode gibt. Die Farbbestimmung von fast weissen Papieren stösst noch auf Schwierigkeiten, besonders bei Anwesenheit von optischen Aufhellern. Kürzlich wurden Studien über die Messung von Blättern durchgeführt, die beträchtliche Mengen von fluoreszierendem Material enthielten. Die Glanzmessung lässt sich am schwierigsten durchführen, was die Unvollständigkeit der Kenntnis dieses Gebietes erklärt. Es wurde von wenig bekannten Forschungen über Glanz berichtet, die während der vergangenen 15 Jahre am PATRA-*Institut und durch eine Gruppe japanischer Wissenschaftler vorgenommen wurden.*

Introduction

BY optical properties of paper, we mean those properties that govern its visual appearance—that is, in general, its colour, opacity and gloss. The surface smoothness of the paper is also of importance in this connection, though it is by no means identical with gloss and is not strictly an optical property. Nevertheless, it is so closely connected with the scattering of light from the paper surface that it cannot be ignored.

The optical properties of paper depend almost entirely upon the fact that it consists of a network of doubly refractive transparent fibres, having a mean refractive index of about 1·56 immersed in air as medium. Repeated reflection and refraction of rays of light takes place at the numerous fibre/air interfaces within the sheet. If the gaps between the fibres are filled with some transparent liquid having a refractive index of around 1·56, the reflections and refractions at the interfaces are almost entirely suppressed and the optical properties of the sheet as we know them disappear.

The transmission of light through plane parallel layers of heterogeneous materials such as paper was described in some detail in a paper that I read to the Technical Section 21 years ago.(1) Despite the passage of this length of time, I think the description there given is still substantially true. The repeated reflections and refractions at the interfaces produce a scattering of light as it enters the sheet. Because of the scattering, some of the light is reflected back in the direction of the source. Some passes through the sheet, though it may emerge at a quite different angle from that at which it enters. Another fraction of light is absorbed and disappears, being converted into heat. Various mathematical analyses have been made of this process, varying somewhat according to the initial assumptions that are made about direction of incidence of the original beam, also what simplifying assumptions have to be made in order to render the system mathematically tractable. (For example, the fibres are usually replaced by an aggregate of uniform spheres of similar refractive index.) The analysis that has been most widely used and that appears to hold good with tolerable accuracy for a wide variety of papers is that originally introduced by Kubelka and Munk in 1931.(2) In this analysis, the optical properties of the paper, as far as its colour and opacity are concerned, are expressed by means of two constants—an absorption coefficient and a scattering coefficient. Other mathematical expressions generally reduce to similar form. Depending on the magnitudes of these coefficients, the appearance of the sheet in any given thickness will vary.

For a paper to have good opacity and whiteness, the scattering coefficient must be large; indeed, the hiding power of the sheet depends entirely upon the fact that a beam of light entering the sheet is broken up beyond recognition before it has penetrated any appreciable depth. As the scattering coefficient is reduced, the sheet becomes more translucent and the whiteness decreases. Finally, when the scattering coefficient is very low, the sheet approximates to a transparent paper. The absorption coefficient, on the other hand, governs the colour of the paper. If the absorption coefficient is constant for all wavelengths, a white or grey paper results, the paper being greyer the greater the absorption coefficient. Generally speaking, however, the absorption coefficient is not constant, but is a function of wavelength, being greater for blue light than for red. The result is a sheet of pale yellow colour. Dyestuffs have, of course, the property of altering the absorption coefficient considerably for certain wavelengths and this selective absorption produces the colouration of the sheet. A tinted transparent film would have a very low scattering coefficient and a high absorption coefficient, the latter varying with wavelength according to the colour of the film.

Opacity

PROBABLY, the simplest of the optical properties of paper to standardise is the opacity. Twenty one years ago, the situation was still chaotic. In the paper referred to,(1) however, preference was expressed for measurement by means of printing opacity and reasons were given for the choice. This method has become a British Standard.(3) Basically, printing opacity is the ratio of the reflectance of a solid black printed area viewed through one thickness of the paper under test to the reflectance of a pile of the unprinted paper. This ratio is 100 per cent for a perfectly opaque paper, zero for a perfectly transparent paper. The conditions of measurement have to be carefully specified.(4, 5)

Printing opacity seems to have stood the test of time well and gives a satisfactory measure of the tendency of papers to exhibit show-through. It is unfortunate, however, that no completely satisfactory apparatus complying with the B.S. specification is available on the British market.

The specification lays down that the samples shall be illuminated by a defined beam of light from an incandescent lamp at a colour temperature between 2 400°K and 2 800°K, incident at an angle of not more than 20° from the normal to the surface of the paper. All the reflected light must be observed by using an integrating sphere or cube. The total area of the three holes (for the sample, entry and exit of the light) shall be less than 4 per cent of the diffusing surface of the integrating chamber. It is further laid down that the walls of the integrating chamber shall have a reflectance between 85 and 95 per cent and shall be white. The photocell, upon which the reflected light falls and the measuring apparatus associated with it, must give a reading proportional to the light emerging from the integrating body and shall have a spectral response approximating to that of the human eye.

The specification was, in fact, framed to include the most commonly used low price, British-made instrument available. This is satisfactory for many purposes, but the integrating chamber is far from an ideal integrating sphere and the photometer leaves something to be desired. A standard instrument was constructed for PATRA during the war years and is still in use for accurate work, but it never went into production owing to lack of demand. The need for a standard instrument is, however, still felt and it now seems likely that the Zeiss Elrepho instrument will be adopted for the purpose.

In order to keep the opacity of paper high, it is essential to make the scattering coefficient as high as possible. This means encouraging the interreflection of light between the fibres of the paper. Anything that tends to decrease this tendency such as wet calendering and excessive beating increases the transparency of the sheet. The opacity can be increased by the use of fillers: the higher the refractive index of the filler, the greater is the scattering coefficient and the more effective it is. It is now well known that titanium dioxide is about the best available filler for giving opacity to paper. The opacity of paper can also be increased by increasing the absorption coefficient. This, however, will impart a distinct colouration to the sheet or at least grey it appreciably. There are limits therefore to what can be accomplished in this direction. Generally speaking, of two sheets of identical structure, the darker one will appear more opaque than the lighter.

Before leaving the subject of opacity, it is well to note that anything that fills the pores of the paper with transparent liquid will tend to reduce the opacity. Printing inks containing oils that penetrate deeply into the sheet have the effect of reducing locally the opacity of the paper in the areas printed: thus, the tendency of print to show through to the reverse side of the sheet is increased. This effect is known as strike-through. It is not only governed by the properties of the paper, but also by the ink used and the printing conditions. It is most noticeable with newspaper printing.

Colour and whiteness

SPECIFICATION of the colours of papers has always been necessary in the printing and papermaking trades and, as the near-whites form a particularly important group, the need is constantly felt for some single-figure measure of whiteness or brightness to indicate how nearly printing papers approach the ideal white.

A review of existing methods of measuring and expressing colour with particular reference to the near-whites was made by me in 1938(6) and again in 1942.(7) Despite the passage of nearly 25 years, the position has not altered fundamentally. Progress has been rather in matters of detail—firstly, the development of improved instruments for the measurement of reflectance values; secondly, a continued search for methods of expressing whiteness in terms of a single figure; thirdly, adaptation of colorimetric methods to include papers containing fluorescent or optical bleaches. The inclusion of such fluorescent materials greatly complicates normal colorimetry.

Despite the fact that it departs from the recommendations made by the International Commission on Illumination for illuminating and viewing specimens, there is a lot to be said for the method in which the specimen is illuminated diffusely and viewed normally or, conversely, illuminated normally and viewed diffusely. These conditions are recommended in the British Standards specification for measuring the printing opacity of paper.

Moreover, adoption of such conditions enables calculations to be made on the basis of the Kubelka-Munk equations.

An important theoretical and practical study of the influence of the optical geometry and absorption coefficient on diffuse reflectance values has been made by Stenius.(8) Stenius measured the reflectance of a number of papers using four different geometries. These were—(*1*) nearly normal incidence and diffuse viewing as given by the General Electric recording spectrophotometer; (*2*) normal incidence and 45° viewing; (*3*) 60° incidence and diffuse viewing; (*4*) diffuse incidence and normal viewing as in the Elrepho reflectometer. These different geometries give different values for the reflectance. Only the fourth geometry—diffuse incidence and normal viewing—gives reflectance values that are consonant with the Kubelka-Munk theory; the divergence in the four methods increases as the colour of the specimen becomes more pronounced—that is, as its absorption rises. There is a great deal to be said for choosing a geometry that does give results in accordance with the Kubelka-Munk theory, since, if this is so, the values for the absorption and scattering coefficients can be calculated from the reflectance values and, once these coefficients are known, the reflectance of sheets made from the same stock (but of different thickness) can be calculated with a fair degree of accuracy.

The Elrepho reflectance photometer conforms to these desired conditions of diffuse incidence and normal viewing fairly closely; it is also photometrically accurate and has good sensitivity. An important instrumentation study of this photometer was made by Dearth, Shillcox and Van den Akker,(9) who, in addition to carrying out a thorough appraisal of its performance, compared the reflectance values with those obtained on the General Electric recording spectrophotometer. Appreciable differences in the two sets of results were found, which could be attributed in the main to differences in the optical geometry of the two instruments. The discrepancies are likely to become greater with more glossy papers.

Probably, because of its close conformity to theoretical requirements, the Elrepho is the instrument around which the present SCAN standards for opacity and brightness are being written. It is likely that international standards will employ this instrument rather than the EEL opacimeter or the GE brightness meter.

Whiteness

A RECENT comprehensive survey of the problems connected with the description and measurement of white surfaces was made by the Inter-Society Colour Council Sub-Committee on problem 19, White Surfaces.(10)

The report deals with the physical and psychological properties of white surfaces and the various ways in which such surfaces can depart from pure white. Instruments available in the U.S.A. for measuring near-whites are reviewed, as are the main formulae propounded for assessing whiteness. As colour is a three-dimensional property, it is theoretically impossible to express in a single number the way in which a colour departs from pure white. Nevertheless, in practice, most papers are in fact desaturated yellows of lightness somewhere between 70 and 95 per cent. Thus, a certain simplification is possible and the fact that whiteness figures can be made to work at all depends on this simplification. Unfortunately, of the many formulae proposed, the simple ones convenient for practical work turn out to be inaccurate or misleading, whereas those that seem to be more promising from the psychological point of view are invariably too complicated for practical work. What is well established, however, is that a small increase in the yellow component of a paper is far more noticeable than a general reduction in reflectance in which the neutrality of the colour is maintained. This is undoubtedly the reason that the addition of a small quantity of blue pigment or dye to a yellowish stock may improve its whiteness, even though the physical reflectance of a paper has been reduced.

The best of the various whiteness formulae are critically compared by Hunter in a recent paper.(11) Those propounded by Hunter, Judd, Selling and MacAdam are basically similar, though they differ in the values of the constants adopted. Finally, Hunter recommends the following equation for general use—

$$W = 100 - \left\{ \left[\frac{220(G-B)}{G+0{\cdot}242B} \right]^2 + \left[\frac{100-G}{2} \right]^2 \right\}^{1/2}$$

where G and B are the reflectances through green and blue filters, respectively, obtained in an instrument described. The green-minus-blue reflectance difference corresponds to the yellowness of the sample and receives about four or five times the weight of the green reflectance alone in determining the visual appearance of whiteness. Hunter's paper is probably the best study that has been made of the subject to date, but it will be seen that the form of equation recommended is not well adapted to general use, though W may be determined graphically from the given values of G and B.

Optical bleaches

THE introduction of optical bleaches to papermaking pulps in recent years has, of course, complicated colorimetry considerably. The action of these optical bleaches is well known: they absorb light in the ultra-violet region of the spectrum and re-emit it as blue light in the visible. This blue light is added to the yellow light reflected by the paper itself and the results are a greatly improved neutral or blueish white. In this way, they differ fundamentally from the ordinary blue dyes that are added to paper stock, which can do no more than to depress the yellow end of the reflectance curve, whilst maintaining that at the blue, producing a more nearly neutral colour, but one of lower reflectance throughout the visible spectrum.

Unfortunately, the use of optical bleach is not without its disadvantages. The first of these is that the optical bleaches are seldom permanent, but fade in strong light, so that the original poor colour of the paper is soon restored. The second drawback is that the action of the bleach depends upon the ultra-violet content of the incident light. They are ineffective in light containing little ultra-violet; thus, the appearance of the paper will change in a marked manner according to the quality of the illumination under which it is viewed—that is, according to whether it is in north skylight or an artificial light. Generally speaking, its colour in artificial light will be poor. A detailed study of the action of these fluorescent whitening agents and the measurement of their relative efficiency has been made by Allen.(12) He explains that fluorescent whitening agents may improve the appearance of cloth or paper in two ways—the first, a blueing effect, changes the shade of the paper away from yellow towards blue; the second, the lightening effect, is responsible for increasing the luminance of the sample generally. The effect of a change in colour towards blue is much more noticeable visually than a general increase in luminance for any fluorescent whitener. Allen goes on to suggest various ways in which the relative efficiency of fluorescent whiteners may be measured.

The colorimetry of fluorescent materials presents two serious difficulties at the moment. The first is that no standard source of light with an agreed ultra-violet content has yet been set up (the CIE Standard 'A', 'B' and 'C' sources are not specified for their ultra-violet content). The second—and greater—difficulty is that, when such surfaces are illuminated by monochromatic light, the reflected light is no longer monochromatic, but there is an extended spectrum; therefore, the reflecting properties of the surface cannot be expressed adequately by means of a plane diagram. Theoretically, their colours can be expressed completely only by means of a solid spectrophotometric diagram in which the X axis represents the wavelength of the incident light, the Y axis the wavelength of the reflected light and the Z axis the intensity. Work along these lines was done some years ago by Donaldson,(13) but it is extremely time-consuming and almost useless for practical purposes. The line of approach that is therefore almost universally adopted is to use some standard source of known ultra-violet content and to analyse the light reflected from a sample both with and without the ultra-violet light component of this source incident upon the specimen. This can be achieved by means of an ultra-violet filter in the incident beam.

An illuminant for the colorimetry of fluorescent materials has been suggested by W. Harrison.(14) He stresses that the colorimetry of specimens containing fluorescent whitening agents requires an illuminant with ultra-violet content to at least 3 600 Å. Single sources based on a filament lamp do not satisfy this requirement. He suggests the use of two sources: one is equivalent to Illuminant C, provided by a filament lamp screened with an O.B.8 filter; the second is the same lamp screened with an O.X.1 or modified O.X.1 ultra-violet filter. (These filters are supplied by Chance Brothers Ltd.) The practical arrangement is to employ interchangeable filters over the same lamp and take the readings consecutively; the readings are added together. The O.X.1 filters transmit a certain amount of deep red, which may be objectionable. This may be removed by combining the filter with a thin layer of O.B.10 filter. The O.B.10 filter also absorbs some ultra-violet, but the effect of this absorption can be reduced by raising the colour temperature of the lamp to 3 000° K. The arrangement finally adopted in Harrison's fluorescence measuring unit is to use the lamp at 3 000° K with an O.B.8 filter, giving Illuminant C chromaticity, supplemented by the ultra-violet light through an O.X.1 plus O.B.10 filter. The lamp is a 12 volt, 100 watt projector lamp slightly underrun at 3 000° K.

The Harrison fluorescence measuring unit has been examined by J. M. Adams.(15) It can be used either as a colorimeter for fluorescent materials using the light source just described or as a fluorimeter in which the sample is illuminated by the light from a mercury vapour lamp at a wavelength of 365·5 millimicrons. The fluorescent light excited by this radiation can be analysed by means of the filters provided with the instrument.

J. M. Adams(16) has recently described a modification of the Sheen abridged spectrophotometer so that it may be used with fluorescent papers. The ordinary tungsten filament lamp can be replaced by a high pressure xenon arc lamp and a small fan is added for cooling purposes. The ultra-violet content of this arc is rather high, but is conveniently reduced by means of an ordinary glass filter, 2 mm thick. The Sheen abridged spectrophotometer is found in practice to give readings close to those obtained from a prism spectrophotometer and the filters are fitted in the path of the reflected light, so that any difference in the spectrophotometric curves when the xenon arc and tungsten lamp are used can be attributed to fluorescence. This has been confirmed in practice, since non-fluorescent specimens give practically the same curve for both illuminants, whereas strongly fluorescent boards or papers give appreciably greater reflectances in the blue and green regions of the spectrum. The differences in the curves show the degree of whitening given by an optical bleach and how the colour of a fluorescent material is affected by the ultraviolet content of the lighting under which it is viewed.

A somewhat similar investigation using a modified Elrepho photometer was carried out by Friele.(17) Friele reached the conclusion that the Elrepho photometer equipped with a xenon arc lamp and suitable filters was adequate for colour measurements on optically bleached materials. He found that there was wide difference of opinion from one observer to the next about the whiteness grading of such materials and found further that most of the whiteness formulae already advanced broke down badly when the materials concerned were fluorescent. For practical, single-figure grading of whiteness, he reached the conclusion that the TAPPI brightness method was as good as any, although it needed some revision; but this method failed completely when the departures from white were in the green or reddish direction. In an appendix to his paper, Friele advanced a new formula for whiteness that gave better agreement with experimental results. He concludes, however, with the significant sentence, 'The formula is therefore only given to stimulate further thoughts on whiteness evaluation and cannot be recommended for use, the confusion with respect to this subject being already intolerable.'

It seems, therefore, that we are far from the end of the road so far as whiteness measurements and the evaluation of optically bleached materials are concerned.

Gloss

GLOSS may, perhaps, be loosely defined as the degree of polish seen on the surface under examination. Although important, it is no easy property to define accurately or to measure and was the subject of a monograph published in 1945.(18) This summarised all the known work on the subject to date; since then, a number of papers have been published that have carried our knowledge a good deal further.

Physically, gloss is closely connected with the reflection of a collimated beam of light that falls upon a paper surface. Some of this light is reflected from the fibres in the surface of the paper, as if these fibres constituted an assembly of minute mirrors oriented at all possible angles to the surface. Because of the method of formation of the sheet and particularly if it is supercalendered, there is a preponderance of fibres oriented near the plane of the sheet, so that some degree of specular reflection takes place. The rest of the light, however, penetrates more deeply into the paper and is reflected and refracted from the fibres in a way that has already been explained. Some of this light emerges again from the surface, but probably in a very different direction from that of incidence. Consequently, there is a great deal of diffuse reflection. The total of the light reflected from the surface is thus made up of specularly reflected and diffusely reflected components, but it is impossible physically to distinguish them and no complete analysis of the problem has been made.

The question of which gloss meter should be adopted for practical work can only be answered experimentally. The technique of the PATRA group of workers has been to have a series of papers of varying degrees of gloss ranked by a group of independent observers. This ranking can usually be established with a fair degree of confidence and the problem then resolves itself into finding some physical measurement or combination of measurements that will rank the papers in the same order as the observers. This goal has only been partially achieved. The first paper in this series was concerned with the study of the Ingersoll glarimeter.(19) It was found that, although there was a broad general relation between the Ingersoll readings and visual assessments of gloss of a series of papers, there were important discrepancies that could not be attributed to sampling, instrumental or personal errors. It was concluded that the glarimeter is not a satisfactory gloss meter except for rough work and routine checking. This work was next extended to cover the Bausch & Lomb and Sheen gloss meters.(20) Measurements were also made on the papers with a goniophotometer and the measurements were used to calculate the various gloss numbers that had been suggested in the literature from time to time. It was found that none of these was entirely satisfactory. For the range of papers examined, the best grading was given by direct measurement of the intensity of the specularly reflected light. The angle of illuminating and viewing the specimens influenced the grading considerably, the best results being obtained at 45°, not the 60° or 70° frequently used in commercial gloss meters. The best rank correlation was obtained with the goniophotometer set at angles of incidence and reflection of 45°. The next best was given by the Sheen gloss meter, which operates at the same angles, but has less accurately defined incident and reflected beams. Other methods gave less satisfactory results.

These observations were followed up further and a later paper(21) showed that the luminance factor of the paper (that is, its brightness) affected the visual assessment of gloss quite appreciably, the darker papers appearing more glossy for the same specular reflectance. The empirical formula below was eventually found to give good correlation with visual assessments for a range of low gloss and medium gloss papers—

$$G = S(a+10/L)$$

In this formula, G is the measure of visual gloss, S is the relative intensity under 45° conditions of illuminating and viewing, L is the luminance factor and a is a constant found by trial to be 0·2. A somewhat similar empirical formula can be derived for use with the Sheen gloss meter.

It is seen that, even for the comparatively easy case of low gloss and medium gloss papers, two parameters are necessary in order to express the gloss in satisfactory terms. When we come on to the problem of high-gloss papers, the task is much more difficult. A study of high-gloss, machine-glazed papers gave serious difficulties.(22) The difficulty of obtaining satisfactory agreement between observers increases as the gloss of the surfaces increases and as the colours and surface textures of the papers to be compared diverge. Moreover, alterations in the conditions of illuminating and viewing the specimens may bring about a considerable shift in the ranking of certain papers. It was concluded that it is impossible to establish any gloss scale that will be satisfactory to all observers under all conditions of illuminating and viewing. The most that one can hope to do is to set up a convention that will be reasonably satisfactory to a majority of observers under average commercial conditions of judging papers and it must be recognised that individual disagreements with this convention will be inevitable and frequent.

It was found further that the visual assessment of high-gloss papers appears to be influenced by the 'peakiness' of the polar reflection curve. Curiously enough, very sharply peaked polar reflection curves seem to cause a downgrading by the observers of the papers concerned. As a result of laborious trial and error, the following empirical equation was found to fit the experimental results tolerably well—

$$G = S(0{\cdot}390 + 0{\cdot}325 \tanh 20/L) - 100 \tanh [0{\cdot}1155\,(P-1)/100]^3$$

Here, G, S and L have the same meanings as before, P is a measure of the peakiness, here arbitrarily defined by the ratio of the intensities at the specular angle and at 5° nearer the normal than the specular angle. The equation has, of course, no physical significance and, although it fitted our results quite well, it is doubtful whether it is of universal application and, in any case, is rather too complicated to be of practical use. A further complication is that many papers are of uneven finish, consisting of high gloss islands surrounded by regions of lower gloss. Such papers tend to be ranked rather lower by observers than the measurements of a goniophotometer would suggest.

Our views on gloss meters based on all this work were summarised in a paper read to the Oil & Colour Chemists' Association in 1953.(23) In this paper, the whole problem was reviewed and certain recommendations were put forward for the design of gloss meters. These recommendations are not, so far as I know, observed in any commercial gloss meter available today.

Apart from this group of papers, little interest in gloss measurement appears to have been aroused in Europe and the U.S.A. Hopkins and Highton,(24) however, describe a modification of the Ingersoll glarimeter in which the amount of polarised light is measured photoelectrically. The observer errors associated with the earlier instruments were therefore removed and the reproducibility of measurements improved. The basic objections to the use of the Ingersoll method established by our earlier work remain.

The only other extensive work on gloss measurement done within recent years is by a group of Japanese workers whose work here is too little known.

Those seeking to explain the optical properties of paper in terms of the structure of paper will be interested in the analysis attempted by Barkas,(25) in which the light scattered from the surface of paper is broken down mathematically into specular and diffuse components. Barkas's treatment is geometric and assumes the existence of specularly reflecting and diffusely reflecting facets in the surface of the paper, inclined at all angles to the surface. The specular reflecting facets probably correspond to individual fibres in the surface of a sheet; the diffuse facets are a mathematical device to express the effects of multiple interreflections and refractions within the surface of the paper before the light finally emerges from the surface. Barkas showed that his analysis was applicable to certain low-gloss surfaces and this conclusion was at first confirmed by the PATRA workers. Investigation of papers of higher gloss, however, showed that the analysis soon broke down, as some of the components had negative values. The same conclusion has been reached in a more recent investigation by Kurita, Yano, Nara and Hasunuma.(26) Their main conclusions are that, for highly glossy papers, the analysis breaks down, as it gives negative values for the diffuse components; although the analysis is formally admissible for less glossy papers, there is no sound physical ground for assuming the existence of two independent components—specular and diffuse—as the specular component varies considerably with the absorption of the paper. The authors state that the reason the analysis does not explain the diffuse reflection phenomena is because the so-called specular reflection does not follow Fresnell's formula. From this and other work, it is evident that the reflection characteristics of paper cannot be explained simply in terms of geometrical optics.

Some interesting work on the reflection of polarised light from paper has been carried out by Tanaka.(27) When light polarised in the plane of incidence is reflected from the surface of paper, part of it remains polarised and the rest is completely depolarised. It can be assumed that the polarised component is specularly reflected and the depolarised component diffusely reflected. In this way, it is possible to isolate the two components and measure their relative intensity. Tanaka concluded from his experiments that this assumption was tenable. He found that with coated paper the shape of the intensity distribution curve of the diffusely reflected component is circular and closely obeys Lambert's Law. With uncoated paper, the shape diverges somewhat from the circular. The main substance to produce the diffuse reflection is clay. Tanaka found further that the regularly reflected component does not follow Fresnell's law of reflection; he considered that this is because reflection from paper cannot be explained by geometrical optics and, consequently, Barkas's analysis cannot be applied to paper. He gives also an explanation of the experimental fact that the direction of the maximum intensity of reflected light is not always the same as the direction of specular reflection.

In the second paper,(28) Tanaka reports measurements made on the reflection characteristics of printing paper, both white and dyed with red ink, made with white and monochromatic beams of light. The magnitude of the specular component of coated paper is hardly affected by dyeing, which gives credence to his theory. For uncoated paper, the polar curve deviates somewhat from the circle predicted by Lambert's Law and the specular component has some colour. That the specular component does not follow Fresnell's formula for coated papers, he suggests may be the result of the interference effect of fine clay coated on them. He studied the effect of calendering on the reflection characteristics and found, as expected, that calendering increased the specular component and decreased the diffuse component to nearly the same extent.

Because of its complicated structure, paper is not an ideal material for measurements to be subjected to theoretical analysis and Tanaka later made similar measurements on ground glass as a reflecting surface.(29) Ground glass may be assumed to have a large number of reflecting facets oriented at all angles, the diffuse component arising from multiple reflections between these. Tanaka found that when the surface is not too rough and the angles of incidence and reflection are not too great, the specular component follows Fresnell's formula. However, when the surface is finely ground and the angles of incidence and reflection are large, the phenomenon of *sheen* is observed. He found that the distribution of the relative area of mirror facets having various inclinations to the mean surface resembles a normal distribution, but the kurtosis is rather larger. When the angle of incidence is about 70°, masking of the mirror facets by adjacent facets is already appreciable. The diffuse component is attributed mainly to multiple reflections. The maximum value of the diffuse component has a linear relation to the logarithm of the root mean square height of the roughness and the larger the angle of incidence or the coarser the roughness of the surface, the larger is the value of the diffuse component. This work is carried further by a second paper.(30) Using the apparatus and methods previously reported, he measured the reflection characteristics of several kinds of ground glass and again found that the distribution of the relative area of the mirror facets having various inclinations to the mean surface resembles a normal distribution, but the kurtosis is larger. When the angle of incidence and reflection is greater than 50°, maskings of the mirror facets occur and the coarser the surface, the greater becomes the masking. If the distribution of the mirror facets is represented by a normal distribution function having a standard deviation of about 18° and the masking by an exponential function, the reflection characteristics of ground glass can be adequately explained by the combination of these two functions. This is an interesting paper, though the conclusions are probably not applicable to paper surfaces on account of their much greater complexity of structure.

When light falls upon a matt surface, in general the reflected light is diffused in all directions. Near glancing incidence, however, some of the light is specularly reflected causing a lustre that is noticeable at glancing angles and is often known as *sheen*. This phenomenon will be familiar to those who have used matt distempers. The surface so treated appears matt until it is illuminated and viewed at near glancing angles, when a gloss appears, which is usually reddish in colour. The phenomenon of sheen has received attention by the Japanese group of workers. Hasunuma and Nara(31) report that the relation between the critical angle of the regular reflection θ and the root mean square value of the surface roughness of metals and glasses is—

$$\cos \theta = k\lambda/2h_a$$

where λ is wavelength, h_a and k are constants defined by the profile of the surfaces. The constant k is approximately $\frac{1}{3}$ for glasses and steels, $\frac{1}{2}$ for chromium-plated surfaces. This is explained in a later paper,(32) in which the observed formula is derived from a theory similar to Kirchhoff's diffraction theory. Scattered light is separated into specularly reflected and diffusely reflected components. The specularly reflected component corresponds to sheen and, if certain assumptions are made, its intensity can be calculated.

The authors carry these studies a stage further in a third paper.(33) Taking curves of intensity against angle by means of a goniophotometer, they found that strongly reflected light was observed only beyond the critical angle and then in the vicinity of the angle of specular reflection. This component is considered to be the cause of the sheen gloss.

Hasunuma and Nara investigated also the scattering of light from chromium-plated aventurine surfaces.(34) The results were analysed on the assumption that the surfaces are made up of randomly distributed small facets, the scattering of light being due to regular reflection from these facets.

Nara(35) described a new gloss meter that determines the *distinctness of image* gloss of samples. This is done by comparing the intensity of light reflected at the specular angle to that reflected at an angle deviating slightly from the specular angle (by about 45 seconds of arc). Angles of incidence ranging from 40° to 80° were investigated. It is claimed that this new gloss meter generally gives results in accordance with visual judgment, but it is obvious that it would only be of use for very highly polished surfaces. It is stated that it is not suitable for bent or curved surfaces, since the curvature produces errors.

An interesting paper on the gloss of machine-glazed papers was published by Nishiwaki. (36) This is virtually a continuation of an investigation of machine-glazed papers by the PATRA group of workers—in fact, samples of the same papers were used for measurements. The set of papers used was regraded by 36 Japanese observers and some discrepancies with the British and Swedish gradings were noted. These discrepancies may well have been due to fading or alteration of gloss of the papers concerned with time. The method of measurement was based upon a method described in a previous paper by Fukushima. In principle, the goniophotometer is used with the illuminator and receptor axis kept at a constant angle, in this case 90°, throughout the experiments. The surface of the specimen is slowly rotated so that its normal gradually approaches the receptor axis from the initial position of specular reflection. In this way, the intensity of the light, which is initially at a maximum denoted by I_0, decreases rapidly at first, then more slowly as the angle of rotation increases. For many papers, the slowly decreasing part of the curve is found to be expressed approximately by $I_2 \exp(-\beta\theta^n)$, where the two constants I_2 and β depend on the sample and the other constant n on the optical geometry of the goniophotometer (in the one used, n approximated to $n=2$). If, therefore, $\log I$ is plotted against θ^n, the slowly decreasing part of the curve is approximately a straight line and can be extrapolated backwards to $\theta=0$. This extrapolated value is denoted by I_2, which is assumed to be due to the diffuse component of the reflected light. The quotient I_0/I_2, where I_0 is the intensity at the specular angle, is assumed to be a measure of contrast gloss and is denoted by Fukushima by the symbol *Gnf*. Following the practice of the PATRA workers, Nishiwaki plots log *Gnf* against the visual assessments of his observers and claims that a linear relation results, with the exception of two papers that appear to have been graded much too high visually. The correlation coefficient between the visual assessments and the gloss number was 0·986. This is a valuable paper in which no claim is made to finality.

Nishiwaki(37) has carried out a similar examination of a set of coloured papers, including some white. These included kraft, unbleached sulphite, glassine, flint and photographic papers. He again found that log *Gnf* was correlated with the visual gradings of 13 observers, except for three white papers that appeared anomalous. The 13 observers chosen were inexperienced and the anomalies with the white papers disappeared when gradings were made by two experienced observers. Nishiwaki(38) has published two further interesting papers on the effect of colour on visual gloss. Specimens used were coloured papers wound round a glass cylinder about 6 cm in length and 4 cm in diameter, overlaid by one layer of transparent ciné film developed without being exposed. The specular reflectance from these specimens was substantially the same in all cases, but the visual glossy appearance differed. The specimens were ranked by a panel of ten observers and a factor δ defined by $\delta = 2 - \log Y + 10\Delta$ was found to correlate very highly with the rankings by visual evaluation. In this equation, Y is the luminance factor and Δ the chromaticity difference from neutral on the MacAdam UCS diagram. It will be noted that δ is not a gloss number in itself; it has the nature of a correcting factor for the colour of the specimens. It shows that the visual gloss increases with the darkness and saturation, that is, with the deepness of the colour. In an extension of this work,(39) Nishiwaki examines the effect of hue on gloss and reaches the conclusion that hue has very little effect on visual gloss under ordinary viewing conditions. The texture of the surface has, however, a significant effect on the visual gloss, although further work is needed to establish the effect of texture quantitatively.

Summary

It will be seen from this survey that measurement of the opacity of papers is approaching finality, in so far as anything in science and technology ever approaches finality. A standard method of specification has been laid down and instruments in conformity with this specification are either available or being prepared. Moreover, it is possible to express opacity in terms of optical constants of the paper that are related directly to the structure and optical properties of the fibres.

When we come to gloss, the position is nothing like so satisfactory. A considerable amount of research work on gloss has been done in various parts of the world, most of which is ignored by the designers of gloss meters. Comparison of the relative gloss of papers closely similar in colour, texture and composition is fairly straightforward; comparison of papers that differ widely in colour and texture is a problem of great difficulty to which no complete answer has yet been found. In the meantime, those interested in gloss measurements may reconsider with advantage the recommendations laid down in the author's earlier paper(23) and the methods of Prof. Fukushima and co-workers also deserve a trial.

For colour measurement, satisfactory instruments are in use, but no final agreement has yet been reached about the best way of measuring the colours of the increasing number of papers containing optical bleaches. The controversy about measuring whiteness of papers continues; the choice lies between formulae that give reasonably good results, but are complicated to apply, and those that are simple, but misleading or of very limited application. In any case, the research worker still has plenty to keep him occupied.

REFERENCES

1. Harrison, V. G. W., *Proc. Tech. Sect. B.P. & B.M.A.*, 1940, **21**, 67–173
2. Kubelka, P. and Munk, F., *Zeitschrift f. Techn. Physik*, 1931, **12**, 593–601
3. B. S. 2923: 1958—Printing Opacity of Paper
4. Harrison, V. G. W., and Poulter, S. R. C., *Brit. J. appl. Phys.*, 1950, **1** (1), 13–17
5. Colour, opacity and gloss sub-committee, *Proc. Tech. Sect. B.P. & B.M.A.*, 1955, **36** (3), 617–639
6. Harrison, V. G. W., *PATRA Research Report No. 2*, 1938; *PATRA Research Report No. 3*, 1939
7. Harrison, V. G. W., *Proc. Phys. Soc.*, 1942, **54**, 86–98
8. Stenius, Åke S:son, *J. Opt. Soc. Amer.*, 1955, **45** (9), 727–732
9. Dearth, L. R., Shillcox, W. M. and Van den Akker, J. A., *Tappi*, 1960, **43** (2), 230A–239A
10. Hunter, R. S., *J. Opt. Soc. Amer.*, 1958, **48** (9), 597–605
11. Hunter, R. S., *J. Opt. Soc. Amer.*, 1960, **50** (1), 44–48
12. Allen, E., *J. Opt. Soc. Amer.*, 1957, **47** (10), 933–943
13. Donaldson, R., *Brit. J. appl. Phys.*, 1954, **5**, 210–214
14. Harrison, W., *J. Sci. Instr.*, 1959, **36** (5), 234–236
15. Adams, J. M., *PATRA Printing Laboratory Report No. 36*, 1960
16. Adams, J. M., *Printing Tech.*, 1960, **3** (2), 5–16
17. Friele, L. F. C., *Die Farbe*, 1959, **8** (4/6), 171–186
18. Harrison, V. G. W., *PATRA*, 1945
19. Harrison, V. G. W., *Proc. Tech. Sect. B.P. & B.M.A.*, 1947, **28** (2), 443–481
20. Harrison, V. G. W., *J. Sci. Instr.*, 1949, **26** (3), 84–90
21. Harrison, V. G. W. and Poulter, S. R. C., *Brit. J. appl. Phys.*, 1951, **2**, 92–97
22. Harrison, V. G. W. and Poulter, S. R. C., *Research*, 1954, **7** (4), 128–136
23. Harrison, V. G. W., *J. Oil Col. Chem. Assoc.*, 1953, **36** (400), 569–579
24. Hopkins, L. F. and Highton, A. P., *Proc. Tech. Sect. B.P. & B.M.A.*, 1955, **36** (2), 339–354
25. Harrison, V. G. W., *Brit. J. appl. Phys.*, 1950, **1** (2), 46–53
26. Kurita, T., Yano, H., Nara, J. and Hasunuma, H., *J. appl. Phys. Japan*, 1955, **24** (8), 318–324
27. Tanaka, S., *J. appl. Phys. Japan*, 1956, **25** (5), 207–213
28. Tanaka, S., *J. appl. Phys. Japan*, 1958, **27** (10), 600–604
29. Tanaka, S., *J. appl. Phys. Japan*, 1957, **26** (3), 85–91

30. Tanaka, S., *J. appl. Phys. Japan*, 1958, **27** (12), 758–762
31. Hasunuma, H. and Nara, J., *J. appl. Phys. Japan*, 1953, **22** (11/12), 389–392
32. Hasunuma, H. and Nara, J., *J. Phys. Soc. Japan*, 1956, **11** (1), 69–75
33. Hasunuma, H. and Nara, J., *J. appl. Phys. Japan*, 1955, **24** (11), 457
34. Hasunuma, H. and Nara, J., *J. Phys. Soc. Japan*, 1957, **12** (10), 1 117–1 122
35. Nara, J., *J. appl. Phys. Japan*, 1957, **26** (9), 452–454
36. Nishiwaki, J., *J. Phys. Soc. Japan*, 1957, **12** (1), 53–57
37. Nishiwaki, J., *J. appl. Phys. Japan*, 1956, **25** (9), 370–374
38. Nishiwaki, J., *J. appl. Phys. Japan*, 1959, **28** (5), 267–272
39. Nishiwaki, J., *J. appl. Phys. Japan*, 1959, **28** (5), 272–276

Transcription of Discussion

DISCUSSION

PROF. R. H. PETERS: To what extent is the Kubelka-Munk function obeyed for dyed papers? In the region over which the function is obeyed, have you calculated the absorption spectra of the dyestuff and, if so, does the result show reasonable agreement with spectra measured in solution? Have you had any experience with spectrophotometers other than the GE spectrophotometer? If so, how do the reflection spectra differ from those obtained on the GE instrument?

DR. V. G. W. HARRISON: I have not myself done work on the Kubelka-Munk equations. Quite a lot of work has been done in the past and this is adequately reviewed by Judd.[1] I think the answer yes can be given to your first question, at least in part. On the reflection spectra side, we have had experience with a number of spectrophotometers and colorimeters that we have in our laboratory. In our work, we use a Uvispek spectrophotometer, which is a modification of the Beckman spectrophotometer. This is slow, but reasonably satisfactory. We have also a couple of photoelectric colorimeters that express the colours in terms of CIE units with tolerable accuracy. There are still differences among instruments that have yet to be resolved.

DR. A. B. TRUMAN: In view of your plea for someone to develop further the work of Dr. Barkas, may I ask for details of the references to Barkas's work?

DR. HARRISON: I am sorry not to have given them explicitly, but they can be found in one of my references(25) and in his main paper.[2] I am very well aware that universities have not got an enormous reservoir of students, but Barkas's analysis is to me a particularly irritating problem, because it does seem to be amenable to mathematical treatment, yet it is difficult to find someone with the time to do it.

MR. N. C. UNDERWOOD: Could the well-established work on least detectable colour differences be extended to the fields of opacity and gloss?

When measuring the opacity of a sheet of poor formation, the scale of

---

[1] Judd, D. B., *Colour in Business, Science and Industry* (John Wiley & Sons Inc. and Chapman & Hall Ltd., 1952), 314–350

[2] *Proc. Phys. Soc. (Lond.)*, 1939, **51**, 274–295

the variations may be smaller than the currently used measuring apertures. How will the observed reading correlate with practice when the subjective results depend on the lowest rather than on the highest opacity?

DR. HARRISON: Some work on the least perceptible differences in opacity was done before the war by Farebrother.[3] Speaking from memory, something like $\frac{1}{2}$ per cent is probably a practical, fairly well established limit to what can be detected. I do not know for gloss; there are extraordinary differences of opinion about the glossiness of the surface of very highly finished papers and those differing in colour. It is somewhat better with low gloss papers, but with those that Steenberg sent us, we had some suggestion of a bimodal distribution of visual gradings, showing sharp differences according to whether one judges the high spots on the surface or takes a general mean over the whole surface. I think you are right about the formation effect and that the weak spots in the paper are taken into account rather than the total opacity: the mean might not necessarily give therefore the information you are looking for.

THE CHAIRMAN: Would Mr. Farebrother care to add a word?

MR. T. H. FAREBROTHER: In the opacity range over about 90 per cent contrast ratio, the least perceptible difference was found to be less than 1 unit under the best possible conditions for comparison. My recollection is that the value found was 0·7 unit.

DR. J. A. GASCOIGNE: In the paper industry, we are plagued with trying to relate every reflectance to GE values: what is Dr. Harrison's opinion upon reflectance standards provided with various instruments? Opal glass secondary standards are supplied for the Elrepho instrument and I wonder whether Dr. Harrison thinks these are better or worse compared with paper standards, as the latter are considered more realistic in some laboratories.

DR. HARRISON: These opal glass standards can form quite a reasonable secondary standard for reflection measurement, provided they are handled carefully, washed and looked after. While not a primary standard, they form a much more convenient working standard of rather greater permanence than paper.

PROF. H. W. GIERTZ: A question of interest when dealing with the opacity of paper is whether light is scattered only by the external surfaces of the fibres or also by internal capillaries and hollow spaces.

---

[3] *Proc. Tech. Sect. P.M.A.*, 1937, **18** (1B), 147–171

DR. HARRISON: I think it is very likely. Anything that will scatter or absorb light, whether from the surface or within the fibres or between the fibres, will contribute to the opacity and colour of the sheet and irregularities that have been very apparent in the photomicrographs have as important a part to play in light scattering as the interaction between the fibres themselves.

PROF. GIERTZ: May I ask Dr. Van den Akker what he thinks about it?

DR. J. A. VAN DEN AKKER: We have no experimental evidence on the point, but I am of the opinion that the submicroscopic capillaries are too small to scatter a significant amount of light.

DR. HARRISON: Whatever these very small obstructions may be, they will scatter also by defraction.

MR. H. W. EMERTON: It needs to be emphasised that the dimensions of the pores within the fibre wall are very much smaller than many of those between fibres and are only a fraction of the wavelength of light. The scattering in this case will therefore be of the Rayleigh type.

PROF. G. JAYME: There is a great deal of strong evidence that the capillaries in the cell wall of late wood fibres are considerably smaller than in early wood fibres. This could be proved, for example, by secondary fluorescence.

MR. D. H. PAGE: We have some direct evidence of this effect of yield on scattering within the fibre. When we started this work on bonding, we used polarised vertical illumination with the object of getting sufficient contrast to see the bonded area. We told Kallmes about this and he managed to see the bonded regions without polarising the light. We could hardly believe this, because we could not see the bonded areas in any great detail with unpolarised light. We went to the trouble of taking our microscope along to his laboratory (because he said his microscope was better than ours) and it turned out of course that the difference was not in the microscopes, but in the pulps. With our pulps, polarised light was essential, but with Kallmes' pulp it was not, due apparently to a difference in the optical perfection of the body of the fibre. We were using a whole range of commercial furnishes, but I believe that his were never-dried fibres from a lightly cooked pulp of relatively high yield.

DR. O. J. KALLMES: Yes, that is true.

MR. PAGE: I think this shows that there must be a real effect of chemical treatment and possibly drying on the internal scattering power of individual fibres.

DR. KALLMES: The opacity and brightness of paper originate primarily from the free fibre lengths, especially in the case of softwood pulps. A preliminary attempt to calculate relative bonded area from geometric considerations is given in our second contribution to this symposium.

MR. Z. J. MAJEWSKI: Papers with a high degree of fibre orientation show a higher density: their opacity is thereby reduced.

DR. VAN DEN AKKER: An observation made by Howard Leech is of considerable interest here. He dried fibres by the solvent replacement technique, which leaves the structure of the fibre in an expanded condition (as determined, for example, by the gas adsorption technique). He compared the appearances under a microscope, using dark field illumination, of fibres dried in this way with ordinary water-dried fibres and observed much stronger light scattering from the expanded fibres. In other words, this is a case in which some of the microscopic fissures in the fibre are big enough to scatter light. It would be very interesting for someone to continue this kind of study and make observations corresponding to various stages of expansion of the fibre wall.

DR. HARRISON: I have no further comment to make, except that it has been very gratifying to me to see how these different properties of paper are beginning to link up. Optical studies may give a little information on the structure of the sheet and, on the other hand, the opticians have much to learn from those engaged on strength properties.