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TRANSACTIONS AND COMMUNICATIONS

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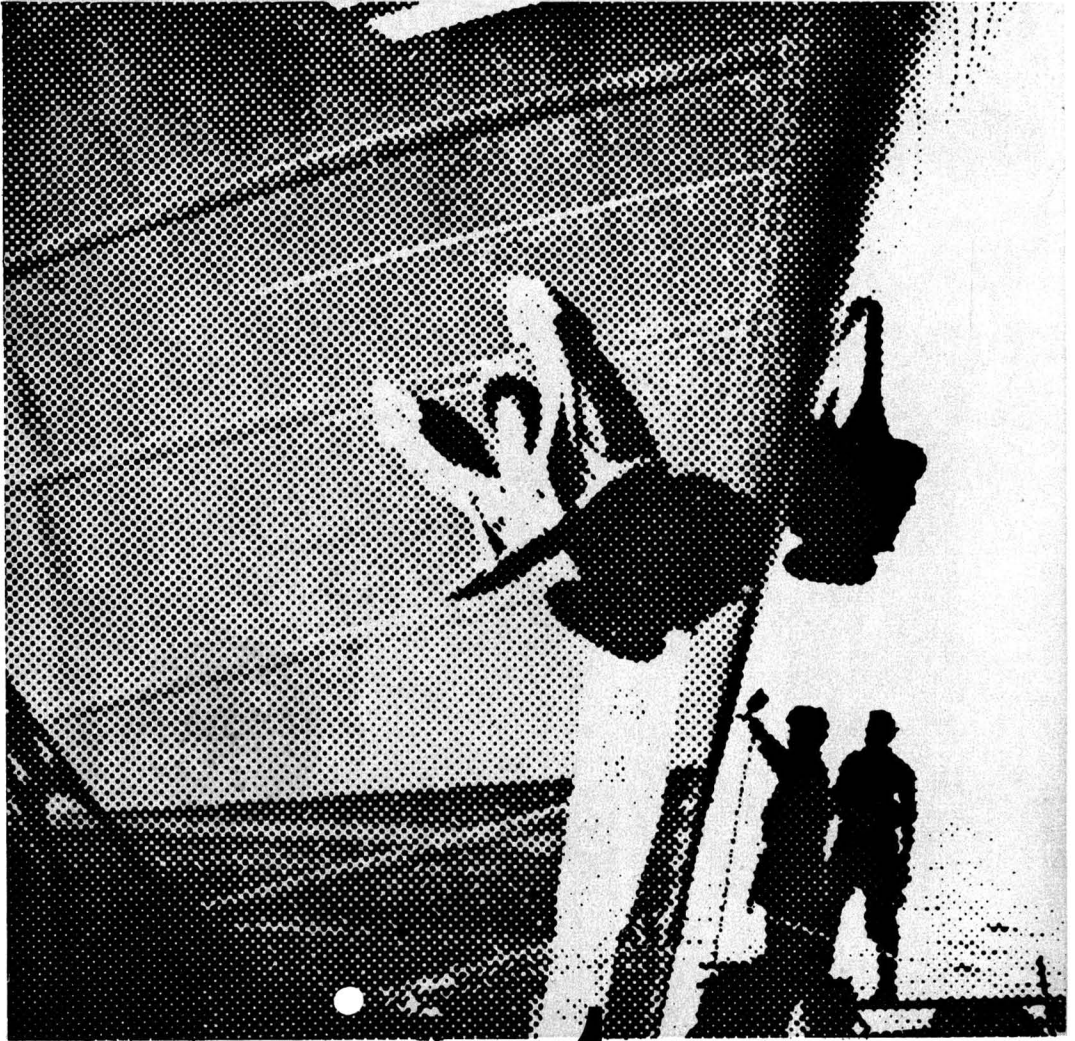
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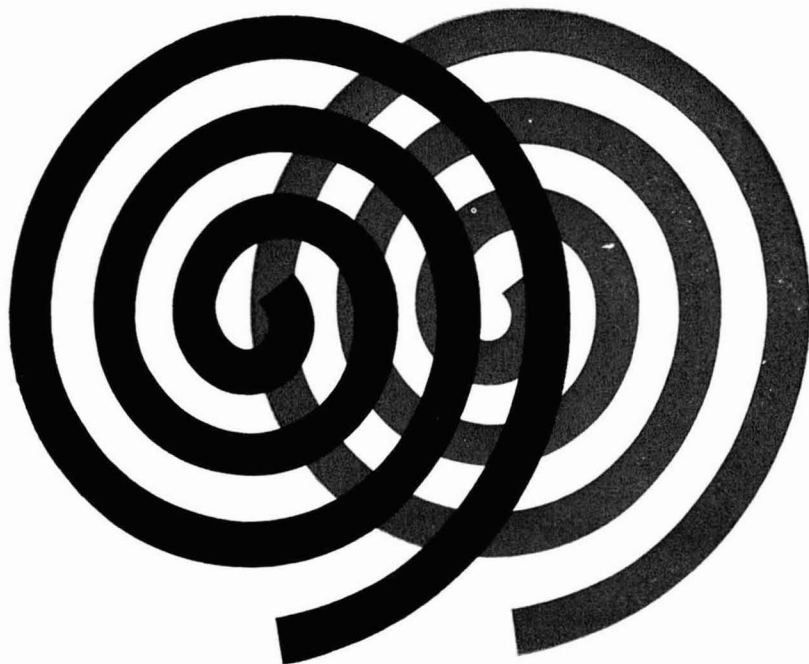
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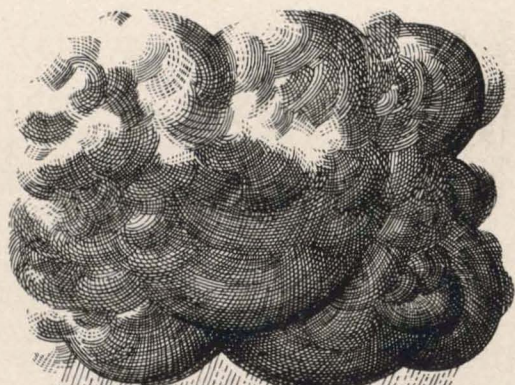
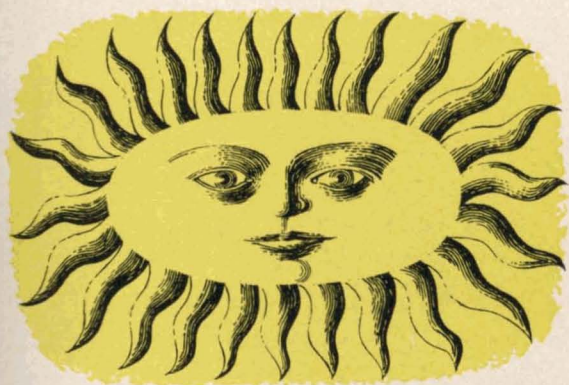
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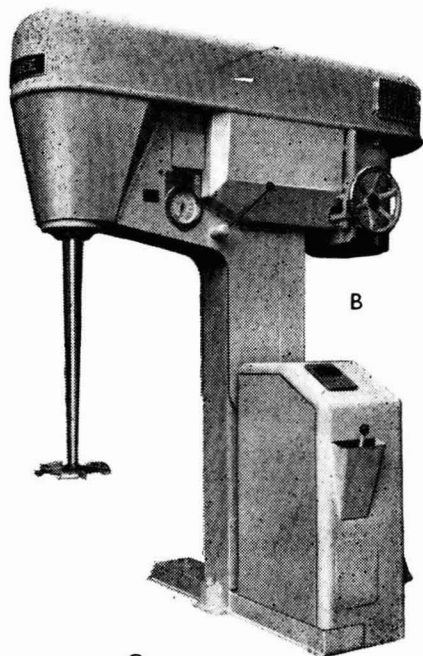


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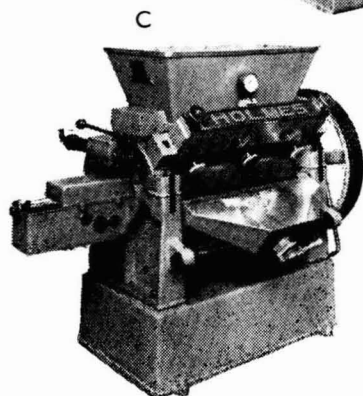
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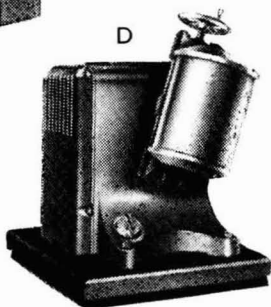
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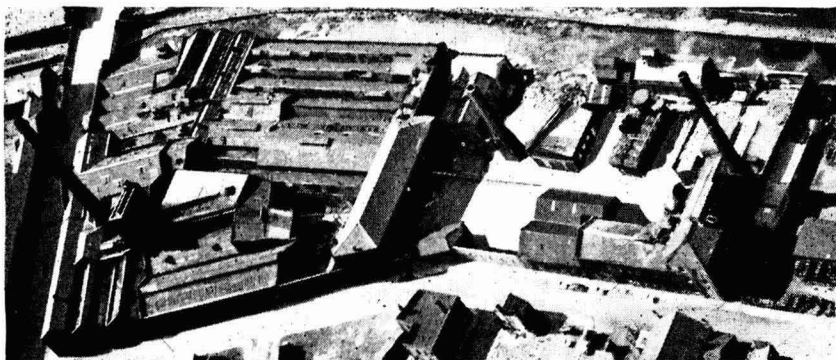
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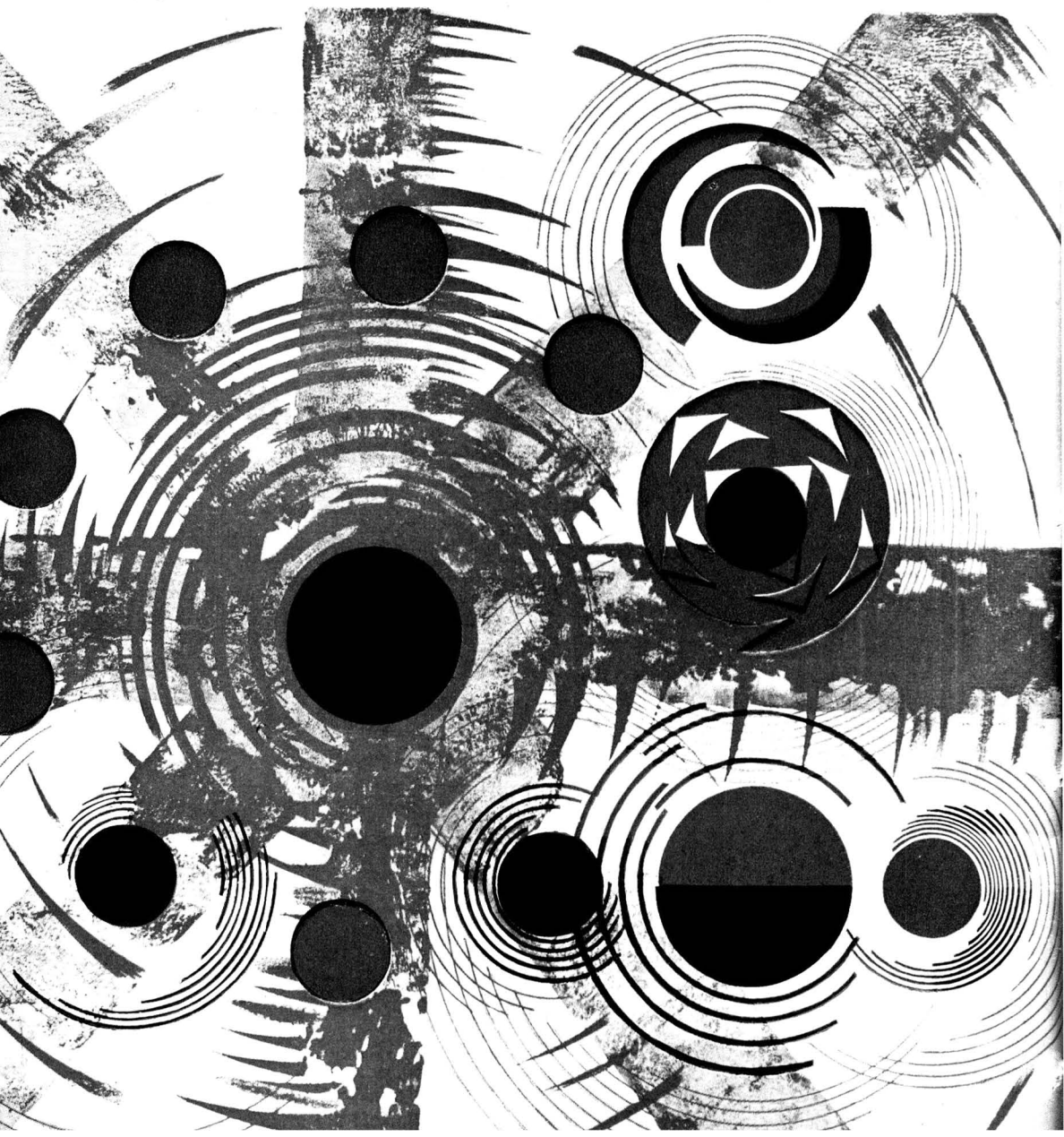
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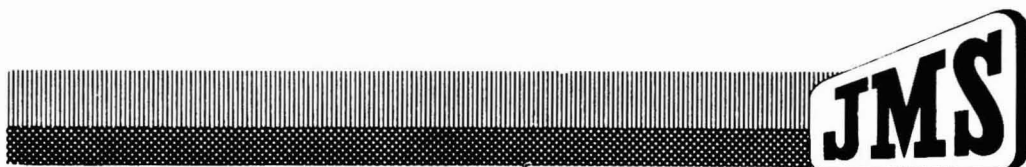


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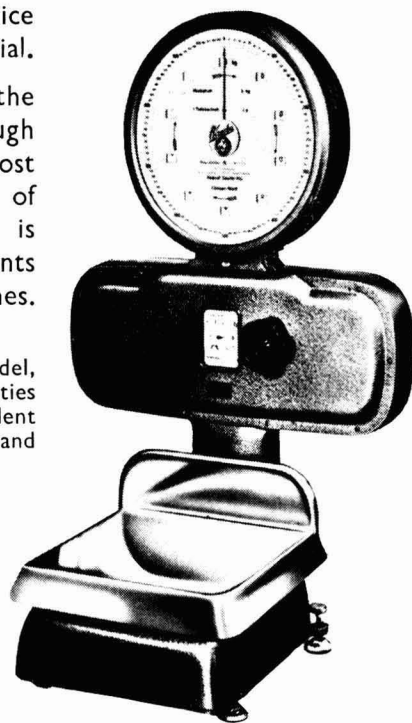
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1 Colour Matching Lamps should imitate—

North-sky daylight

Total daylight (i.e., sunlight + skylight)

2 If north-sky daylight is preferred, which curve should be chosen?

Illuminant C

Abbot-Gibson 7,400°K

Any observations on any aspects of colour-matching lamps will be welcome.

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Colour-matching Lamps— An Enquiry Into Members' Wishes

EXPLANATORY NOTE

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The colour-rendering properties of any light source are defined precisely by its spectral energy distribution, that is, by the proportion of the total energy emitted by the lamp at each wavelength in the visible spectrum. The energy distribution in the ultraviolet region is also important where fluorescent colours or fluorescent brightening agents (optical bleaches) are present; the lack of sufficient ultraviolet radiation is a common failing among some present daylight lamps and should be rectified in a new specification.

There is no such thing as an "ideal" spectral energy distribution for detecting small colour differences, except possibly an equal-energy light source, which would radiate equal amounts of energy at all wavelengths between specified limits. However, such a light source is of academic interest only, since it does not correspond to any natural light.

A *standard* light source which could be universally accepted would, however, be of great value. Many of the problems of colour matching arise from the existence of metameric pairs, that is, pairs of patterns which appear to have the same colour under one light but no longer match under another type of illumination. The existence of a standard reference lamp would minimise disputes as to whether two patterns are an acceptable match, although such disputes would not be eliminated, since differences in the colour vision of individual observers can be as important as differences in illumination.

The purpose of an "artificial daylight lamp" is to imitate some aspect of daylight, and the main difficulty arises in defining the kind of daylight which is to be imitated. It is obvious that the colour of the light coming from a brilliant blue sky is different from that from a grey, overcast sky, while the light in late evening is different from that at mid-day. Discussion of the different phases of daylight is simplified by the use of "colour temperatures". When any substance, such as a piece of iron, is heated in the dark, it first becomes visible as a dull red colour, which then changes to orange and finally to white, or bluish white, as the temperature is raised. The colour of the light can therefore be defined in terms of the temperature of the source—the bluer the light, the higher the temperature.

The natural light normally used by professional colourists is that coming from a north-facing window or skylight, but even here the colour temperature varies widely, from about $5,000^{\circ}\text{K}$ on a very dull day with an overcast sky to over $15,000^{\circ}\text{K}$ on a clear day with a blue sky. North-sky daylight is chosen by colourists not so much for its colour, but because it is fairly constant in intensity, direct sunlight being intermittent and glaring.

On the other hand, in everyday use, coloured materials are viewed under illuminations which differ even more widely in colour temperature, from ordinary tungsten light at about $2,600^{\circ}\text{K}$ to north-sky daylight at over $15,000^{\circ}\text{K}$. In addition, modern developments in fluorescent and discharge lighting introduce further complications, as the spectral energy distributions of these lights are different from any phase of daylight or tungsten light. Naturally, the special conditions of viewing under artificial light do not directly affect the specification of an "artificial *daylight* lamp". It is probably true to say that the most common conditions under which the majority of coloured materials are viewed is "total daylight", which is a combination of light coming directly from the sun together with light coming from the rest of the sky.

Surprisingly enough, the colour quality of total daylight is fairly constant with a colour temperature of about $6,000^{\circ}\text{K}$. Such light is just as good as any phase of north-sky daylight for detecting small colour differences, and its more constant colour makes it much superior to the north-sky light used by colour matchers. If a metameric match cannot be avoided, then, if the patterns are arranged to match under a lamp conforming to the spectral energy distribution of total daylight, they will still match under most daylight conditions. Furthermore, total daylight has a colour temperature which lies in the middle of the wide range of colour temperatures encountered in daily life.

The first question to be answered, therefore, is whether an artificial daylight lamp should be designed to imitate either (a) total daylight or (b) north-sky light. If a decision is made to imitate total daylight, the specification should be relatively simple. Total daylight has the same spectral distribution as sunlight outside the atmosphere, and there is a proposed standard curve of such radiation (Moon, *J. Franklin Inst.*, **230**, 583 (1940)); this curve extends into the ultraviolet, and lamps conforming to this distribution would be suitable for the visual examination of fluorescent material. The colour temperature of this light is 6,200°K.

On the other hand, if north-sky daylight is preferred, then there are two spectral-energy distribution curves which might be imitated by an artificial daylight lamp. These are, first, the standard (Illuminant C) established by the International Commission on Illumination (C.I.E.) in 1931, which has a colour temperature of 6,740°K, and, second, a distribution computed by Abbot and Gibson as representing a moderately overcast north-sky daylight with a colour temperature of 7,400°K. The chief merits and demerits of these two possibilities can be briefly summarised as follows—

Illuminant C

Advantages

- (1) It is a widely used standard source for colorimetry, and is likely to remain so for many years.

Defects

- (1) It is seriously deficient in ultraviolet radiation as compared to any phase of daylight, and cannot be used for the inspection of fluorescent materials.
- (2) The spectral energy distribution of Illuminant C is determined by a set of liquid filters used in conjunction with a tungsten filament lamp. Although this illuminant was intended to represent north-sky light, the actual energy distribution is determined partly by the liquid filters employed, and there seems to be no cogent reason why a daylight lamp, which almost certainly will not employ liquid filters, should be required to duplicate this particular curve.

Abbot-Gibson 7,400°K

Advantages

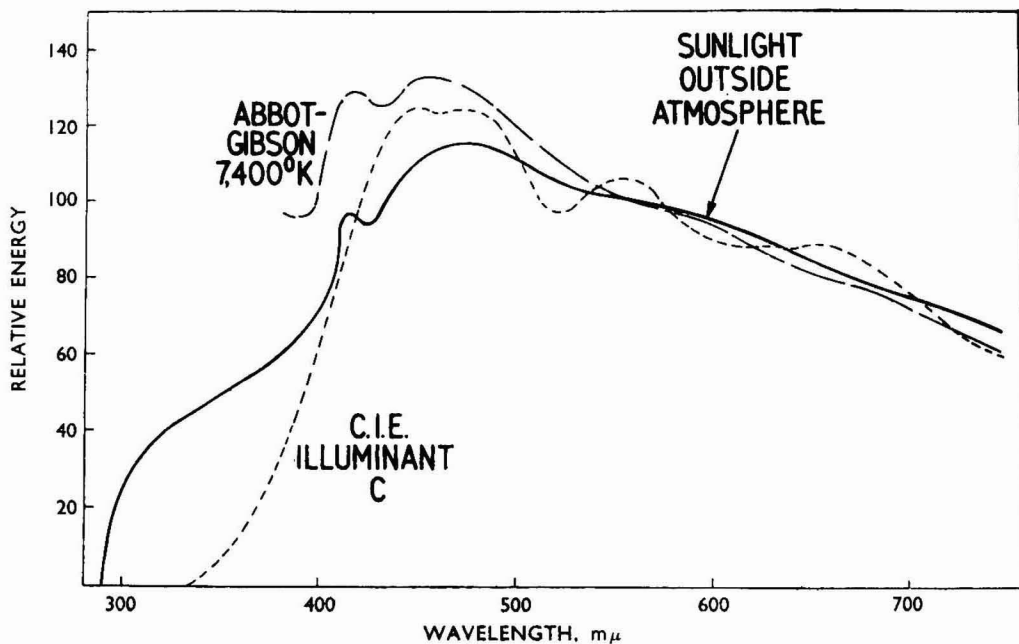
- (1) The energy distribution defined by this curve has been found to approximate more closely than Illuminant C to a moderately overcast north-sky daylight in the U.S.A., and this is probably true also in the U.K. and in other countries.

- (2) The curve is computed from measurements of solar radiation and not of a particular light source; although at present it extends down to only 380 millimicrons ($m\mu$), it could probably be extended to 300 millimicrons, which would define the ultra-violet characteristics for the examination of fluorescent specimens.
- (3) The curve has been adopted as a standard for colour-matching lamps in the U.S.A.

Defects

- (1) No convenient tables are yet available whereby spectrophotometric curves could be converted into colorimetric data representing the appearance of samples under this illuminant. It is doubtful, however, whether differences between the calculated values obtained from the Abbot-Gibson curve and those obtained from Illuminant C would be of practical significance.

The three spectral-energy distribution curves discussed are illustrated in the diagram.



ADDITIONAL NOTE ON THE PAINT INDUSTRY

By Miss D. L. Tilleard

How does the question affect the paint industry? There is a trend in the industry towards using artificial sources of light for colour checking, largely because these can be maintained constant, and are available for 24 hours of the day, if need be. High-power tungsten lamps, with blue filters, are available for this purpose but their power consumption is high and for lighting large areas the more efficient fluorescent lamps are commonly used, frequently with additional tungsten lamps to provide more energy at the red end of the spectrum. The fluorescent lamps do not have the smooth distribution of energy through the spectrum which is obtained with daylight, there being strong mercury lines, and the Paint Research Station has consistently recommended that where any metamerism may be involved, a final check should be made under natural daylight.

In spite of the deficiencies of artificial lighting of this kind it is becoming commonly adopted and closer standardisation of such sources would clearly be of value to the paint industry.

The first step to be taken by the B.S. Committee is to decide on a spectral energy distribution representing the kind of daylight which is considered most suitable for colour matching, and then to ask the lamp manufacturers to reproduce this within agreed tolerances.

The views of the paint industry and O.C.C.A. members are sought by the Committee on whether this chosen type of daylight should represent:—

- (a) North-sky light, which is variable but may on a clear day be very blue (corresponding to high colour temperatures, 7,400°K or higher),
- or (b) Total-sky light, which is less variable, and corresponds to lower colour temperatures, around 6,000°K.

It is traditional in the paint industry to speak of colour matching being carried out ideally under north-sky light, but it seems likely that the only feature of importance is that the glare of direct sunlight should be removed. In practice, the light from a north-facing window will not normally be that from a blue sky, but may include reflection of sunlight from white clouds, or more frequently may be completely diffused, giving a lower colour temperature, probably nearer 6,000°K.

The proposal at present before the committee, viz., to standardise on an energy distribution representing total-sky light (about 6,000°K) rather than one representing north-sky light (7,400°K) offers the advantage that the lower colour temperature is closer to the ordinary conditions of daylight viewing, therefore any metameric matches made under it will be acceptable under most daylight conditions.

Further measurements have now been made in this country on the distribution of energy from north sky and total sky, and once the

desired colour temperature has been established, a smoothed version of one of these curves could be adopted as standard.

Before a final decision is reached there are other aspects of the question which must also be considered. One is the connection with colorimetric measurements. Such measurements are being used in the paint industry to a much greater extent than in the past, and with the development of instrumental matching may become even more important. It would seem to be desirable that such measurements should be made in reference to an illuminant which, if required, could be exactly reproduced in the laboratory so as to provide a visual check on the measurements. At the present time, with Illuminant C as the standard source, this is possible; colorimeters are calibrated to give results in terms of Illuminant C and tables and computers are available giving results in terms of Illuminant C, and this illuminant can be set up, to a reasonably close approximation, with a tungsten lamp and liquid filters.

The present demand for a source with a higher content of ultraviolet, particularly to deal with fluorescent materials, may result in the next few years in a C.I.E. decision to adopt either a modified form of Illuminant C or a completely new standard illuminant with the desired ultraviolet characteristics. Whatever the decision made it is likely that the same principle will be adhered to, viz., that the source should be exactly reproducible in the laboratory for colour inspection as well as colour measurement.

Whilst there are obvious advantages in such correspondence between measurement and inspection, it is likely that in many types of matching in the paint industry such correspondence may not be a matter of first importance. The industry is concerned mainly with matching to standards, rather than the reproduction of precise colorimetric values. If measurements and calculations are made with Illuminant C and some other illuminant is used for inspection, there are two cases to consider. If the match is completely non-metameric as is frequently the case, it will remain a match under all illuminants and the choice of inspection illuminant is unimportant. If the match is metameric the position is more difficult because the match will vary with illuminant. To correspond with instrumental checks the inspection illuminant should be the same as that specified for measurement, but it may in practice be less important that the match should hold under Illuminant C than under some recognised illuminant chosen to represent average daylight.

It is hoped at the next meeting of the B.S. Committee (December 19th, 1961) to put forward the views of O.C.C.A. members and of the Paint Industry on the matter of the choice of a standard for artificial daylight, and comments from members will be welcomed, in particular as to the first decision to be taken, viz., whether the aim should be to reproduce North-sky light or Total-sky light.



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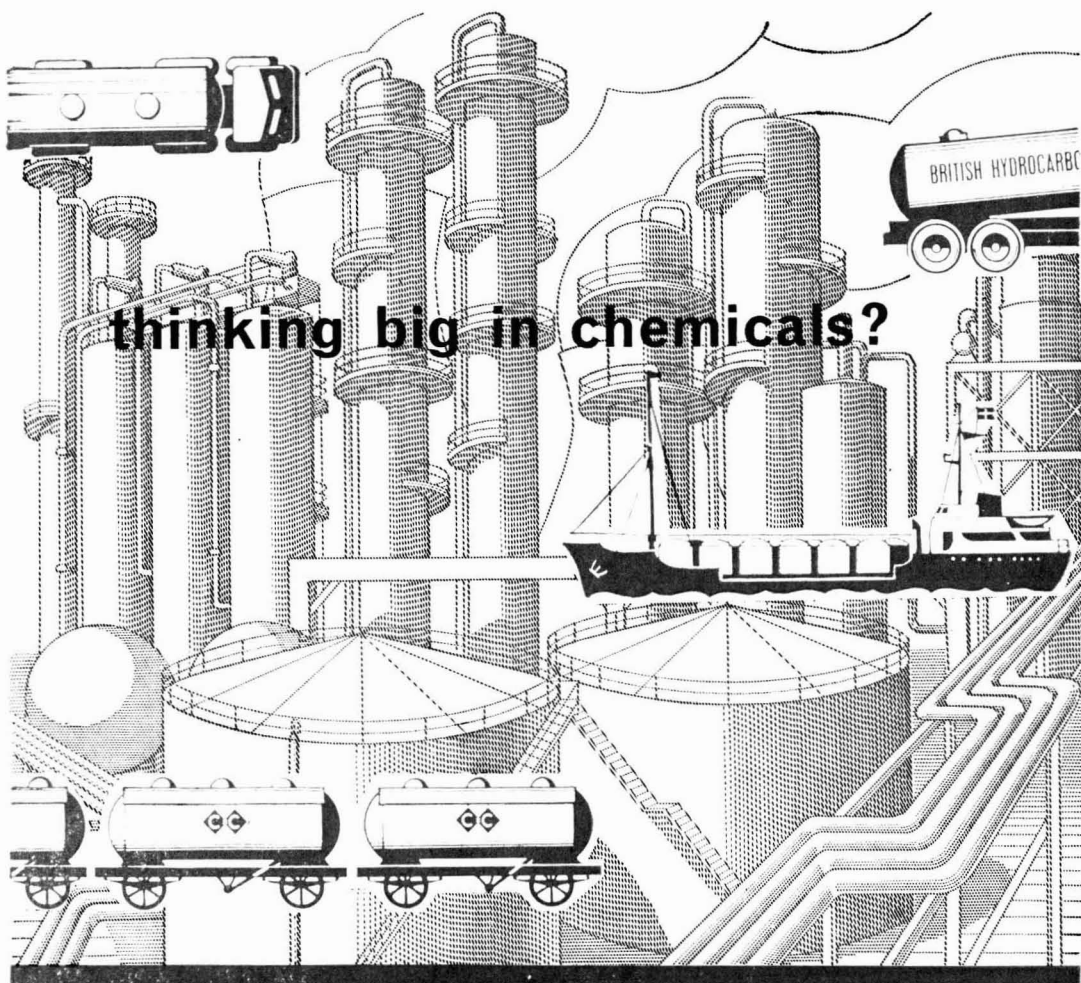
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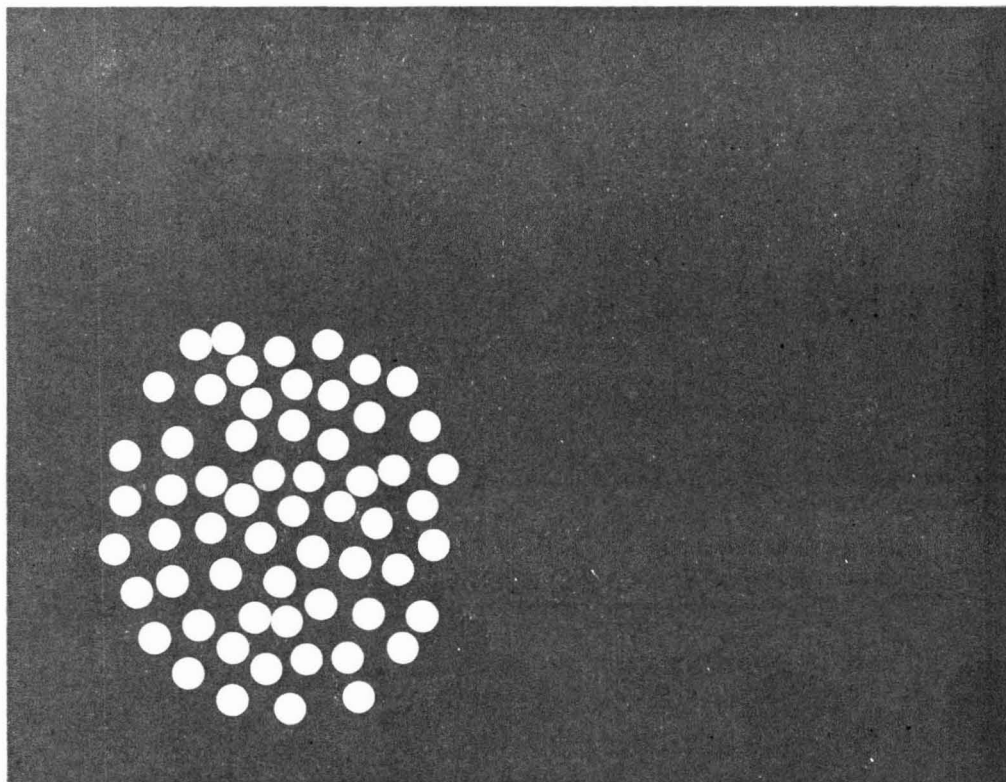
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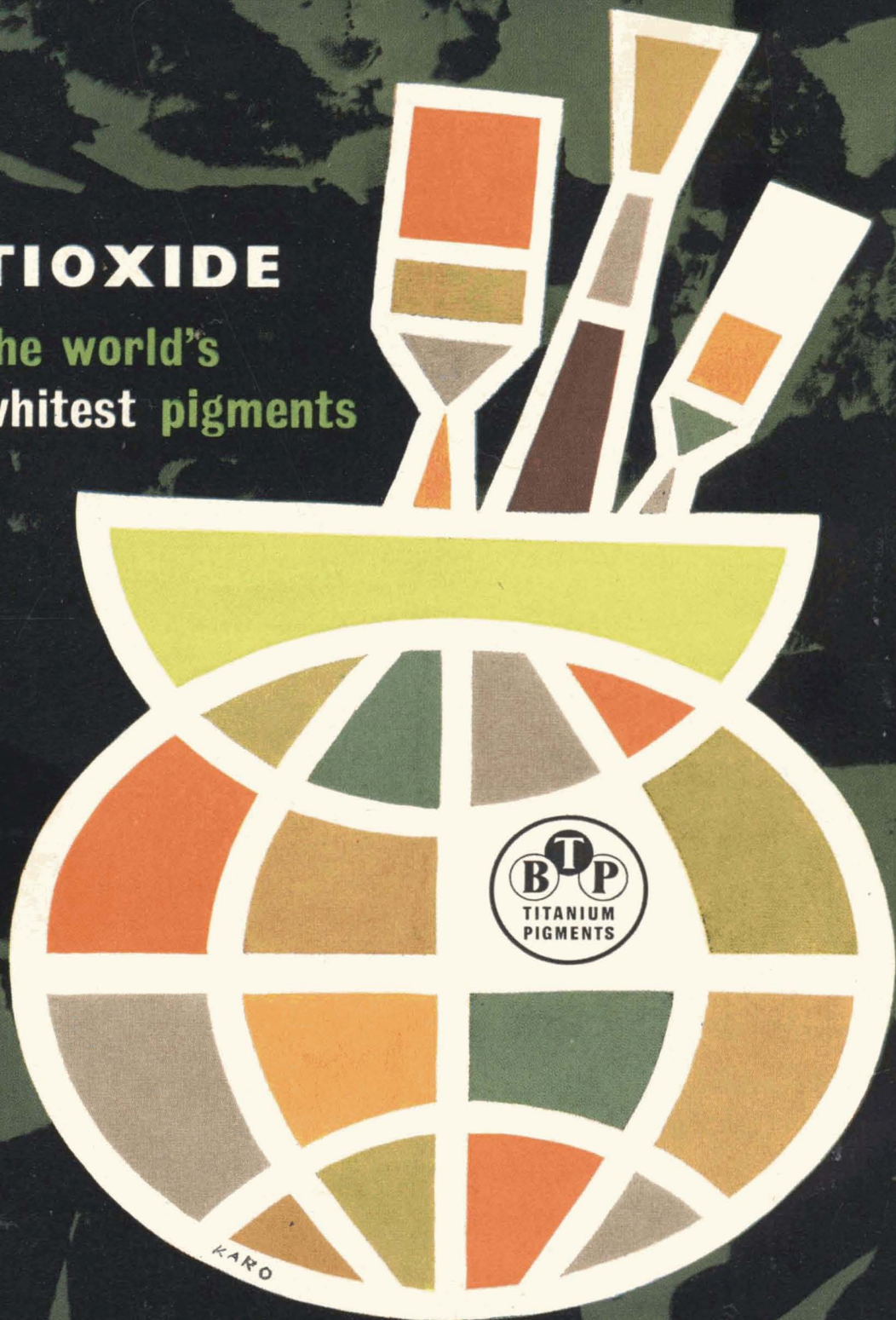
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No. 10

TRANSACTIONS AND COMMUNICATIONS

The Correlation of Service Behaviour of Paint with Observed Physical Characteristics in Air Drying Paint for Structures

By F. G. DUNKLEY and D. P. EARP

*British Railways Research Department (Chemical Services),
Protective Coatings Laboratory, Cavendish House, Park Street, Derby*

Summary

The work described in this paper is a series of *ad hoc* investigations, carried out over a period of some years, into the causes and cure of certain paint failures. In the main the studies are of the characteristics of alkyd resin varnishes and paint media, which arise from the water sensitivity of this class of resin.

The characteristics studied include primarily adhesion, especially inter-coat adhesion, water absorption, and the mechanical properties of extensibility, tensile strength, and recovery. The variation of these properties with different oil length, oil modification, change in polyhydric alcohol used, and the influence of minor additions of other resins, has been studied. Some work on the rapid assessment of paint durability is also described.

INTRODUCTION

The study of the service behaviour of paint and its correlation with the physical properties measured in the laboratory is complicated by the intrusion of the effect of many factors whose influence is unknown, and sometimes, unsuspected. These effects often modify the service performance of a paint, so that results obtained under what are apparently identical circumstances differ widely and the relationship between the observed physical properties and service performance is masked. The influence of surface preparation on paint performance has only to be mentioned for it to be seen how great, but unpredictable, such external influences can be. The study is really one of trends and tendencies.

In the course of the authors' work, probably a unique opportunity is afforded for observing the performance of paint in service, on bridges, railway rolling stock and buildings, in all the varied atmospheric conditions which this country provides. On the scale of the authors' observations, the trends and tendencies, which link up performance in the field with laboratory determined physical properties, are more clearly seen. It is the task of the British Railways Paint Laboratory to determine this relationship and to attempt to define the qualities of the paints used by British Railways in such a way that the most suitable compositions are employed. The work involved brings in the measurement

of physical properties and it has long been the aim of the Laboratory to develop a system of physical tests which would permit a paint to be defined entirely in terms of its physical properties—a situation which does not exist at this stage.

This paper describes a number of discontinuous *ad hoc* investigations which have been carried out, and in fact are continuing, to relate service performance with determined physical characteristics. In most cases, accepting the position as it stands, since the correct interpretation of the physical characteristics of a paint in terms of its performance can only be achieved with a knowledge of the chemical composition, the influence of changes in chemical composition and molecular arrangement has been concurrently studied. The paper describes the methods used, and gives some details of the laboratory work which has taken place over a number of years, and has in some cases enabled modification of the specification to be made in order to eliminate the deficiencies experienced in paint performance.

As far as buildings and structures are concerned, early experience with medium and short oil alkyds showed that while their performance on well prepared surfaces was excellent, an excessive number of flaking failures occurred in practice. This was attributed to the high tensile strength of these media which, when the paints based on them were applied over either wood or steel surfaces, on which loose material was present, caused severe lifting and flaking owing to the strong contraction in drying and ageing. In bad cases this flaking failure occurred within a few months of application, but more often it did not develop until after a year or more. This form of failure was not only premature, but also presented a most difficult surface for refinishing. It was found that by using a low tensile strength copal ester varnish, and reinforcing it with a proportion of alkyd resin (sufficient to give a phthalic anhydride content to the medium of 10 per cent), a durable yet tolerant paint medium was obtained. As the result of the adoption of this reinforced oleoresinous media, flaking failures have largely disappeared, and although some gloss retention has been sacrificed, the ultimate failure in the form of chalking presents a surface which can be readily refinished. A further reason for reducing the glycerol phthalate content of the paint was to avoid the tendency shown by the medium-long and short oil alkyds to give poor intercoat adhesion when the coating sequence is interrupted beyond a period of, say, forty eight hours. Numerous cases of flaking of the enamel coat were experienced when the alkyd media were used without a proportion of a fossil resin varnish.

The same solution of the problem in painting carriages, where the surface is excellent and the conditions under which the paint is applied are controlled, was not appropriate. In these circumstances it was felt that the full potentialities of the long to medium oil alkyds should be exploited, and the proper course to adopt would be to investigate any difficulties which arose, such as adhesion failures, with a view to tracing the cause and relating it to some physical property of the film, which could then be controlled. The method of approach has been fully justified by events and the medium oil alkyd (26 per cent phthalic anhydride) adopted for locomotives has been found fully satisfactory. Increased durability and adaptability to workshop conditions have been the outstanding features of their performance, to which may be added enhanced dirt resistance and resistance to cleaning agents. The same type of medium was selected for

road vehicles, and a four-coat process was employed. The medium for the primer was the same alkyd used for the remaining coats, but it proved to be rather unsatisfactory because of poor chip resistance. This particular difficulty has been overcome by substituting a copal ester based primer, without loss of durability in the system.

A somewhat longer oil medium has been employed for carriages (23 ± 3 per cent phthalic anhydride); initially this was used for both undercoating as well as finishing, but the system had low impact resistance. Taking this to indicate lack of balance between adhesion and cohesion, the undercoating as well as the primer were put on an oleoresinous medium. The performance of the system has been good generally, the durability has been excellent, so much so that the repainting of carriages is no longer determined by the onset of surface degradation, *i.e.* loss of gloss or checking. However, in spite of the improvement effected, the incidence of impact damage caused by barrows and flying ballast, and the development of corrosion at windows, door edges, *etc.*, remains. A spasmodic failure by loss of adhesion will be referred to later.

In the case of wagons, the use of alkyd based paints was allowed only as an alternative to be adopted where their relatively rapid drying characteristics were necessary to accommodate the needs of the production line—a policy dictated by the knowledge that the wagon body plates could not be brought to a rust-free condition. The first choice in the circumstances was a very long oil copal ester medium. This policy has been shown to be correct since, although from laboratory tests on clean plates the intrinsic durability of the alkyds has been found to be at least twice that of the oleoresinous paints, the latter have in practice given better service showing in this performance, their greater tolerance of the poor surface conditions demonstrating the better wetting and adhesion afforded by the linseed oil medium.

Two specific problems have required special study, the first and most important being the difficulty in ensuring satisfactory adhesion for the varnish coating used in revarnishing carriages when alkyd resin varnish is employed. Earlier experience had shown that complete loss of adhesion could occur, and as a result it was decided that it was necessary to use a copal ester varnish. It was felt that this must be regarded as a temporary measure, and because the durability given by the best oleoresinous varnish is much inferior to that of the alkyd resin varnish in use for new or completely repainted vehicles, a quick solution was sought. The second problem is an occasional, but very frustrating, failure of varnish adhesion on the new or completely repainted car. This failure develops quite soon after painting and is quite different from that which sometimes occurs when the enamel beneath the varnish has begun to chalk.

The smog of a few years ago may be remembered because of its effects on delicate chests, but the authors remember that immediately afterwards many complaints were received that the varnish was peeling from the carriages; a programme of investigation was opened. As the result of the investigation which followed, the loss of adhesion was traced to the excessive water sensitivity of the alkyd resin varnish. A similar incidence of failures has not occurred since that time, although occasional failures do occur. Work carried out in the laboratory has shown that the combination of a number of circumstances

is necessary to give rise to actual adhesion failures. At normal times the proportion of vehicles affected, although probably less than 0.5 per cent, is considered to be excessive, but this does not outweigh the other advantages possessed by the alkyd resin systems.

A difficulty encountered in this work was the absence of any acceptable method of measuring adhesion and as a necessary preliminary to the study, a method of carrying out adhesion measurements was necessary, and eventually the method referred to in this paper was developed. The present form of the apparatus represents a compromise, but it does enable a measure of the adhesion to be made. The apparatus will be described in some detail later.

It was felt that it would not be profitable to study adhesion in isolation, and a number of other properties was included in the investigation. The properties thought to be of most direct interest in this connection were water absorption, rate of evaporation of water from the saturated films, extensibility and recovery of detached films. It was an almost inevitable extension of this work that the durability of the varnishes under test should be simultaneously assessed.

TEST METHODS

Water Absorption

The method used for the measurement of water absorption was that described by Kendall¹. The test was applied to varnished tinned steel strips, and also to detached films.

Adhesion

The method of measuring the adhesion was that developed in the Laboratory, and the apparatus employed is an adaptation of the Gardner Tensiometer, the forces being measured by a strain gauge technique or a spring. The instrument used, illustrated diagrammatically in Fig. 1, is a modified form of

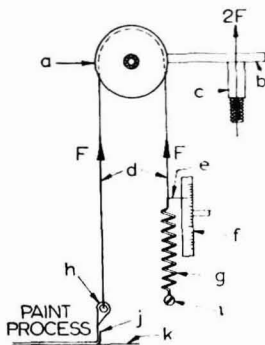


FIG. 1. DIAGRAMMATIC SKETCH SHOWING METHOD OF DETERMINING PAINT ADHESION USING A MODIFIED GARDNER PARKS TENSIO METER

- | | |
|--|-----------------------------|
| (a) Pulley (on ball race). | (e) Indicator arm. |
| (b) Crosshead extension bar fitted to Gardner Parks Tensiometer. | (f) Force extension scale. |
| (c) Screwed rod, motor driven to give constant rate of crosshead rise. | (g) Spring |
| (d) String. | (h) Bar. |
| | (i) Rigid support. |
| | (j) Adhesive tape and film. |
| | (k) Panel. |

the Gardner Tensiometer, from which the standard components other than the motor, gearing and crosshead had been removed. The crosshead is fitted with a side arm which in turn carries a ball-bearing mounted pulley over which a string is passed to transmit the load from the crosshead as it rises to (i) the panel under test, and (ii) the spring on the instrument. Since these forces are equal, any force measured by the extension of the spring is directly related to the actual force exerted by the adhesive tape attached to the paint panel and looped over a bar attached to the string. The measure of the adhesion is taken when the rate of stripping of the film equals the rate of application of the force which is denoted by the steady reading of the pointer attached to the spring on the extension scale. The force is read from calibration curves of the various springs and is the force required to overcome the force of adhesion and is applied at right angles to the paint film.

Reproducibility of the results is good over all ranges where the adhesion of the tape is greater than the adhesion of the paint system, which is obviously the limiting factor of the method, and the figure obtained is that of the adhesion at the weakest point in the system. Normally the strength of the adhesion of the paint system studied is, in the dry condition, greater than that of the adhesive tape to the surface; consequently, "dry" adhesion of systems cannot always be measured successfully. This limitation is not for the purposes of this investigation an important one, since in practice it is only when the film is wet that the adhesion falls to a dangerously low level and adhesion failures develop.

The specimens used in the adhesion tests were prepared by applying two coats of the sample to tinned steel strips and clean, scale-free mild steel panels. In addition, mild steel specimens were prepared by applying the sample as the final coats in the one coat priming paint, one coat undercoating paint, one coat black enamel and two coat varnish system. This system corresponds to the standard process used in accelerated weathering tests for carriage or locomotive varnishes. After ageing for the prescribed period, the specimens were immersed in water at room temperature for periods up to eight days.

The method of test adopted is to attach a length of pressure sensitive tape to the specimen, immediately after its removal from the water, and wiping dry. The speed of rise of the crosshead is adjusted to give a constant rate of stripping ($\frac{1}{2}$ in./min.), the load being applied vertically to the panel surface. In order to standardise the test conditions, the area of varnish covered by the tape is isolated by cutting through to the base metal.

After periods of immersion of the specimen, the adhesion of the tape to the test film is greater than that of the weakest interfacial adhesion of the paint/metal system, and on separating the tape from the specimen, part or all of the film adheres to it. The tape can be filed as a permanent record of the position of failure. This method of removing the film has the advantage that it does not modify the film in any way as could happen with an adhesive and it is preferable, especially for the purpose of investigating interfacial or inter-coat adhesion, to the grid or other cutting devices, which only reveal information about the paint/metal adhesion. The values of the adhesion figures

obtained fall off with continued immersion, but there is often a tendency to give a fairly steady figure before the eight-day immersion period.

When the adhesion test is conducted on a multi-coat system, failures may occur at any level, from the metal/primer interface to that of the final coat. The position of the failure as well as the value of the measured adhesion is of practical importance, the former in test conditions depends to some extent on the speed with which the tape is removed; if the tape is snatched off the failure lies nearer to the surface than if a slow pull is used. The explanation of this seems to be in the ability of the film gradually to dissipate the energy involved through its various levels. An alternative apparatus designed to ensure rapid application of the stripping force similar to the snatch technique has been designed, and measurements have in fact been made on an instrument built on the lines of the *Charpy* Impact test instrument, *i.e.* using a falling pendulum as the source of energy. The force necessary to strip a definite area is assessed by the difference in free energy of the pendulum when falling freely from a definite position and that shown after overcoming the adhesion of the paint film over the test area. This is shown by the maximum rise of the pendulum past the centre point in both cases, the absorbed energy being calculated from the difference obtained. Although this method has been used in a number of experiments the results were not sufficiently reproducible with the available apparatus, but the method appeared to have possibilities, and at a convenient opportunity further consideration will be given to the design of the apparatus and to the interpretation of the results obtained.

Although the main objective of this paper is to describe work on varnishes, it might be mentioned in passing that other *ad hoc* investigations on adhesion using the same technique have been carried out on paints and paint systems. Experiments in connection with assessing methods of surface preparation have been carried out with interesting results, but they have shown that the problem is extremely complex. It would seem that adhesion is subject to influence in quite unexpected ways; for example, the adhesion of priming paint is apparently affected by pigment particle size (indicated by the Hegman gauge) rising to a maximum at 4-5 and falling off with decreasing particle size. Among other factors observed is the increase in adhesion given by refined linseed oil when substituted for boiled linseed oil in primers, and even small additions of white lead or zinc oxide modify the adhesive properties of priming paints. In many cases the effects noticed were of considerable magnitude and although the reasons for them are not clear, some avenues of investigations were suggested.

Among the earlier experiments a series of varnishes submitted by various manufacturers for approval against railway specifications were examined with the results given in Table I. These results seemed to indicate a possible relationship between adhesion and water absorption properties. The relative orders of merit for film adhesion and the inverse order of merit for water absorption have in fact a Spearman ranking coefficient of 0.75. Unfortunately, those varnishes which contain sufficient phenolic and rosin derivatives to give significant improvement in adhesion were later shown to have poor durability, but in the meantime, encouraged by these results to pursue the line of investigation, further work was carried out with a series of varnishes of which rather more information on the composition was available.

TABLE I
PRELIMINARY RESULTS SHOWING POSSIBLE RELATION BETWEEN FILM ADHESION AND
WATER ABSORPTION DATA

Description of Varnishes	Water absorption data (8 days' immersion)				Adhesion	
	Attached film		Detached film		Stripping Force (8 days' immersion) (g./in.)	Inverse order of merit
	Amount absorbed (%)	Order of merit	Amount absorbed (%)	Order of merit		
Oleo-resinous	19.3	10	34.2	10	510	10
Standard alkyd	5.5	2	6.5	4.5	30	5
Alkyd "A"	8.9	7.5	6.4	4.5	20	3
Alkyd "B"	7.2	3	7.1	6	15	1
Alkyd "C"	9.3	9	4.9	2	20	3
Styrenated alkyd	8.3	4	9.0	8	100	7
Modified alkyd (rosin/phenolic)	8.6	5.5	5.4	3	40	6
Modified alkyd (10% P.A. rosin/phenolic)	8.9	7.5	8.7	7	220	8
Modified alkyd (phenolic)	3.7	1	3.2	1	20	3
Tung oil/phenolic	8.7	5.5	18.6	9	300	9

Durability

The durability of the materials was assessed on two coats of varnish applied to the standard painting systems using an accelerated weathering equipment, a detailed description of which, and its correlation with outdoor weathering, has been given in detail elsewhere^{2, 3, 4}, the total exposure being fourteen weeks.

A method of drawing up a durability rating of the materials was devised using a "Points System" based on the assessment of gloss, checking and splitting. In order to allow for the different speeds of the onset of these failures and the different rates of deterioration, points were allocated at the quarter, half and three-quarters life inspections, as well as the final inspection. The final order of merit was based on the summation of the total points thus awarded, the lower the points awarded the better the performance. The system of points is given in Table II. In addition, a further twenty points was added for each additional type of failure, such as loss of adhesion, and presence of rust staining if such abnormal behaviour was experienced. The film was rated as "good" if no failure occurred, while the rating "poor" was intended to coincide with the condition at which repainting would be necessary in service.

The various materials were classified into four main groups as shown in the summarised classification of durability results (Table III). Materials placed

TABLE II
ALLOCATION OF POINTS FOR FAILURES

Rating	Film property under consideration		
	Gloss	Checking	Splitting
Good.. ..	0	0	0
Good/Fair	4	2	4
Fair	8	4	8
Fair/Poor	16	8	16
Poor	24	12	24
Poor/Bad	32	16	32
Bad	40	20	40

TABLE III
CLASSIFICATION OF MATERIALS WITH REFERENCE TO DURABILITY
(ASSESSED ON BLACK PROCESS)

Group 1			Group 2			Group 3			Group 4		
Ref.	% P.A.	Description	Ref.	% P.A.	Description	Ref.	% P.A.	Description	Ref.	% P.A.	Description
A3	22.5	Glycerol/linseed	A2	23.0	Glycerol/linseed	A1	22.0	Glycerol/linseed	D3	28.0	P.E./linseed/tung
A4	22.0	"	C5	36.0	Glycerol/soya bean	B1	20.0	P.E./linseed	E1	34.0	Glycerol/linseed with rosin
A5	22.0	"				B2	20.0	"	E2	19.0	P.E./linseed/D.C.O. with rosin
C1	27.0	Glycerol/linseed/tung	E8	25.0	Glycerol/linseed/tung	B3	22.1	"	E3	19.0	Glycerol/linseed/tung with rosin/phenolic
C4	31.0	"	F2	27.0	Glycerol/linseed/tung with 16% styrene	B4	22.1	"			
D1	21.0	P.E./tobacco seed				G5	25.0	Glycerol/linseed	B5	23.0	"
D4	15.0	P.E./soya bean	G7	33.0	Glycerol/linseed/tung				C2	16.0	Glycerol/D.C.O.
E5	24.5	Glycerol/linseed/tung containing phenolic				F1	22.0	Glycerol/linseed/D.C.O. with 10% styrene	C3	18.0	Glycerol/D.C.O./linseed
			D2	25.0	P.E./tobacco seed						
G1	15.0	P.E./linseed oil				F3	25.0	Glycerol/linseed/tung with 24% styrene			
G2	16.5	Glycerol/linseed				F4	21.5	Glycerol/linseed/tung with 36% styrene			
G3	18.0	"									
G4	22.0	"									
G5	22.0	"									
G6	30.0	"									
Points Rating											
90-120			120-140			140-190			190-370		

in Group 1 are those possessing "good" durability with no appreciable evidence of splitting and checking failures. Group 2 materials possess "fair" durability, showing inferior appearance owing to a greater loss of gloss, with probably some signs of splitting. Materials in Group 3 possess "poor" durability owing to increased tendency for splitting and deep checking failures, whilst Group 4 materials show "bad" durability and in many cases these specimens were withdrawn from the test before the remainder had reached a suitable end point.

Mechanical Properties

The assessment of the mechanical properties of the film was carried out on detached films taken from amalgamated tinned steel specimens prepared by the application of two coats of the varnish, after ageing for approximately six months in the Laboratory. The size of the detached test specimen was 1 cm. in width and approximately 1-1½ in. in length. The film thickness was measured and recorded, the precise length of the specimen between the jaws being measured to the nearest 1/1000 in. The specimens were clamped between the jaws of the apparatus shown diagrammatically in Fig. 2; the upper jaw was capable of movement perpendicularly with the minimum of friction, whilst the lower jaw remained fixed. The load was applied to the film by adding a suitable weight to the pan; the extension produced in the film at pre-determined intervals was assessed by measurement of the distance between the jaws, using a vernier caliper capable of reading to within 1/1000 in. After fifteen minutes the added load was removed from the weight pan and the film was allowed to remain in a "slack" condition at room temperature and humidity. The recovery was measured at intervals up to fifteen minutes after the removal of the load by use of the vernier calipers. Care was taken to ensure that no tension was applied to the film during this measurement.

The apparatus used in this investigation was extremely simple and considerable improvement could clearly be introduced. Even in this form, however, the reproducibility of the results on a number of films was found to be quite satisfactory and within 5 per cent, providing that the applied load was greater

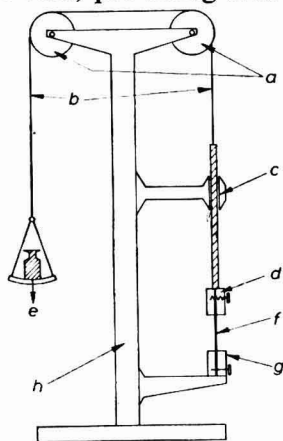


FIG. 2. DIAGRAMMATIC SKETCH SHOWING METHOD OF DETERMINING MECHANICAL PROPERTIES OF DETACHED FILMS

- (a) Pulleys. (b) String. (c) Guide. (d) Upper jaw (movable). (e) Weight.
 (f) Test film. (g) Lower jaw (fixed) (h) Support.

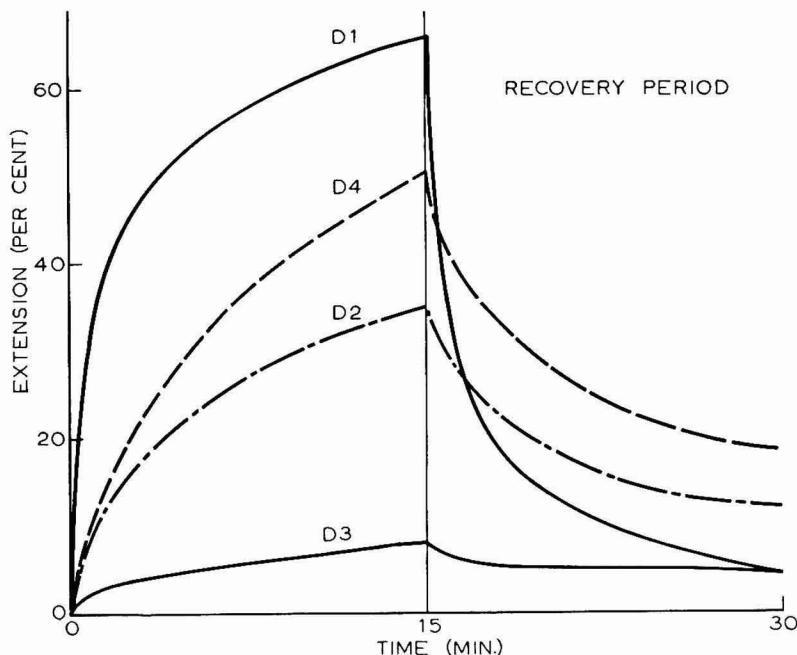


FIG. 3. TYPICAL EXTENSION/TIME CURVES FOR ALKYD VARNISHES [D1, D2, D4—100G. LOAD, D3—300G. LOAD]

than 50 g. and that the extension produced exceeded 25 per cent of the original film length. From the results obtained the extension/time curves were drawn for given loads, of which those illustrated in graphical form are typical (Fig. 3). After a period of fifteen minutes, the extension of the film under load was found (E_{15}) and the modulus of the film was calculated and expressed in dyne/cm.² The final extension of the film was obtained by allowing the extended film to "recover" (after removing the load) for a period of fifteen minutes. The final length of the film (E_F) is then obtained by measurement and the recovery expressed as follows:

$$\% \text{ Recovery} = \frac{E_{15} - E_F}{E_{15}} \times 100$$

MATERIALS TESTED

In order to trace possible relationships between composition and the properties being measured, a wide range of typical alkyd resins and media was selected for the tests, the range of phthalic anhydride contents being adjusted to cover that in use on railway rolling stock. The resins were grouped in the manner indicated below; the number of samples selected in each series is shown in brackets.

These materials were adjusted to a working viscosity by the addition of white spirit, following the addition of the driers (0.5 per cent lead and 0.05 per cent cobalt, expressed on the resin solids). In all cases the film examined consisted of two coats of the prepared varnish applied to the appropriate surface. An indication of the degree of polymerisation was obtained by the measurement of the viscosity of the solution at the same concentration.

Series	Description of resin
A	Oil modified alkyd resins containing 20-23 per cent phthalic anhydride, using glycerol and linseed oil (5)
B	Oil modified alkyd resins as for Section A using pentaerythritol as the polyhydric alcohol (5)
C	Oil modified alkyds with varying phthalic anhydride content, using various types of oils with glycerol as the polyhydric alcohol (5)
D	Similar to Series C, but using pentaerythritol as the polyhydric alcohol (4)
E	Alkyd resins with other modifications involving the use of rosin derivatives and phenolic resins of different types (8)
F	Styrenated alkyd resin media
G	A range of oil modified alkyd resins with varying phthalic anhydride content, mainly based on glycerol and linseed oil (7)

RESULTS

Water Absorption

In contrast to the earlier encouraging results, it was found that there was little or no evidence of correlation between the percentage water absorption and the adhesion of either the detached or attached films (Fig. 4).

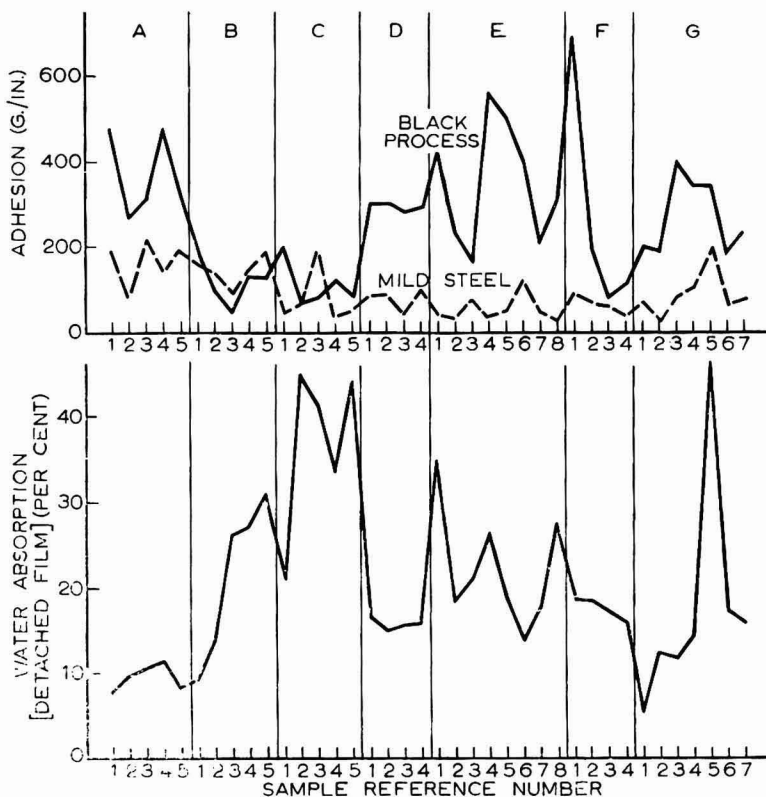


FIG. 4. COMPARISON OF FILM ADHESION AND PERCENTAGE WATER ABSORPTION (AFTER EIGHT DAYS' WATER IMMERSION) (A, B, C, etc., indicate series of test.)

TABLE IV
ADHESION TEST RESULTS SHOWING RECOVERY OF FILM ADHESION
OF FILMS IMMERSSED IN WATER 4 DAYS

Material	Description	Film adhesion (4 days' immersion) (g./in.)	Film adhesion (g./in.) measured after varying drying periods (hr.)									
			1	2	3	4	5	6	7	24	72	
D1	21% P.A. content—pentaerythritol tobacco seed oil alkyd	340	540	660	490	—	900	No failures			—	
E5	24.5% P.A. content—phenolic modified glycerol/linseed/tung oil alkyd	290	380	440	560	—	615	655	820	950	No failure	
F1	22.0% P.A. content—alkyd containing 10% styrene	120	340	520	—	520	570	570	—	No failure		
G2	16.5% P.A. content—glycerol/linseed oil alkyd	320	360	380	390	—	410	425	445	1050	No failure	
G4	22.0% P.A. content—glycerol/linseed oil alkyd	380	420	450	—	530	580	625	—	No failure		
G7	33.0% P.A. content—glycerol/linseed/tung oil alkyd	435	520	560	—	600	650	690	—	980	No failure	

TABLE V
INFLUENCE OF REPEATED IMMERSION AND DRYING OUT OF FILM ADHESION
UNDER WET CONDITIONS

Material	Number of repeated immersions	"Wet" film adhesion (g./in.) after immersion for stated periods (days)		
		1	2	8
D1	0	450	550	300
	24	450	475	300
E5	0	700	650	500
	24	500	410	325
F1	0	575	700*	700*
	24	475	400	200
G2	0	425	350	200
	24	400	400	300
G4	0	525	450	350
	24	525	400	200
G7	0	1,000	750	250
	24	1,200	600	350

* Indicates failure from enamel coat while remainder fail from undercoat.

Adhesion

The general conclusions to be drawn from the tests on the various alkyd materials can be summarised as follows. The film adhesion of alkyd processes is considerably reduced when subject to moisture. The loss of adhesion persists while the film is wet, but on drying the original film adhesion may be approached; this fact is shown very clearly in Table IV. In Table V is shown the effect of twenty four cycles of repeated immersion and drying (occupying alternate days) on adhesion, measured after the final cycles, for periods of immersion up to eight days test, using as a control the results obtained on duplicate panels not subjected to the above treatment.

A comparison of the adhesion of the varnishes when applied to the substrates employed, namely, tinned steel, mild steel and black enamel, shows only poor correlation. The value of adhesion depends to a great extent on the nature of the substrate, and materials giving good adhesion to painted surfaces sometimes give inferior adhesion on metal surfaces.

With regard to the effect of composition on adhesion in paint systems, the following indications can be seen.

- (i) With alkyds of similar phthalic anhydride content and based on linseed oil, the substitution of pentaerythritol in place of glycerol causes reduced adhesion (Series A and B, Table VI).

TABLE VI

ADHESION TEST RESULTS OBTAINED ON VARNISH SAMPLES CONTAINING 20-23% P.A. WITH LINSEED OIL (AFTER 8 DAYS' WATER IMMERSION)

Material	Fatty acids (%)	Minimum film adhesion (g./in.) after 8 days' immersion			
		Tinned iron	Mild steel	Black paint process	
				Adhesion	Position of failure
<i>A series (glycerol alkyds)</i>					
1	60.4	13	187	480	} Mixed, enamel and u/c Undercoat
2	66.8	20	80	270	
3	70.0	27	210	310	
4	65.0	54	145	480	
5	65.0	85	185	310	
<i>B series (pentaerythritol alkyds)</i>					
1	67.5	36	159	180	} Primer
2	68.0	33	142	100	
3	62.3	51	95	50	
4	62.3	36	148	130	
5	67.0	40	184	130	

TABLE VII

ADHESION TEST RESULTS OBTAINED ON VARNISH SAMPLES OF VARYING COMPOSITION WITH DIFFERING OILS (AFTER 8 DAYS' IMMERSION)

Material	Fatty acids (%)	P.A. (%)	Type of oil	Minimum film adhesion (g./in.) after 8 days' immersion			
				Tinned iron	Mild steel	Black paint process	
						Adhesion	Position of failure
<i>C series (glycerol alkyds)</i>							
1	58	27	Linseed/tung	90	43	200	Enamel
2	77	16	D.C.O.	170	67	65	
3	67	18	D.C.O./linseed	150	186	80	Metal
4	55	31	Linseed/tung	95	40	115	
5	48.6	36	Soya bean	52	53	95	
<i>D series (pentaerythritol alkyds)</i>							
1	66	21	Tobacco seed	31	86	300	Enamel
2	63	25	Tobacco seed	19	86	300	Undercoat
3	57	28	Linseed/tung	23	39	280	Mixed failure
4	55	15	Soya bean	38	92	290	Undercoat
<i>Series A</i>				40	160	370	—
<i>Series B</i>				40	145	120	—

- (ii) With alkyd media based on other types of drying and semi-drying oils, such as tung oil, dehydrated castor oil and soya bean oil, with glycerol (Series C, Table VII), the adhesion is somewhat inferior to those based on linseed oil alone (Series A). However, in the case of resins containing pentaerythritol (Series D), the use of tobacco seed or soya bean oil gives practically comparable results to those of glycerol/linseed alkyds.
- (iii) The incorporation of other resins in the oil modified alkyds, such as rosin and phenolic derivatives, gives a tendency for the adhesion failure to occur higher in the paint system, e.g. in Series E the failure occurs mainly at the enamel/varnish interface (Table VIII).
- (iv) The styrenated alkyds (Series F) tend to give failures lower in the system, except in the case of the material containing the least styrene.
- (v) The comparison of the results with Series G (Table IX), in which the phthalic anhydride was varied, gives no evidence (for the group of glycerol/linseed alkyds) that a direct relationship between adhesion and phthalic anhydride content exists. This result was somewhat unexpected in view of experience and, because it seemed probable that other

TABLE VIII
ADHESION TEST RESULTS ON VARNISH SAMPLES OF VARYING COMPOSITIONS
AFTER 8 DAYS' IMMERSION

Material	Fatty acids (%)	P.A. (%)	Description	Minimum film adhesion (g./in.) after 8 days' immersion			
				Tinned iron	Mild steel	Black paint process	
						Adhesion	Position of failure
<i>E series (other modifications)</i>							
1	49.5	34	Glycerol/linseed + rosin	36	38	420	Enamel
2	68.0	19	P.E./linseed/ D.C.O. + rosin	21	33	230	Enamel
3	55	19	Glycerol/linseed /tung with rosin phenolic	40	76	160	Primer
4	72	17.5	Glycerol/linseed /tung phenolic	23	37	560	Enamel
5	64.4	24.5	Glycerol/linseed /tung phenolic	17	44	510	Enamel
6	75.3	19.0	Glycerol/linseed /tung phenolic	200	120	400	Enamel
7	63	20.0	P.E./linseed/ tung phenolic	25	50	210	Primer
8	65	25.0	Glycerol/linseed /tung with 100% phenolic	55	30	310	Enamel
<i>F series (styrenated alkyds)</i>							
1	57	22	Glycerol/linseed /D.C.O./10% styrene	25	97	700	Mixed
2	46	27	Glycerol/linseed /tung/16% styrene	12	68	195	Primer
3	42	25	Glycerol/linseed /tung/24% styrene	7	60	75	Primer
4	37	21.5	Glycerol/linseed /tung/36% styrene	17	42	135	Metal
<i>Series A (glycerol/linseed) mean value</i>				40	160	370	—

differences in these materials had masked the effect sought, it was decided to undertake a further series of tests in which not only the actual phthalic anhydride content would be known but also the method of manufacture; the reference to the additional tests is made in Part Two of this paper.

TABLE IX
ADHESION TEST RESULTS ON VARNISH SAMPLES SHOWING MAINLY INFLUENCE OF P.A.
CONTENT AFTER 8 DAYS' IMMERSION

Material	Fatty acids (%)	P.A. (%)	Minimum film adhesion (g./in.) after 8 days' immersion			
			Tinned iron	Mild steel	Black paint process	
					Adhesion	Position of failure
G Series (mainly glycerol/linseed alkyds)						
1*	70	15	18	62	200	Primer
2	50	16.5	12	25	190	Primer
3	65	18	28	90	400	Enamel
4	65	22	28	110	350	Enamel
5	48	25	198	200	350	Enamel
6	56.6	30	9	70	190	Primer
7†	60.0	33	21	80	240	Primer

*This material based on pentaerythritol only.

†This material contains linseed/tung oil.

Durability

The results (Tables X-XIII) obtained show strongly a connection between composition and durability, and may be summarised in the following manner.

- (i) Considerable loss of durability arises from the modification of the alkyd by use of rosin or phenolic materials. Only two of the selected materials in this class, E5 and E8, both containing approximately 25 per cent phthalic anhydride in glycerol and linseed/tung oil, show any promise. All materials containing rosin or rosin/phenolic resins gave bad durability and were withdrawn at an early stage from the test.
- (ii) In the case of materials in Series A and B, both containing 20-23 per cent phthalic anhydride and linseed oil, the replacement of glycerine by pentaerythritol causes a reduction in durability. In fact, the only satisfactory materials using pentaerythritol as the polyhydric alcohol were those using a lower phthalic anhydride content (15 per cent) with a high fatty acid content (*e.g.* material G1), or employing semi-drying oils, such as soya bean and tobacco seed oils (materials D4 and D1).
- (iii) Variations in the nature of the oil used indicate that superior results are obtained on alkyd resins using glycerine in conjunction with linseed oil or linseed/tung oil, the resins containing dehydrated castor oil being definitely inferior in durability. It must be stressed that in all cases the materials were used on the same basic paint process as finishing varnishes. However, if semi-drying oils, such as soya bean oil, tobacco

TABLE X
DURABILITY RATINGS AND FILM PROPERTIES ON VARNISH SAMPLES CONTAINING
20-23% P.A. WITH LINSEED OIL

Material	Points for failures during exposure					Data from extension/time curves				
	½ stage	⅓ stage	¼ stage	Final	Σ Points for failure	Applied load (g.)	Film thickness (microns) (μ)	Extension (15 min.) (%)	Recovery (%)	Force to provide 100% extension (dyne/cm. ²) × 10 ⁻⁶
A Series (glycerol alkyds)										
1	6	20	48	68	142	100	50	20	53	98
2	6	12	36	52	126*	100	45	40	70	55
3	4	12	36	52	104	50	39	62	97	20
4	10	20	40	52	122	100	37	98	91	27
5	12	12	40	52	116	50	41	64	92	19
B Series (pentaerythritol alkyds)										
1	8	20	52	84	164	100	26	55	85	69
2	8	20	48	76	152	100	30	50	80	65
3	12	28	48	68	156	50	22	46	87	49
4	12	20	60	84	176	50	22	44	85	51
5	16	32	60	80	188	100	35	31	63	90

*Includes +20 points for rusting failure.

seed oil, *etc.*, are used, pentaerythritol resins give superior results, provided that the phthalic anhydride content is not excessive.

- (iv) The use of styrenated alkyd resin varnishes does not appear to yield a greatly improved durability, and from the limited results it would appear that the styrene content should not exceed 16 per cent (*cf.* material F2 (16 per cent styrene) against F3 (24 per cent) and F4 (36 per cent), all based on glycerol/linseed/tung oil type resins). The lower durability of material F1, even though of a lower styrene content, is attributed to the use of dehydrated castor oil in the resin.
- (v) The nature of the influence of the phthalic anhydride content seems to depend on the other constituents of the resin, but for glycerol/linseed or linseed/tung oil type resins, the durability seems to be unaffected by variations of the phthalic anhydride content between the limits of 16.5-33 per cent (Series G).

Mechanical Properties

The results obtained are given in Tables X-XIII. A number of points of general interest arise, *e.g.* the extent to which the mechanical properties are influenced by the chemical constituents of the materials, and also how the film properties can be affected by the method of manufacture (Fig. 5).

In Table X (Glycerol Alkyds) the material A1 possesses an inferior film recovery (53 per cent) and an appreciably higher modulus compared with similar materials A3, A4 and A5 (recoveries 91-97 per cent), whereas the

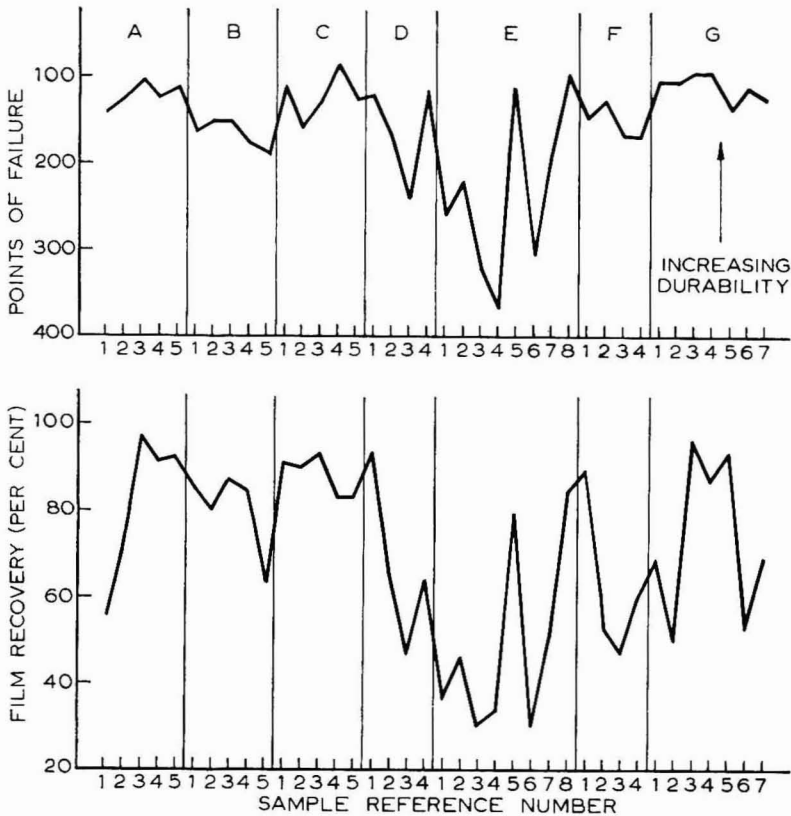


FIG. 5. COMPARISON OF FILM DURABILITY AND FILM RECOVERY (A, B, C, etc., indicate series of test.)

materials in Table X, based on pentaerythritol, show a higher modulus figure than the typical numbers of Series A materials and the average recovery figure is approximately 80 per cent. Similarly, in Table XI the figure for the modulus is, in a number of cases, considerably lower for films prepared from the different oils, in spite of higher phthalic anhydride content. In Table XIII the toughness of the material D3 is most striking and is apparently due to the use of linseed/tung oil in its composition, as compared with the semi-drying oils used in the other materials. In Table XII (Rosin and Phenolic Modified Alkyds) the bulk of the samples show high figures for the modulus and possess low recovery characteristics (30-50 per cent), whilst in the Table indications are to be seen that increasing styrene content gives increasing resistance to extension. Finally, in Table XIII it is apparent that the amount of phthalic anhydride present in the glycerol type alkyds has very little influence on the film properties involved in the tests.

In a number of cases, the film reaction with varying applied loads was investigated and it was immediately apparent that the films failed to obey exactly the ideal relationship between applied load and the extension produced by the load, although quite good agreement was obtained for the percentage recovery in all cases, unless the film prematurely broke during the fifteen

TABLE XI
DURABILITY RATINGS AND FILM PROPERTIES OF VARNISH SAMPLES OF VARYING COMPOSITION WITH DIFFERING OILS

Material	P.A. content (%)	Type of oil	Points for failure during exposure					Data from extension/time curves						
			‡ Stage	‡ Stage	‡ Stage	Final	Σ Points for failure	Applied load (g.)	Film thickness (μ)	Ex-tension (15 min.) (%)	Recovery (%)	Force to produce 100% extension (dyne/cm. ²) × 10 ⁻⁶		
C Series (Glycerol alkyls)														
1	27	Linseed/tung	4	16	44	48	112	100	60	66	91	25		
2	16	D.C.O.	8	20	36	56	160**	10	26	34	90	1.5		
3	18	D.C.O./linseed	6	12	36	56	130*	20	26	34	93	1.5		
4	31	Linseed/tung	10	12	32	36	90	50	41	42	83	19		
5	36	Soya bean	10	10	36	52	128*	50	28	28	83	2.0		
D Series (Pentaerythritol alkyls)														
1	21	Tobacco seed	6	12	48	56	122	100	39	66	93	38		
2	25	Tobacco seed	12	20	56	80	168	100	37	36	66	74		
3	28	Linseed/tung	20	48	72	100†	240	500	39	29	47	430		
4	15	Soya bean	12	20	36	48	116	100	37	51	63	52		

*Includes +20 parts for "rusting".

**Includes +40 parts for "rusting" and "adhesion" failures.

†Specimen D3 withdrawn from exposure test before majority of specimens.

TABLE XII
DURABILITY RATINGS AND FILM PROPERTIES OF VARNISH SAMPLES WITH VARYING MODIFICATIONS

Material	P.A. content (%)	Description	Points for failure during exposure				Data from extension/time curves					
			‡ Stage	§ Stage	¶ Stage	Final	Σ Points for failure	Applied load (g.)	Film thickness (μ)	Ex-tension (15 min.) (%)	Recovery (%)	Force to produce 100% extension (dyne/cm. ²) × 10 ⁻⁶
E Series (Alkyds with other modifications)												
1	34	Glycerol/linseed + rosin	16	44	100*	100*	260	200	37	7.5	37	710
2	19	P.E./linseed/D.C.O. + rosin	11	32	80	100*	223	200	56	26.5	46	130
3	19	Glycerol/linseed/tung with rosin phenolic	36	84	100*	100*	320*	200	41	20.0	30	240
4	17.5	Glycerol/linseed/tung and phenolic	84	84	100*	100*	368	200	37	7.0	34	760
5	24.5	" " " "	6	10	32	64	112	100	43	80	79	28
6	19.0	" " " "	32	72	100*	100*	304	200	52	7.9	30	480
7	20.0	P.E./linseed/tung/phenolic	8	16	68	100*	192	200	52	24.0	51	160
8	25.0	Glycerol/linseed/tung with 100% phenolic	6	10	36	48	100	100	52	61.0	84	31
F Series (Styrenated alkyds)												
1	22	Glycerol/linseed/D.C.O./10% styrene	12	20	48	68	148	100	56	97	89	18
2	27	Glycerol/linseed/tung/16% styrene	8	24	44	52	128	100	52	42	53	45
3	25	" " " 24% "	12	32	48	76	168	200	47	63	47	66
4	21.5	" " " 36% "	10	12	56	80	168	200	56	16	60	220

*Specimens withdrawn from exposure test before majority of specimens.

TABLE XIII
DURABILITY RATINGS AND FILM PROPERTIES OF VARNISH SAMPLES, SHOWING MAINLY INFLUENCE OF VARIATIONS ON P.A. CONTENT

Material	P.A. content (%)	Points for failure during exposure				Σ Points for failure	Date from extension/time curves				Force to produce 100% extension (dyne/cm. ² $\times 10^{-4}$)
		$\frac{1}{2}$ Stage	$\frac{1}{3}$ Stage	$\frac{2}{3}$ Stage	Final		Applied load (g.)	Film thickness (μ)	Extension (15 min.) (%)	Recovery (%)	
G Series (mainly glycerol/linseed alkyds)											
1*	15.0	6	12	36	52	106	75	40	69	65	
2	16.5	6	12	44	44	106	41	54	50	89	
3	18.0	6	10	32	48	96	75	72	96	18	
4	22.0	6	12	32	48	96	56	90.5	87	19	
5	25.0	10	24	48	56	138	49	108	93	9	
6	30.0	10	20	40	44	114	37	86	53	60	
7**	33.0	10	20	40	56	126	60	50	69	64	

*Material based on pentaerythritol only.

**Material contains linseed/tung oil.

minutes' loading period. A further point observed during a number of supplementary tests carried out on detached films was the effect produced by the use of comparatively low loads applied for fifteen minutes followed by the recovery period of fifteen minutes, and repeated until finally the film broke. The break occurred at extensions as were obtained by single but greater loads, and in fact by choice of loads, the break could be produced at the film's maximum extension, practically instantaneously, or after varying periods of time.

Although no evidence was obtained to indicate any close correlation between "adhesion" and the mechanical properties measured, these properties assume great special interest as it appears from Fig. 5 that there is considerable correlation or relationship between the ability of the film to recover its original state after removal of the load causing extension and the film durability. The durability also seems related to the force required to produce 100 per cent extension (film modulus) which should not be excessively large ($30\text{--}40 \times 10^6$ dyne/cm.²), as in the case of the materials in Series E, except E5 and E8 which seems to be of the order necessary for "good" durability. However, the films of very low modulus seem to possess lower durability as illustrated by materials C2 and C3.

SUMMARY OF RESULTS—PART ONE

The method described for quantitative assessment of adhesion has enabled some interesting data to be collected, and has revealed the extreme sensitivity of alkyd finishes to water, whether they are exposed to high humidity (in excess of 90 per cent) or completely immersed. The loss of adhesion persists while the film is wet, but is apparently entirely restored when the film has again become dry; this was demonstrated by measuring the adhesion of an alkyd resin system after it had been subject to twenty-four cycles of wetting and drying. The substrate has a considerable influence on the measured adhesion, and the relative adhesion of different materials varies with the substrate. As may be expected, the chemical composition of the alkyd, the modifying oil, the polyhydric alcohol, and any modification, affects adhesion but generally in an unpredictable way.

Perhaps the most interesting outcome of this work may prove to be the apparent relationship traced between the modulus of the film, its extensibility and power of recovery and its durability. The apparatus used in these tests is open to criticism, but the existence of some measure of correlation between the properties is most strongly indicated. Provisionally it would appear that for good durability a modulus not greater than 30×10^6 , and a percentage recovery, after extension to 100 per cent, of not less than 90 per cent, is necessary for oil modified alkyds. If further work confirms this relationship, a valuable tool for the assessment of paint durability will have been forged. No correlation could be found between the mechanical properties referred to and adhesion.

PART TWO

The results obtained in Part One of this paper were considered to be of interest and of some value, but also to leave a number of questions unanswered. It was decided, therefore, to carry out a further series of tests in which materials of a more limited range of compositions would be compared, but about which

TABLE XIV
COMPOSITION AND PROPERTIES OF GLYCEROL/ALKYD VARNISH MEDIA AS SUPPLIED

Material	Ingredients used (%)			Properties of medium		
	Oil/fatty acid	Glycerol	P.A.	Solids (%)	Viscosity (Stokes)	Acid value
G1	75	8.5	16.5	58	4	9.5
G2	70	10.5	19.5	58	4.6	7.4
G3	65	12.5	22.5	55	4.6	8.2
G4	60	14.5	25.5	55	7.5	5.0
G5	52.6*	20.8	30.0	50	7.0	10.0

TABLE XV
COMPOSITION AND PROPERTIES OF PENTAERYTHRITOL ALKYD VARNISH MEDIA AS SUPPLIED

Material	Ingredients used (%)			Properties of medium		
	Fatty acid	Penta-erythritol	P.A.	Solid (%)	Viscosity (Stokes)	Acid value
P1	68.5*	16.5	15.0	6.0	3.5	2.0
P2	64.0*	18.0	18.0	60	5.5	3.5
P3	60.0*	19.5	20.5	55	6.0	4.5
P4	55.0*	21.0	24.0	53	7.0	7.0
P5	51.0*	22.5	26.5	50	7.0	7.0

*Signifies medium prepared by fatty acid process whilst remainder were prepared by monoglyceride process.

TABLE XVI
COMPOSITION AND PROPERTIES OF MIXED GLYCEROL/PENTAERYTHRITOL ALKYD MEDIA

Material	Ingredients used (%)			Properties of medium		
	Oil	Penta-erythritol	P.A.	Solid (%)	Viscosity (Stokes)	Acid Value
PG1**	75.1	9.1	15.8	62	5	6.0
PG2	67.0	12.0	21.0	55	5	6.0

**Made by lengthening the PG2 material after esterification with a mixed linseed/tung stand oil.

much more detailed information concerning their composition and method of manufacture was available. The samples in this series were of a predetermined phthalic anhydride content; with either glycerol or pentaerythritol as the polyhydric alcohol, the details of manufacture were also known.

The various materials tested possessed the compositions and properties given in Tables XIV-XVI.

The resin solutions were used as varnishes after the addition of naphthenate driers to give 0.5 per cent lead and 0.05 per cent cobalt, calculated on the resin solids, as in the original tests.

Two other varnishes were included in the tests, namely, an alkyd resin varnish to B.R. Specification No. 1, Item 7 (containing 26.8 per cent phthalic anhydride), and a copal ester varnish to B.R. Specification No. 6, Item 4. The same properties were studied using the same methods as in Part One.

- (a) Water Absorption and Evaporation Rate.
- (b) Adhesion,
- (c) Durability, and
- (d) Mechanical Properties.

RESULTS

Water Absorption

The results (Table XVII) confirm that the percentage of water absorbed by the film is independent of the phthalic anhydride content for both the glycerol and pentaerythritol resins, although the latter as a class showed rather lower absorptions than the glycerol resins which, in their turn, were considerably lower in water absorption than the oleoresinous varnish.

Significantly, however, there is a definite relationship between the evaporation rate and the phthalic anhydride content. Increasing phthalic anhydride content is accompanied by increasing evaporation rate, with the pentaerythritol resins giving higher rates than the glycerol resins. This could be interpreted as indicating either that the water within the film is more loosely held or, alternatively, that the films become more permeable with increasing phthalic anhydride content; the latter assumption is clearly untenable.

The oleoresinous varnishes have characteristics in strong contrast with the alkyds, possessing the lowest evaporation rates but the highest water absorption, as is evident from the table below.

Material	Water absorption	Evaporation rate (K)
Glycerol alkyd varnishes (average)	15.5	0.16
Pentaerythritol alkyd varnishes (average)	11.7	0.28
Oleoresinous varnishes	55.0	0.07

Water absorption tests were also carried out on a Standard painting system. Owing to the difficulty of obtaining strictly comparable films with the multi-coat system, it was realised that the actual percentage water absorption figure would have only limited significance, but it was felt that the evaporation constants of the processes might well be of interest.

TABLE XVII

WATER ABSORPTION DATA FOR DETACHED FILMS OF VARNISH AFTER 8 DAYS' IMMERSION
(Age of film—3 months)

Material	Type of varnish	Water absorption test data		
		Loss in weight of material from dry film after 8 days' immersion (%)	Evaporation constant (K)	Water absorption (corrected) (%)
G1	Glycerol alkyds (see Table XIV)	3.8	0.16	15.9
G2		2.8	0.14	17.6
G3		3.7	0.13	14.5
G4		1.9	0.18	13.3
G5		2.5	0.21	16.3
P1	Pentaerythritol alkyds (see Table XV)	4.1	0.20	11.8
P2		3.8	0.25	11.2
P3		2.8	0.31	10.6
P4		2.6	0.32	11.8
P5		2.7	0.36	11.7
PG.1	Mixed alkyds (see Table XVI)	3.3	0.32	13.8
PG.2		3.6	0.30	12.3
Std. I	B.R. Spec. 1, Item 7	1.8	0.26	14.6
Std. II	B.R. Spec. 6, Item 4	4.8	0.07	55.0

The results (Table XVIII) indicate that the water absorption of the processes finished with the pentaerythritol type alkyds is somewhat less than that of the other materials, but the differences are less marked than for the detached films of the varnish. It is, however, interesting to note that the relative values for the evaporation constant (K) obtained for the processes show that the constants for the pentaerythritol alkyds are considerably lower than those of the other materials; an effect which is, in fact, opposite to that observed on the detached films. This lower evaporation constant is a result of the different characteristics of the pigmented undercoats and the varnish. Although the low permeability of the pentaerythritol varnish resulted in a lower water absorption, it may be assumed that the bulk of the moisture so absorbed was held rather tenaciously by the undercoatings, thus accounting for the low evaporation rate.

TABLE XVIII

WATER ABSORPTION DATA ON MULTI-COAT SYSTEM (2 COATS VARNISH)
AFTER 8 DAYS' IMMERSION

Material	Type of varnish	Water absorption test data (full process)		
		Loss in weight of dry film after 8 days' immersion (%)	Evaporation constant (K)	Water absorption (corrected) (%)
G1	Glycerol alkyds (see Table XIV)	2.1	0.022	12.4
G2		1.6	0.021	10.8
G3		1.9	0.021	12.4
G4		1.6	0.016	11.0
G5		1.6	0.018	12.9
P1	Pentaerythritol alkyds (see Table XV)	1.4	0.012	11.5
P2		1.2	0.008	9.5
P3		0.95	0.004	9.9
P4		0.95	0.005	9.4
P5		1.0	0.009	11.2
PG.1	Mixed alkyds (see Table XVI)	1.1	0.012	10.8
PG.2		1.3	0.011	13.3
Std. I	B.R. Spec. 1, Item 7	1.3	0.008	10.7
Std. II	B.R. Spec. 6, Item 4	1.8	0.027	13.1

Adhesion Measurements

In tests involving the Standard paint process failures were found to occur at various levels within the process and the point of failure is indicated in Tables XIX-XXI. The minimum intercoat adhesion of the system is also given in these Tables. After preparation the specimens were aged for a period of three months under laboratory conditions before being subjected to the various adhesion tests. In addition, after six months' ageing and twenty-four hours' extra exposure to ultra-violet light, some of these same panels were overcoated with a further two coats of the same varnish in order to assess the adhesion of each material when used for revarnishing weathered processes. The adhesion of each system was measured both before and after periods of immersion in water. In the latter case, tests were carried out immediately after the specimens had been withdrawn from water and again after complete drying out.

TABLE XIX
ADHESION OF VARNISHES ON BLACK PROCESS IN "DRY" CONDITION

Material	Type of varnish	Minimum adhesion of black process (original state) (g./in.) (showing also position of failure)	Minimum adhesion of revarnished specimen	Adhesion (g./in.) of original film after 8 days' water immersion followed by "drying out"
G1	Glycerol alkyds (see Table XIV)	NF—750	Slight V/V—900	NF—800
G2		NF—750	V/V—650	NF—700
G3		NF—750	V/V—500	NF—650
G4		NF—750	V/V—250	NF—850
G5		NF—650	Slight V/V—1,000	NF—600
P1	Pentaerythritol (see Table XV)	NF—550	NF—950	NF—725
P2		Slight failure —900	Partial failure —900	NF—700
P3		V/V—300	V/V—550	V/V—475
P4		V/V—250	V/V—450	Enamel failure or V/V—450
P5		V/V—350	V/V—225	Enamel failure or V/V—500
PG.1	Mixed alkyds (see Table XVI)	Partial varnish failure—525	V/V—900	Partial enamel failure—650
PG.2		V/V—625	V/V—600	Partial enamel failure—600
Std. I	B.R. Spec. 7, Item 1	NF—625	V/V—675	NF—625
Std. II		B.R. Spec. 6, Item 4	NF—725	NF—1,100

V/V—Varnish/Varnish failure. NF—No failure.

The results quoted in Table XIX are based on the mean adhesion figures obtained on duplicate panels, the tests being carried out in the dry state. The results apply to the panels after ageing in the Laboratory for three months, and the following points emerge. Varnish adhesion failures occurred in six out of seven specimens finished with the alkyd varnishes containing pentaerythritol.

The minimum inter-coat adhesion of a duplicate series of specimens, subsequent to a period of eight days' water immersion followed by a thorough drying period, was comparable to the initial figures, although it will be seen from Table XX that the adhesion whilst still wet was considerably lower.

The tests carried out on the revarnished specimens show the superiority in adhesion of the oleoresinous varnish (Specification 6, Item 4). The figures for the pentaerythritol alkyd materials show that in revarnishing, the applied film possessed an adhesion equivalent to that of the exposed varnish, as did

TABLE XX

ADHESION OF BLACK PROCESS UNDER "WET" CONDITIONS RESULTING FROM WATER IMMERSION FOR VARYING PERIODS

Material	Panel	Minimum adhesion of film after varying periods of water immersion-black process, showing position of failure								Minimum adhesion of revarnished process	
		1 day		2 days		4 days		8 days		After 1 day	
		Adhesion (g./in.)	Position	Adhesion	Position	Adhesion	Position	Adhesion	Position	Position of failure	Adhesion (g./in.)
G1	A	450	Mixed	150	Item 1	125	Item 1	50	Item 1	} V/V	10
	B	170	Item 1	100	Item 1	100	Item 1	50	Item 1		
G2	A	180	Item 1	30	Item 1	20	Item 1	15	Item 1		4
	B	120	Item 1	25	Item 1	30	Item 1	25	Item 1		
G3	A	75	Item 1	25	Item 1	20	Item 1	20	Item 1		3
	B	700	Item 6	420	Mixed	300	Item 1 & Item 7	60	Item 1		
G4	A	550	Item 6/7	400	Item 1	275	Mixed	180	Item 1		5
	B	450	Item 6/7	400	Item 6	300	Mixed	170	Item 1		
G5	A	175	Item 1	150	Mixed	150	Item 1	200	Item 1		7
	B	475	Item 6/7	300	Item 6/7	300	Item 6/7	200	Item 1		
P1	A	400	Item 7	200	Item 7	200	Item 7	180	Item 7		7
	B	400	Item 7	200	Item 7	200	Item 1	200	Item 1		
P2	A	300	Item 7	125	Item 7	120	Item 7	120	Item 7		2
	B	325	Item 7	135	Item 7	120	Mainly Item 7	140	Item 1/7		
P3	A	200	Item 7	120	Item 7	80	Item 7	55	Item 7	2	
	B	125	Item 7	100	Item 7	70	Item 7	65	Mixed Item 1/7		
P4	A	90	Item 7	100	Item 7	100	Item 7	60	Item 7	1	
	B	120	Item 7	100	Item 7	100	Item 7	70	Item 7		
P5	A	80	Item 7	70	Item 7	80	Item 7	60	Item 7	5	
	B	100	Item 7	90	Item 7	70	Item 7	75	Item 7		
PG.1	A	260	Item 7	190	Item 7	180	Item 7	90	Item 7	2	
	B	220	Item 7	180	Item 7	150	Item 7	90	Item 7		
PG.2	A	250	Item 7	150	Item 7	200	Item 7	50	Item 7	3	
	B	180	Item 7	165	Item 7	200	Mixed Item 1/7	100	Item 1		
Std. I	A	400	Item 6	350	Mixed	50	Item 1	45	Item 1	3	
	B	420	Item 7	330	Mixed	100	Item 1	50	Item 1		
Std. II	A	575	Item 6	310	V/V initially	200	Mixed Item 7/1	170	Item 7/1	18	
	B	560	Item 6/7	350	„	240	Item 1	160	Item 1		

Key of paint process

- Item 1. Primer for metal.
- Item 6. Black sealer undercoat.
- Item 7. Black enamel varnish.

the same varnish applied to the paint. On the other hand, the glycerol alkyds (G1-G5 inclusive) show that in revarnishing the film fails to adhere to the weathered varnish as effectively as it had done in the original. The two types of varnish therefore show dry adhesion of the same order when applied over weathered varnish. Some evidence is available that the increasing phthalic anhydride content has detrimental results on the varnish adhesion, whether the alkyd is based on pentaerythritol or glycerol. The only exception is material G5 (in the glycerol alkyd range), which was prepared by the "fatty acid process" as opposed to the "monoglyceride process" used for materials G1-G4 inclusive.

A further series of experiments was carried out on the same specimens after varying periods of water immersion. The tests were carried out in duplicate and some differences were obtained, particularly in the initial stages of the immersion. In the case of the revarnished specimens, it was found that the failures occurred extremely rapidly at low loads, which made it unnecessary to proceed beyond the one day immersion period. The results obtained are summarised in Table XX.

A number of points emerge from a consideration of Table XX.

- (i) When revarnished panels were tested, the additional coats were readily removed after only one day's immersion.
- (ii) The failure of adhesion of materials based on pentaerythritol takes place from the paint system (after eight days), whilst in the case of the other materials the failure occurs from the metal primer.
- (iii) In the case of the glycerol alkyds, the phthalic anhydride content influences the adhesion, which starts at reasonably high level, falling for materials G2 and G3, followed by increasing figures for G4 and G5. In the case of the pentaerythritol alkyds, the adhesion falls off as the phthalic anhydride content increases.

TABLE SHOWING INFLUENCE OF PHTHALIC ANHYDRIDE CONTENT ON FILM ADHESION

Range of phthalic anhydride content (%)	Adhesion test results on Black process (g./in.)			
	Glycerol alkyds		Pentaerythritol alkyds	
	After 8 days' immersion	" Dry "	After 8 days' immersion	" Dry "
15-19.5	50-20	No failure	190-130	No failure
20.5-24.0	40	"	60-65	300*
25.5-30.0	175-200	"	65	350*

* Failure between varnish/varnish coats.

- (iv) Considering the results as a whole, it would appear that the materials can be grouped into approximately the following three classes after eight days' water immersion.

<i>Superior adhesion</i> (above 100 g./in.)	{ G4, G5 P1, P2 and Std. II (oleoresinous varnish)
<i>Intermediate adhesion</i> (50-100 g./in.)	{ P3, P4 and P5 PG1 and PG2
<i>Inferior adhesion</i> (less than 50 g./in.)	{ G1, G2 and G3 Std. I (alkyd resin type varnish)

TABLE XXI
FILM ADHESION AFTER WEATHERING TEST

Material	Type of Varnish	Adhesion values after 8 days' immersion showing position of failure and load/inch to cause failure after accelerated weathering		Order of merit after exposure	Order of merit before exposure (see Table XX)
		Adhesion (g./in.)	Point of failure		
G1	Glycerol alkyds (Table XIV)	220	Item 6	11	11
G2		190	Item 6	13	14
G3		180	Item 6	14	13
G4		280	Item 6	9	3
G5		300	Item 6	8½	1
P1	Pentaerythritol alkyds (Table XV)	400	No failure	2½	2
P2		400	No failure	2½	5
P3		370	Item 6 and some failure from metal	6	10
P4		500	No failure	2½	9
P5		430	Item 6 and metal mixed	5	8
PG.1	Mixed alkyds (Table XVI)	300	Mixed failure mainly Item 6	8½	6
PG.2		325	Item 6	7	7
Std. I	Spec. 1, Item 7	230	From Item 6 or Item 7	10	12
Std. II	Spec. 6, Item 4	650	No failure	2½	4

From Table XXI it will be seen that the differences are less marked than those obtained earlier, but some similarity of behaviour persists. The results recorded in Table XXI are those obtained on the same specimens as those referred to in Table XX, but after a period of fourteen weeks accelerated weathering.

Evaluation of Durability

For this section of the investigation a series of panels was prepared using the same paint process as described in previous sections. The metal panels (9 in. × 2 in.) used were of mild steel, as used for railway carriages. The panels, after preparation, were aged for a suitable period before exposure in the usual accelerated weathering cycle for fourteen weeks, or until failures due to checking and splitting were recorded. At the conclusion of the tests the specimens were examined and placed in order of merit of their final conditions. The results obtained for this series of specimens are summarised in Table XXII.

TABLE XXII
DURABILITY OF BLACK PROCESS AS ASSESSED BY ACCELERATED WEATHERING
TESTS OF 14 WEEKS' DURATION

Material	Failure points assessed during exposure					Order of merit	
	¼ Stage	½ Stage	¾ Stage	Final	Failure points progressive total	From points rating	Visual assessment at conclusion
G1	16	34	36	40	126	4½	5
G2	16	24	40	40	120	3	6
G3	20	28	40	44	132	7½	4
G4	18	28	40	40	126	4½	3
G5	18	28	32	40	118	2	1
P1	16	28	40	44	128	6	7
P2	20	28	40	48	136	9	8
P3	16	36	60	120	232	11	11
P4	16	44	80	100	240	13	12
P5	12	36	68	120	236	12	13
PG.1	12	28	32	60	132	7½	9
PG.2	20	36	40	52	158	10	10
Std. I	12	20	32	40	104	1	2
Std. II	44	68	84	104	300	14	14

However, if the orders of merit for durability of the above series are considered from a practical angle, it is possible to class the materials in three main groups.

Group I—Materials of satisfactory durability

These are materials G1, G2, G3, G4, G5, P1, P2, PG1 and the Standard I (S.R. Varnish).

Group II—Materials of inferior durability

This group consists of materials PG2, P3, P4 and P5.

Group III—Material of unsatisfactory durability (compared with the above materials of the alkyd type)

Oleoresinous varnish (Std. II), although this varnish is known to be among the most durable of its type.

From the above results, the following general conclusions can be drawn regarding the effect of composition on durability.

- (i) The alkyd varnishes are of superior durability to the oleoresinous varnish Std. II.
- (ii) Only minor differences exist between the durability of the alkyd varnishes based on the glycerol types within the ranges tested, irrespective of the phthalic anhydride content.
- (iii) In the case of alkyd media containing pentaerythritol, satisfactory materials can be obtained, provided that neither the phthalic anhydride content nor the pentaerythritol content is excessive; for example, materials P1 and P2 possess satisfactory durability, whilst materials P3, P4 and P5 are considerably less durable. This result was expected, and samples P1 and P2 more nearly represent current production formulations.
- (iv) In the case of the mixed alkyds PG1 and PG2, the disadvantage of the increased phthalic anhydride content or pentaerythritol content is also apparent, PG1 being superior to PG2, the former material being classified together with the glycerol alkyds and the latter with the pentaerythritol type materials.

Mechanical Properties of the Detached Films

The results obtained for the present series of tests are summarised in Table XXIII.

Examination of the results shows some points of interest which are outside the range of the likely experimental errors and reproducibility of the test procedure.

- (i) The glycerol alkyds show similar values for the Young's modulus and the percentage recovery of the film, and the effect due to increasing phthalic anhydride content or glycerol content is small.
- (ii) In the case of the pentaerythritol alkyds, the value of the modulus increases with increasing phthalic anhydride or pentaerythritol content. In fact, the material P1 gave a film of very low strength and was not capable of assessment on the apparatus and loads used, whilst P5 material possessed great strength which withstood a load of 500 g. applied to 1 cm. wide film. It will also be seen that the recovery of the film in the selected time interval of fifteen minutes after removal of the load fell to practically 50 per cent with the materials of higher phthalic anhydride and pentaerythritol contents.

TABLE XXIII

DATA OBTAINED FROM EXAMINATION OF EXTENSION/TIME CURVES FOR
SPECIFIED LOADS ON ALKYD MEDIA ETC.

Material	Applied load (g.)	Extension (15 min.) (%)	Recovery (%)	Film thickness (μ)	Force to produce 100% extension (dyne/cm.) $\times 10^{-6}$
G1	20 50*	65 90*	95 Film breaks	37.4	8
G2	20 50*	59 89.3	93 Film breaks	52.2	6.5
G3	20 50	44.3 92.6	92 93	52.2	8.5 10.5
G4	20 50	40.3 78.3	96 90	46.7	10.5 13.5
G5	20 50	41.6 85.1	87 91	37.4	12.5 15.5
P1	20* 50*	29.0 56.4	Film breaks	48.5	16
P2	20 50	26.7 49.5	93 100	37.4	19.5 26.5
P3	50 100 200	10.7 24.1 50.5	50 55 62	44.8	102 91 87
P4	100 200 300	5.4 20.3 42.4	48 49 49	44.8	406 215 155
P5	200 500	7.9 50.1	55 40	37.4	660 260
PG.1	50 100*	34.3 65.5	97 Film breaks	52.2	28 —
PG.2	50 100	34.9 58.9	81 81	37.4	38 45
Std. I Spec. (1/7)	50 100	52.6 94.8	84 80	35.5	26 29
Std. II Spec. (6/4)	50 100	44.6 74.5	65 74	56.0	19.5 23.5

*Indicates film broke under the applied load before 15 min. interval expired.

- (iii) The mixed alkyds PG1 and PG2 also showed a similar loss of the power of recovery with increasing phthalic anhydride and pentaerythritol contents, but this was less marked than for the pentaerythritol class materials.
- (iv) The standard materials showed rather unexpected results since the synthetic varnish showed a lower recovery than expected, whilst the oleoresinous varnish showed higher strength than similar materials previously examined.

SUMMARY OF RESULTS—PART TWO

From the results obtained in the various sections of this investigation, it is possible to connect variations in the chemical composition of alkyd resin media with differences in behaviour. In the water absorption measurements of the films, the alkyds based on pentaerythritol gave lower values than those based on glycerol alkyds, whilst both types were considerably less water absorbent than the oleoresinous varnish. Differences in the evaporation (permeability) constants were found and, although the significance of these results has not been clearly explained, the evaporation rate appears to bear some relation to the phthalic anhydride content. In contrast to this, the water absorption of other types of alkyd resins appeared to be unaffected by variations in the phthalic anhydride content.

In the adhesion tests on the dry films (of the initial process) adhesion failures were obtained with those based on pentaerythritol with higher phthalic acid contents, whilst the glycerol alkyds showed no failures. When the materials were used for revarnishing, however, the differences were less marked between these two classes of material, although the best results were obtained on materials of low phthalic anhydride content. All alkyds showed considerable loss of adhesion when tested in the "wet" state and there was some evidence to relate differences with variations in chemical composition, notably with regard to location of failure, which was at varnish level for pentaerythritol types but primer metal level for normal alkyds. The site of the minimum interfacial adhesion is of considerable practical importance. Service failures are more likely to occur when the adhesion is lowest near the surface, since under damp conditions the outer part of the film may not possess adequate mechanical strength to resist displacement. Cleaning operations, for example, may cause the film to break and result in its stripping; varnish/varnish adhesion is, therefore, of greatest importance.

The effect of composition (within the limits of the test) on durability can be summarised as follows. Glycerol alkyds, irrespective of the phthalic anhydride content, and the pentaerythritol alkyds with low phthalic content, showed satisfactory durability. The pentaerythritol alkyds of higher phthalic content showed somewhat inferior durability to the others except the oleoresinous varnish. Finally, the study of the mechanical properties of the detached films showed considerable differences which could be associated with differences in composition which are described in detail in Table XXIII, the determination being made on the films after accelerated weathering.

The result of the comparisons is summarised below. The minimum adhesion of the paint process finished with the various varnishes under humid conditions

(except for position of failure within the process) could not be correlated with any physical property studied. In the case of the "dry" adhesion data obtained on the original process, there is some evidence to indicate that the materials possessing the highest modulus of elasticity and lowest percentage recovery figures show lower adhesion of the varnish coatings (see results for materials P3, P4 and P5). It is, however, felt that the film property which leads to the loss of adhesion under humid conditions of the alkyd type materials has not been identified in the present series of tests, but the substrate has an effect.

If the alkyd materials alone are considered, it is interesting to note that there is a considerable correlation between the power of recovery of the film after extension, the modulus of the film and the durability, as assessed by the exposure to accelerated weathering tests. In fact, it would appear safe to assume that satisfactory durability will only be given by a material if it shows a high percentage recovery from the extended condition, and provided its modulus does not exceed 30×10^6 dyne/cm.² as illustrated in the following table.

TABLE SHOWING CORRELATION BETWEEN DURABILITY AND PHYSICAL CHARACTERISTICS OF THE FILM

Durability rating	Material	Mean Recovery (%)	Mean modulus of film (Force to produce 100% extension within 15 mins.) (dyne/cm. ²) $\times 10^{-6}$
Group I Materials of satisfactory durability	G1	95	8
	G2	93	6.5
	G3	93	9.5
	G4	93	12
	G5	89	14
	P1	—	16
	P2	96	23
	PG1	97	28
	Standard I	82	28
Group II Materials of inferior durability	PG2	80	42
	P3	55	90
	P4	49	180 (approx.)
	P5	47	250
			„

The oleoresinous varnish (Std. II), which possesses inferior durability, does not completely fulfil the above conditions and is exceptional in its behaviour. This is believed to be due to the fact that the mechanical properties were only measured after ageing in the Laboratory under normal conditions. If different classes of material are to be evaluated, it would obviously be necessary to concentrate on differences in behaviour after exposure to ultra-violet light. It can, however, be assumed that the effect of the ultra-violet light and exposure on the alkyd systems under consideration is similar, and the differences in mechanical properties give a reliable index to the durability of the systems. Early experiments using a wider range of alkyd materials from varied sources gave results which support the above conclusion.

CONCLUSIONS AND DISCUSSION

The work described in this paper has been carried out as part of the normal work of the Laboratory, in pursuance of the aim to evolve a system of physical tests which will enable a paint to be defined in terms of its physical properties. In this instance the immediate aim was to eliminate a weakness in a particular painting system, which otherwise has shown remarkable durability and fitness for its purposes. Considerable progress has been made towards this limited objective, and the area within which the ultimate solution may be found has been defined, and a method of test has been developed. A difficulty which has yet to be solved is that of defining the limiting adhesion necessary; it is clear from the work already carried out that this value must be lower than is generally supposed, since tests carried out in the field have revealed a very low adhesion to exist even when the service performance is entirely satisfactory. It is clear that the location of the interface of minimum adhesion in the film is very important. As it affects the probability of a disturbance of the film which will cause stripping, deep seated cleavage is less important than partial loss of adhesion in the final coat. The method used for the measurement of adhesion referred to in the paper has been quite successfully employed in the measurement of the value of various types of surface preparation of both steel and aluminium, and has enabled a suitable system of surface preparation to be worked out which will ensure good adhesion even to heat hardened aluminium alloy surfaces.

It seems important to differentiate between paint adhesion, as measured by the method described, and impact or "chip" resistance, to which quite different criteria apply. The two properties, though related, do have quite different practical implications. The interesting relationship between film modulus, extensibility, film recovery and durability which has been described could well provide a useful contribution to the methods of assessing paint durability, and it is hoped to develop this further. Other methods which may be developed in connection with this important aspect of paint control and evaluation include the measurement of volume changes in the film under moist conditions, and loss on hydrolysis. A useful indication of durability may also be obtained from the absorption spectra and here again a preliminary test gave quite positive indications of a useful relationship. The technique proved a little difficult, but there is every indication that here is a further useful avenue to explore.

ACKNOWLEDGEMENTS

The authors would like to thank Mr. T. M. Herbert, Director of Research, British Railways, for granting permission to publish this paper, and Mr. F. Fancutt, Assistant Director of Research, for his encouragement and guidance in the work. They would also like to express their appreciation for the contribution made by Mr. Maxwell Wickett, for the considerable part he has played in the design of the adhesion apparatus, and in the conduct of the laboratory work.

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DISCUSSION

DR. E. SUNDERLAND criticised the paper because it presented a large amount of data, which most people unfortunately had not the time to analyse, together with a number of conclusions which most people read and assumed to be well founded. The authors had wisely refrained from drawing too general conclusions, but they had implied that little correlation had been found between the chemical constitution of an alkyd and its physical properties. Further, the authors did not recommend in some cases the use of pentaerythritol or of D.C.O. As an alkyd manufacturer, Dr. Sunderland felt that the chemical constitution of an alkyd resin was definitely related to its physical properties. He believed that most alkyd chemists had long concluded that to express the constitution of an alkyd in terms of the percentages of fatty acid and phthalic anhydride contents tended to hide rather than clarify the nature of the alkyd. Dr. Sunderland said that he would not expect these parameters to show any correlation with physical or other properties. There were, he felt, other more significant parameters.

However, if a number of alkyds were to be studied, as in this case, it would be expected that the percentages would be expressed on a common basis. In Part Two of the paper that was done and percentages were given in terms of the original charge. The only unknown quantity here was the nature of the pentaerythritol, which could, however, be deduced to be monopentaerythritol. In Part One, where the bulk of the data appeared, not only had different bases of calculation apparently been used, but also where mixed oils or resin additions were used no data had been given. In certain series, *e.g.* A and B, the alkyds were not characterised at all; only a single parameter, fatty acid content, was given.

TABLE A

Series	Hydroxyl excess (%) based on	
	Product	Charge
G.1.	— 3	+ 38
G.2.	+ 169	+ 210
G.3.	+ 15	+ 59
G.4.	— 21	+ 21
G.5.	+ 71	+ 103
G.6.	— 29	+ 11
G.7.	— 66	— 25

For Series G (Part One), Dr. Sunderland said that he had calculated the hydroxyl excess on two bases (Table A); in the first column on the assumption that the percentages were based on product, and in the second on the assumption that they were based on initial charge. Those which were underlined were, in his opinion, probable alkyds, and there were two in each column. The other three seemed to him to be highly improbable or even impossible alkyds by either method of calculation, and he could only assume that the figures given were erroneous. He suggested that the authors might have obtained these data from alkyd manufacturers having individual methods of expressing percentages, and he felt that quite a number of

percentage bases could easily be devised. It was clear that a more useful, universally accepted, method of expressing alkyd constitution was required.

Dr. Sunderland reiterated that he would not expect a correlation between percentage composition and physical properties, even if all percentages were on the same basis, so that confusion would inevitably result from the choice of varying bases. However, as the authors had shown a clear correlation between accelerated durability and the elasticity of the film as expressed by its percentage recovery, it seemed worthwhile to attempt a correlation of these properties with alkyd composition. If the correct alkyd parameter were chosen, there appeared to be a significant correlation. He said that he had calculated the esterification functionalities for those alkyds where there did not seem to be much doubt about the percentage basis and had plotted these against percentage recovery. The result, which he illustrated with a slide, showed that up to a functionality of about 1.98, high and substantially constant recoveries were obtained. Above that functionality there was a rapid approximately linear drop in recovery. As a functionality of 2 represented a critical point when considering an alkyd molecular build-up, it was of considerable interest to find a sharp break there. The theoretical and practical implications of this correlation were clearly of great interest.

Dr. Sunderland felt that in papers involving alkyds, the constitution of the resins should at least be given in an unequivocal form so that others might derive benefit from them. Finally, he said that in his opinion the calculation of esterification functionalities should *not* be made by the method presented by Carmody (*Official Digest*, 1958).

MR. DUNKLEY thanked Dr. Sunderland for his remarks and said that the alkyds which the authors used were actually available from manufacturers; they were informed of the compositions and the figures were reliable. Dr. Sunderland had criticised the way in which the compositions were stated, but the authors accepted the alkyds offered to them and tried to determine their constitution and their characteristics. The percentage of phthalic anhydride could be determined with accuracy and reproducibility.

DR. D. P. EARP added that the alkyds used were selected from catalogues; they were from different manufacturers, whether or not any significant results were obtained from them. The authors had aimed to obtain the chemical specifications of the alkyds, but so far it had not been possible.

MR. W. H. MOSS, having noted that the work reported was carried out in the laboratory, asked if the authors had information on the effect of higher or lower temperatures, particularly the lower temperatures, on adhesion. He said that it was commonly thought that, whatever caused damage to a paint, the actual cracking and peeling occurred at low temperatures. Mr. Moss further inquired whether the authors included refrigeration in their accelerated test cycle. There seemed much difference of opinion as to whether it should be included and at what temperatures. His experience was that refrigeration helped in differentiating between various materials.

DR. EARP replied that most of the experiments were carried out at laboratory temperatures of 65-70°F, and no measurements on the effect of lower temperatures on adhesion had been made. The use of a refrigerator had been considered in the past; the idea had been discarded because if a very low temperature refrigerator were used which gave solid carbon dioxide, and the panels were wet, failures occurred which were not comparable with those which occurred in service. If an ordinary refrigerator operating at temperatures a few degrees below zero was used, the panels would be subjected to conditions already covered by other tests.

MR. DUNKLEY added that throughout the laboratory work it was held as a criterion that the types of failure reproduced must have some relation to those which occurred in practice. To have increased or decreased the temperatures used would have meant that the types of failure which occurred in practice would not have been reproduced. It was for that reason that the authors had not found it necessary or desirable to use temperatures other than those experienced in the laboratory. Further, they had found that adhesion failure was likely to occur perhaps less in the winter months than in the summer months; failure to adhere would seem to depend more on humidity than on temperature.

DR. V. R. GRAY said that in his own paper (to be published in the November issue of the *Journal*), which dealt with surface coatings on wood, an attempt had been made to calculate the energy change on application of a surface coating. The figures given in Table I of the authors' paper for the energy of the removal of films were much higher than his own (some 500 to 50,000 times as great) and these were on wet films. Possible explanations of this were the slow development of chemical bonds and electrostatic effects. The latter explanation, which was unlikely to apply to wet films, would lead to variation in adhesion with rate of loading. The former would be more likely on metal than on wood or old varnish.

Commenting on the effect of the rate of application of load, he thought that it was generally agreed that if experiments were carried out at lower rates of loading there was a tendency for lower adhesion values to be obtained. In practice, for instance, the rate of application of load might be quite low, and Dr. Gray suggested that some of the differences in results might have been due to shrinkage or to some slow change in other properties which would result in much lower adhesion values being obtained in those conditions. He asked if the authors had found the adhesion values to be lower on wood than on metals, or low at low rates of loading.

DR. EARP said that in the particular series of tests described measurements had been confined to coatings on various metal panels of steel or alloys. The rates of loading in the first series of tests had been limited, which explained why the strain gauge had been used, but for high rates of loading the Charpy impact type of instrument had been employed. The authors did not correlate the adhesion results obtained by the two different methods, but had found that with the rapid application of load the failures were higher up in the film.

MR. G. PHILLIPS remarked that, unlike Dr. Sunderland, he had found the paper to be extremely interesting and very valuable. There were in the paper many examples where the constitution and the physical properties were quite closely related. Dr. Sunderland had himself calculated values in two different ways although he criticised other manufacturers for doing the same. Mr. Phillips agreed that a system of standardisation was indeed necessary.

With regard to the adhesion tests, he wondered how long it took to set up the apparatus. In Table IV the authors had given figures for film adhesion after four days' immersion and for the film adhesion after varying drying periods. On reading the paper Mr. Phillips said that he had assumed that it would take about ten minutes to make the measurements, so that ten minutes had to be added to the times given. The authors had obtained adhesion values which agreed very closely with those he had obtained and had reported in his own paper (*J.O.C.C.A.*, 1961, **44**, 575 *et seq.*).

DR. EARP said that in general the time required for preparation before the measurements were made was probably about five minutes.

MR. P. F. BRIDLE asked whether Dr. Earp could be more explicit about the nature of the rosin, rosin modified phenolic and pure phenolic modified alkyds mentioned in Table VIII, the value and performance of which had been condemned. He said that the request was prompted by the fact that two of these modified alkyds were

reported as "glycerol alkyds plus rosin", suggesting that rosin was present only as an adulterant. He could not believe that any paint manufacturer used rosin in such a manner, certainly not at its current high price, and therefore he suggested that there would be little value in studying such a system.

DR. EARP said that the information with regard to the rosins and the modified alkyds was given in the manufacturers' catalogues.

MR. BRIDLE commented that Dr. Earp's reply suggested that those two alkyds were in fact described in the literature as "rosin modified alkyds", and in that case their description in the paper seemed to be somewhat misleading.

MR. P. WHITELEY took the opportunity to draw attention to the undesirable multiplication of scales of assessment with which paint technology was burdened. Reference to the literature, he said, would show scales of four, five, six, seven, eight and ten stages. In addition, various methods of rating different properties or defects had been used. The authors of the paper had adopted a particularly unusual system with unequal steps and with a lower rating for checking than for gloss retention. The necessity to think in fresh terms to suit each such paper published made their study far more difficult, and he suggested that the Association, possibly in liaison with the Paint Research Station, make recommendations to simplify and unify the position.

While many authors seemed to find need for fine divisions in assessment, he had difficulty in maintaining consistent judgment of the more subjective properties, even over the scale of five stages which seemed most suitable for research purposes. For paint users he proposed an even simpler scale with only three stages: no visible defect or change; defect visible but not serious; defect serious enough to require repainting. It must also be remembered, he added, that numerical values arbitrarily assigned to subjective judgments on scales of the type "good/fair/bad" did not form a ratio scale and could not properly be treated by ordinary arithmetical or statistical methods. In such ordinal scales, two times two might not equal four, and ten (or any maximum rating) might well be infinity. Great care must therefore be taken in interpreting data such as the authors had presented in the paper, since their summation and averaging were not strictly permissible. However, he admitted to having been compelled to do the same thing himself.

MR. DUNKLEY agreed that methods of test and methods of recording results, on which Dr. Earp and himself held very strong views, should be standardised. It was also most important that they should be as simple as possible, and that questions of order of merit and relative performance and the number of steps should be as few as possible. The system at Derby was originally of four steps, but as the work had proceeded a number of further steps had been added.

At the British Railways' laboratory the authors were dealing with paints and had actually carried out classifying tests, which they could do with reasonable accuracy and in reasonable time; having got the information, they had to correlate it with the performance expected in service. They were able to measure certain properties in the laboratory which, although not directly related to those in practice, could be correlated with them. Mr. Dunkley said that he was quite sure that when a final decision was made and a specification drafted, it would be based somewhat on the lines of the information given in the paper.

The correct interpretation of physical testing results could be achieved only if regard was given to the type of materials to which the tests applied. In all the tests the authors had exposed a standard of the same chemical class for a long period, and in that way had been able to determine a correlation between accelerated tests and actual performance.

Solar Reflectivity of Paints

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Summary

The reflective efficiency of a surface can only be defined if the energy at each wavelength of the source is known. It is not very practical to measure efficiency by the heat absorbed when radiation from the source falls upon it. If the reflectivity is known over the waveband of the source, then the reflective efficiency can be calculated by integrating the energy reflected over the radiation spectrum. The solar energy spectrum approximates to a black-body radiator at 6000°K without radiation below 0.3 μ .

White painted surfaces are less efficient than the best polished metals, but are preferable because of stability and protective value. Rutile titanium dioxide pigmented paints have the best solar reflectivity, despite significant absorption in the ultra-violet region, and can have an efficiency of 77.6 per cent. For any white paint system, increase in radiation wavelength is associated with increased penetration of the film. Since ultra-violet radiation is reflected near the surface a coating containing a non-absorbing pigment, such as basic white lead, can increase the reflectivity of a rutile titanium dioxide paint to 82.6 per cent.

A simple theoretical model of the reflective process has been devised which gives good quantitative agreement with experimental results.

INTRODUCTION

When radiation falls on a surface of a reflective material, it may be reflected as in the ideal case of a polished metal, or it may be partially reflected where the process is one of reflection, refraction and transmission. The last occurs when radiation passes through a paint film containing a "white" pigment which has a different refractive index from that of the medium.

Any radiation which is not reflected from a surface, irrespective of the mechanism, is converted to heat whatever its wavelength and irrespective of the direction of the reflected radiation, *i.e.* there is no difference between diffuse or specular reflection. The efficiency of a reflective surface can be defined uniquely by the amount of energy it absorbs under the stated conditions. In practice the temperature to which a surface will rise above the ambient temperature when radiation falls upon it will be governed by the quantity of heat absorbed, by the heat capacity and conduction of the surface system, and by the amount of heat which is lost by convection and re-radiation. If it is desired to keep the surface as cool as possible, everything is to be gained by having a high surface reflectivity, because this is in addition to what can be effected by optimum design of the structure and conventional methods of heat exchange, such as water cooling. Loss of heat by convection can be appreciable, but it is a function of the design and position of the surface. Loss of heat by true re-radiation can be small by comparison with convection and, because it is proportional to the fourth power of the absolute temperature, only becomes significant at high temperatures. Assuming a constant coefficient of emissivity, therefore, and an ambient temperature of 0°C, the ratio of heat lost at 100°, 200° and 300°C relative to that lost at 20°C is 3.4, 8.7 and 18.9 respectively. Over a temperature range such as this it is not safe to assume constant emissivity since it is well understood, but not yet studied in detail,

that surface characteristics can vary significantly with respect to energy in the different infra-red regions. This paper deals with the amount of heat absorbed from different temperature sources, with particular reference to solar radiation.

The effect of solar radiation on different comparative surfaces has been the subject of many practical studies, but the part played by the surface alone cannot be separated from the heat lost by convection and radiation. By exposing panels at 45° to the vertical to the sun in July-August, Cottony and Dill found¹ the relative temperature rise for various surface coatings (see table below).

Surface coating	Temperature (°C)
Lamp black	100
White paint	28
Canary yellow paint	41
Aluminium paint	63
Slate paint	70
Medium green paint	92
Galvanised iron (unpainted) ..	69

The high efficiency of white paints has been observed² on galvanised steel sun-shades in the Imperial Valley, California, where their use reduced the surface temperature by 28°C .

The idea that high solar reflection is achieved by high efficiency in the infra-red has been found³ to be incorrect in practice by observing the temperature rise and evaporation losses of cans containing petrol. This paper shows how the reflective efficiency of a surface against different sources can be determined by a precise knowledge of the reflectivity over the wave band peculiar to the source.

ASSESSMENT OF REFLECTANCE

Radiation Sources

The reflectivity of a surface is not constant with change in wavelength; the energy at different wavelengths for each source must therefore be known. The energy distribution with wavelength for theoretical black body radiators at 500° , 1000° , 3000° and 6000°K , plotted so that the total energy is constant, is shown in Fig. 1. The energy distribution from the sun is similar to that of a 6000°K radiator, but with no energy below 0.3μ . In order to reflect the most energy from a 6000°K source the reflectivity in the visible region (about 0.5μ) is very important, whilst the reflectivity in the infra-red region (1.5 to 6μ) cannot have much effect, the reverse being the case against a 1000°K source. Therefore, if the sources of radiation are different, each surface must be considered in relation to the significant wavelengths involved. In the case of solar reflectivity, the reflectivity must be known over a wider wave band (from

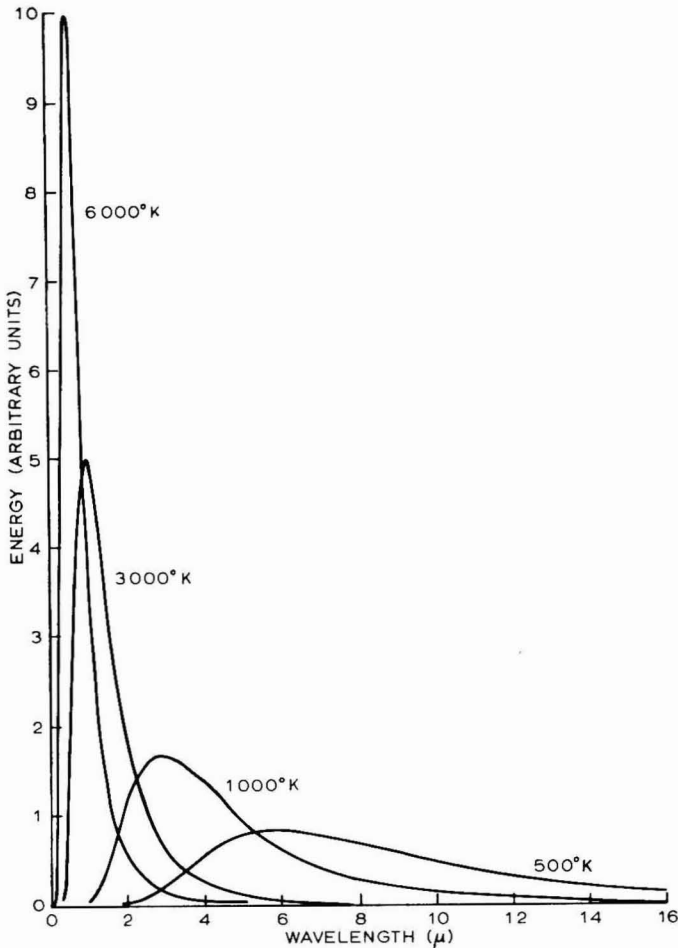


FIG. 1. BLACK BODY RADIATION AT DIFFERENT TEMPERATURES
(total energy the same)

0.3-5 μ) than is usual for white paints, but measurement to 2.2 μ only introduces very little error. No relaxation of measurement can be made in the ultra-violet portion of the spectrum as it will be demonstrated later that this is most significant.

Reflective Efficiency

If different surfaces are exposed under identical conditions in air to the same radiation, then the temperature rise will give a good indication of the reflective efficiency of each system. The part played by the surface only cannot be separated from the loss of energy by convection and radiation, which are not usually constant for samples of different reflectivity. Convection can be minimised by performing the experiment *in vacuo*, but this requires complicated apparatus. If, however, the distribution of energy of the radiation source is known at all wavelengths of the spectrum and the reflectivity of the surface

is known over the same spectrum, then the energy reflected can be calculated at small wavelength intervals. The sum of these is the total energy reflected and its relation to that of the original source is the integrated reflectivity or reflective efficiency (ΣE) against that particular source; it can be expressed as a coefficient or percentage. In practice this process is best done graphically. For materials of high reflective efficiency, it is clearer to plot the integrated absorption against wavelength and to measure the area of this for comparison with that of the total energy of the source. Provided that the total hemispherical reflectivity of the surface is measured, the reflective efficiency is independent of whether the surface is matt or glossy. In practice, however, a glossy surface would only be inferior to a matt one if the geometry of the coated item was such that the energy specularly reflected underwent further reflections with associated absorption.

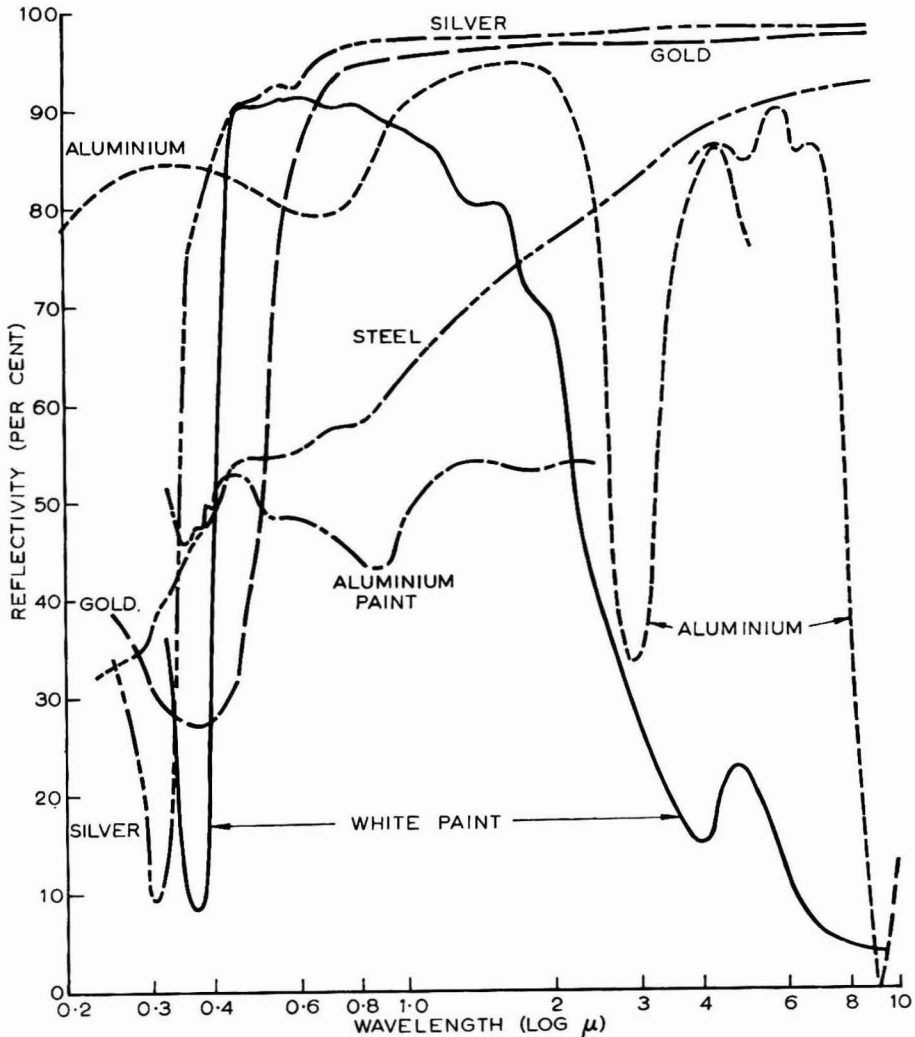


FIG. 2. REFLECTIVITY OF DIFFERENT SURFACES BETWEEN 0.2 AND 10 μ

Surfaces of highest solar reflectivity must appear white to the eye, but with less reflective surfaces the colour to the eye may not be significant. For example, a black anodised aluminium surface has good reflectivity in the infra-red region with an integrated solar reflectivity of 18.2 per cent instead of approaching zero, as would be expected from its visual blackness.

Comparison of Polished Metal and Painted Surfaces

The choice between a polished metal surface and a highly reflective paint is one in which the calculated reflective efficiency (ΣE) for any particular source is the only guide. An accurate or specific value for any particular metal surface or paint cannot be obtained, as this is dependent to some extent upon the "condition" of the reflecting surface, which is difficult to define. This is particularly true of aluminium, which has an absorption band in the 3μ region associated with the oxide on the surface. It will be shown later that the thickness of the paint can have a significant effect. However, the choice between these two types of reflecting surfaces should be made particularly in relation to the temperature of the source.

Typical reflectivity values for gold, silver, steel⁴, aluminium^{5, 6} and white⁷ and aluminium paint at different wavelengths are shown in Fig. 2. The graph connects the reflectivity and the logarithm of wavelength, the latter scale being used for convenience only. The reflective efficiency (ΣE) of these surfaces against different temperature sources is given in Table I. Silver has the best reflective efficiency against sources from 500° to 6000°K (solar). Gold is inferior to silver against solar radiation because of its absorption in the visible region, but against sources between 500° and 3000°K it is comparable with it. Aluminium, which is intermediate between silver and gold against solar radiation, falls off in efficiency with sources of lower temperature.

TABLE I
TYPICAL REFLECTIVE EFFICIENCY OF VARIOUS SURFACES AGAINST DIFFERENT TEMPERATURE SOURCES

Surface	Reflective efficiency (ΣE) against: (%)			
	Solar radiation	3000°K	1000°K	500°K
Silver	91.1	97.3	98.4	99.8
Gold	73.4	94.2	97.2	99.2
Aluminium	84.4	86.1	74.1	73.1
Mild steel	57.0	68.6	85.6	90.3
White paint	78.7	75.2	25.9	22.7
Aluminium paint	48.9	51.4	—	—

On the other hand mild steel, which has a reflective efficiency of 57.0 per cent against solar radiation, progressively increases to 90.3 per cent against a 500°K

source. The solar value for aluminium paint, which at about 50 per cent is typical for paints of this type, does not offer much advantage since it absorbs about twice as much as white paint, although the latter absorbs about twice as much as silver. However, the liability of silver to tarnish rules out its use except in special cases. Polished aluminium can be more efficient than white paint, but since aluminium deteriorates on weathering, or needs paint to prevent deterioration, then a white painted surface would appear to be the best and simplest choice for a surface of good solar reflectivity.

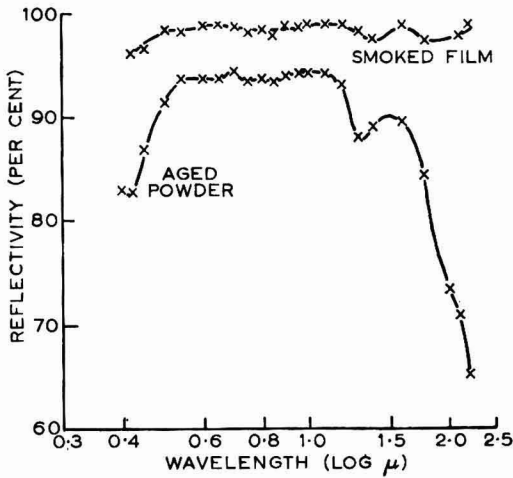


FIG. 3. REFLECTIVITY OF SMOKED AND AGED MAGNESIUM OXIDE

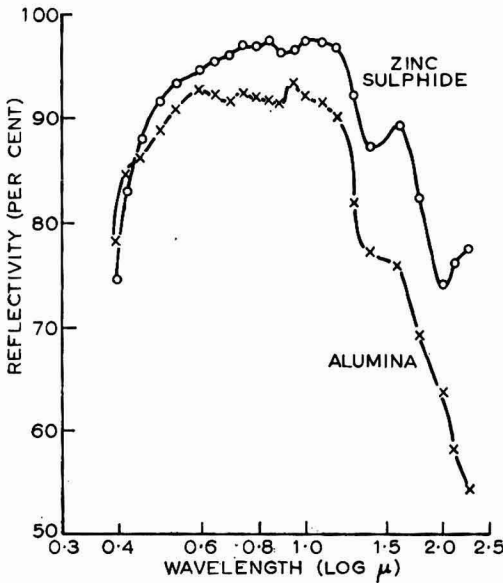


FIG. 4

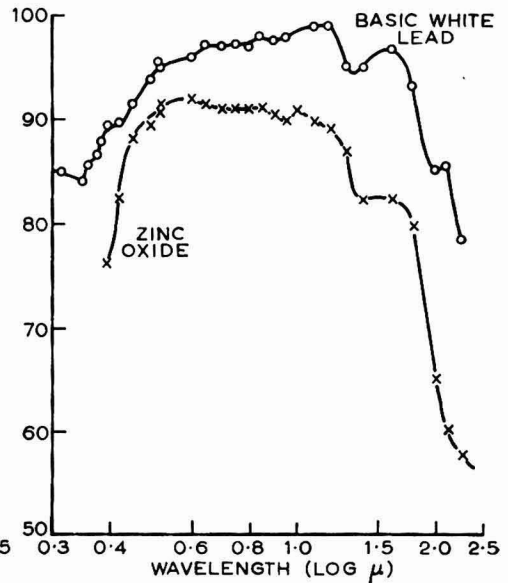


FIG. 5

REFLECTIVITIES OF ZINC SULPHIDE AND ALUMINA POWDER (FIG. 4) AND ZINC OXIDE AND BASIC WHITE LEAD POWDER (FIG. 5)

Comparison of White Pigments

If a white paint shows the best practical surface for solar reflectivity, a knowledge of the different pigments available is most desirable. A preliminary study of likely pigments, either in the form of pressed blocks (bound with a minimum of high grade gelatine) or in a resin medium, was therefore made.

Experimental Method

The sample was placed at the centre of an aluminium sphere coated inside with a good titanium dioxide pigmented oil undercoat, followed by a layer of titanium dioxide bound with gelatine and a layer of magnesium carbonate bound with gelatine. The sample was illuminated with "white" light indirectly by multiple reflections from the inside of the sphere by eight 240 watt (26 volt) tungsten filament lamps assembled in a ring, the plane of which was just behind

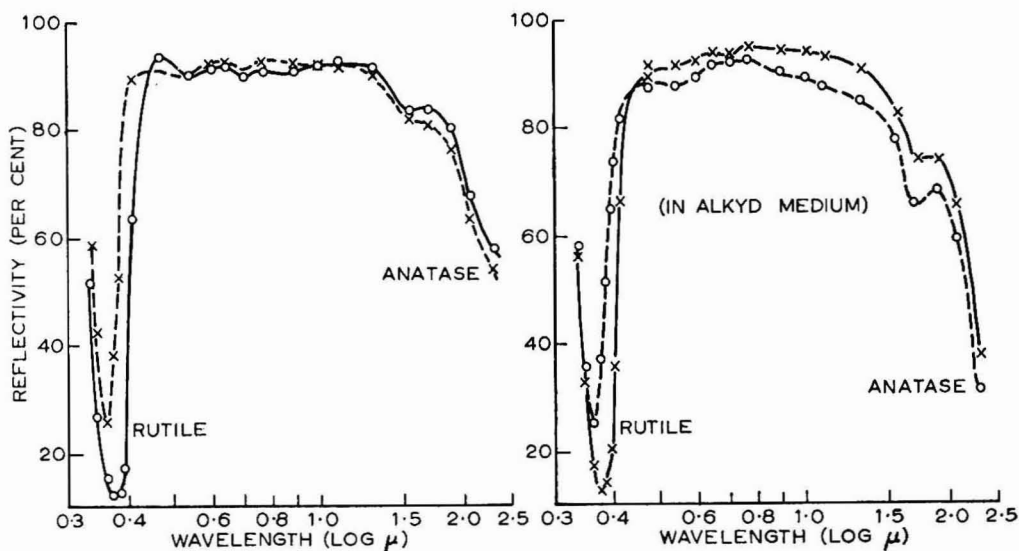


FIG. 6

FIG. 7

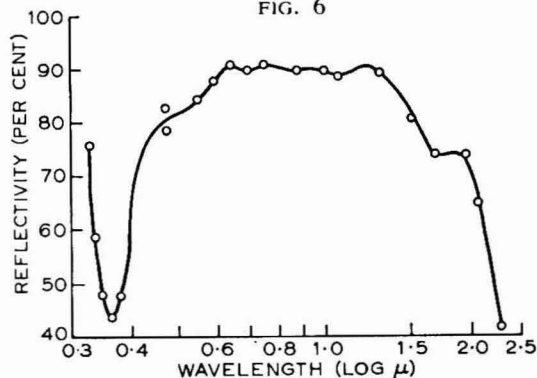


FIG. 8. REFLECTIVITY OF BASIC WHITE LEAD IN ALKYD MEDIUM

(Above)
REFLECTIVITY OF RUTILE AND ANATASE
TITANIUM OXIDE POWDER ALONE (FIG. 6)
AND IN ALKYD MEDIUM (FIG. 7)

that of the sample. The radiation emerging from the sphere opposite the sample was focused on to the entrance slit of a Leiss double monochromator fitted with a quartz prism⁸. A chopper was placed in front of the entrance slit to interrupt the radiation at 800 c/s and the detector used had an amplifier tuned to this frequency. The detector was situated in a light-tight box behind the exit slit of the monochromator. Measurements were made by setting the spectrometer to pass a particular wavelength and comparing the energy radiated from the sample with that reflected by the sphere lining when the sample was not in position. In the ultra-violet region the reflectivity of the lining, the energy emitted from the tungsten lamps, the transmittance of the spectrometer and the sensitivity of the detectors fall off; this method therefore becomes very insensitive below 0.4 μ .

Experimental Results

The reflectivity results obtained with different pigments are shown graphically in Figs. 3, 4, 5 and 6, and for rutile and anatase titanium dioxide and basic white lead in an alkyd medium in Figs. 7 and 8. The integrated reflective efficiency (ΣE) against solar radiation (omitting the region 0.3-0.4 μ where the values of reflectivity in this region are not available) is given in Table II. The dry pigments give good reflectivity, but this is considerably reduced when incorporated in a medium, even for rutile and anatase titanium dioxide, both of which have a very high refractive index for transparent materials.

TABLE II
REFLECTIVE EFFICIENCY OF WHITE PIGMENTS TO SOLAR RADIATION

Pigment	Reflective efficiency (ΣE) (%)	
	Dry pigment	In medium
Smoked magnesium oxide	96	—
Compressed aged magnesium oxide ..	91*	—
Aluminium oxide	87*	—
Zinc sulphide	92*	—
Zinc oxide	88*	—
Basic lead carbonate	95.6	79.7
Rutile titanium dioxide	84.7	77.6
Anatase titanium dioxide	87.6	77.5

*Integrated only between 0.4 and 2.2 μ .

DETAILED STUDY OF RUTILE PIGMENTED COATINGS

Tests of the reflective efficiency of rutile titanium dioxide in three different media have been determined for different film weights. Each finish was applied over different filler coats because it was considered that with lightweight finish

coats the colour of the filler (undercoat) would play an important part in the overall efficiency of the paint scheme. A chromated etch primer was common to all schemes, which were applied to 22 s.w.g. hard aluminium panels. The schemes which were examined are given below; percentages of titanium dioxide are percentages by weight of dry film.

- A. Nitrocellulose containing 20 per cent titanium dioxide on (a) a black cellulose filler, (b) a standard white filler.
- B. Alkyd containing 33 per cent titanium dioxide on (a) a black synthetic filler, (b) a standard grey synthetic filler, (c) a white filler, as (b) but with the trace of black omitted, (d) an extra white filler, as (c) with rutile added.
- C. Epoxy/amine adduct containing 55 per cent titanium dioxide on (a) a black, (b) a cream, or (c) a white epoxide-based filler.

Full details of the panels examined are given in Tables III, IV and V. For convenience these tables record the reflective efficiency (against solar radiation) and in the case of the epoxy/amine adduct (Table V) the calculated reflective efficiency of the top coat and filler, details of which are discussed later.

TABLE III
REFLECTIVE EFFICIENCY OF RUTILE TITANIUM DIOXIDE IN NITROCELLULOSE MEDIUM
(Scheme A)

Etch primer (oz./yd. ²)	Filler		Finish		Reflective efficiency (%)
	Type	Weight (oz./yd. ²)	Type	Weight (oz./yd. ²)	
0.46	Black cellulose	1.26	—	—	4.8
0.41	„	1.24	Cellulose	1.10	52.7
0.47	„	1.27	„	2.10	69.6
0.39	„	1.02	„	3.00	73.7
0.38	„	1.00	„	3.95	74.9
0.52	„	1.00	„	5.00	76.2
0.43	Standard white cellulose	1.28	—	—	62.4
0.39		1.27	„	1.00	70.9
0.39	„	1.14	„	2.08	74.4
0.48	„	1.27	„	3.10	77.4
0.57	„	1.27	„	3.90	77.2
0.55	„	1.14	„	4.98	80.1?

TABLE IV
REFLECTIVE EFFICIENCY OF RUTILE TITANIUM DIOXIDE IN ALKYD MEDIUM
(Scheme B)

Etch primer (oz./yd. ²)	Filler		Finish		Reflective efficiency (%)
	Type	Weight (oz./yd. ²)	Type	Weight (oz./yd. ²)	
0.38	Black synthetic	2.20	—	—	8.7
0.40	„	1.92	Alkyd	1.00	56.9
0.47	„	1.64	„	2.10	70.6
0.50	„	2.60	„	3.15	73.9
0.38	„	1.88	„	4.30	74.8
0.45	„	1.70	„	5.20	75.3
0.43	Standard grey synthetic	1.86	—	—	59.0
0.44	„	1.96	„	1.00	67.7
0.44	„	1.74	„	2.15	73.9
0.51	„	1.90	„	2.82	75.0
0.39	„	1.80	„	3.80	75.9
0.48	„	1.72	„	5.20	76.8
0.43	Standard white synthetic	1.45	—	—	64.8
0.40	„	1.22	„	1.04	68.9
0.50	„	1.22	„	2.10	73.6
0.46	„	1.42	„	3.10	76.1
0.39	„	1.38	„	4.10	77.0
0.43	„	1.48	„	5.20	76.5
0.49	Extra white synthetic	1.46	—	—	60.4
0.38	„	1.27	„	1.08	68.9
0.50	„	0.90	„	1.99	73.8
0.46	„	1.48	„	2.90	74.5
0.47	„	1.29	„	4.32	75.9
0.55	„	1.19	„	4.90	77.1

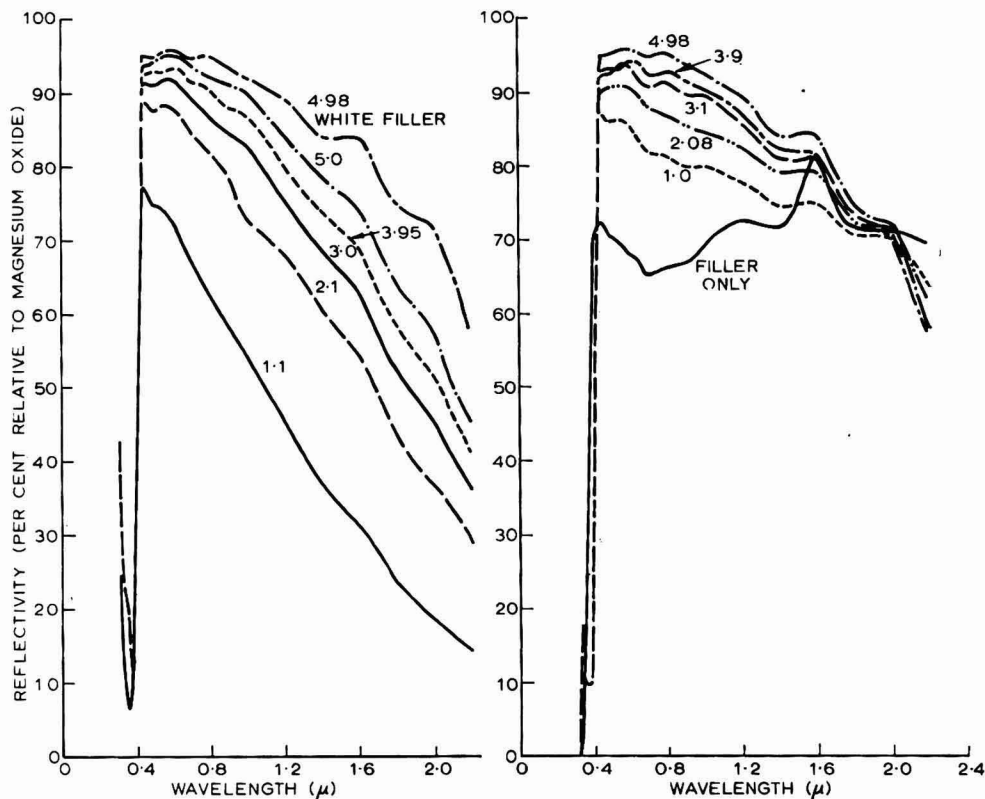


FIG. 9. ON BLACK FILLER (Scheme A (a)) FIG. 10. ON WHITE FILLER (Scheme A (b)) REFLECTIVITY OF RUTILE TITANIUM DIOXIDE IN NITROCELLULOSE MEDIUM. THE FIGURES ON THE CURVES INDICATE THE WEIGHT OF TOP COAT IN OZ./YD.²

The reflectivity of all panels was measured between 0.30 (or 0.32) and 2.2 μ by the method described earlier for the nitrocellulose and alkyd systems, and with a *Beckman Ratio Recording Spectrometer* for the epoxy/amine adduct system. The values obtained for the nitrocellulose paint on a black or white filler for different weights of top coat are shown graphically in Figs. 9 and 10. The plots of Figs. 9 and 10 are directly comparable as the weights of the several top coats are either the same or very similar; Fig. 9, which relates to a black filler, includes for ease of comparison the reflectivity of the maximum weight top coat on a white filler. These measurements were made relative to freshly prepared magnesium oxide and when used to calculate the solar reflective efficiency give results which have to be corrected by a factor of 0.96, because magnesium oxide has an integrated solar reflectivity of 96 per cent of absolute.

The results on a black filler are of academic interest in that they represent the amount of energy reflected by the pigment back to the surface prior to the rays penetrating to, and being completely absorbed by, the black substrate. At moderate and high top coat weights there is little difference between the black and white filler over the visible spectrum (0.4-0.75 μ) where the average pigment size is such as to give maximum scattering. There is, however, a

significant difference as the top coat weight is decreased or the wavelength increased.

For any particular weight of top coat, in general, the proportion of energy which reaches the substrate increases with increase in wavelength. The process can therefore be considered as one of increased penetration of energy as the wavelength increases. This reduction in reflectivity in the infra-red is less significant when the integrated solar reflective efficiency is considered.

* The reflectivity of the white undercoat alone (Fig. 10) is appreciably improved as the wavelength changes from 0.7 to 1.6 μ . This is because the

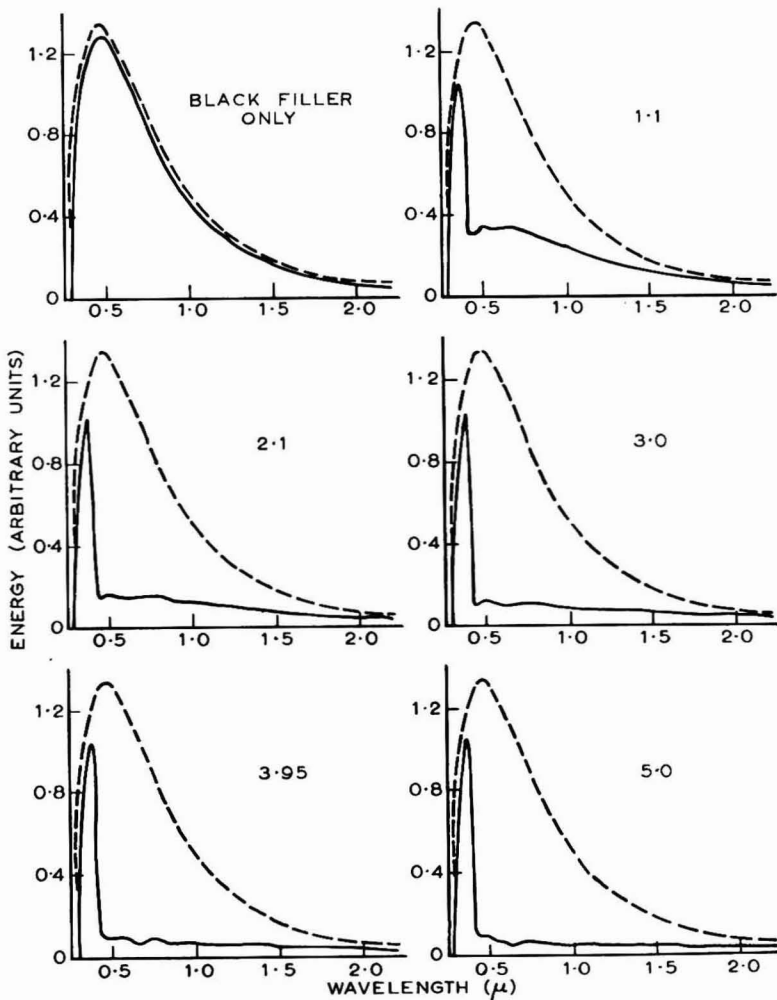


FIG. 11. ABSORPTION OF RUTILE TITANIUM DIOXIDE PIGMENTED NITROCELLULOSE FINISH ON A BLACK NITROCELLULOSE FILLER.

(The figures on the graphs indicate the finish weight in oz./yd.²)

————— Absorption curve of paint scheme
 - - - - - Solar spectrum

coating is comparatively thin to reflect radiation efficiently in this waveband and the energy which reaches the aluminium panel is reflected by it. Since more energy is transmitted to the aluminium as the wavelength increases and since the reflectivity of aluminium improves between 0.7 and 1.6 μ , reaching almost 95 per cent at 1.6 μ (see Fig. 2), then improved reflectivity in this region is to be expected. If a thin top coat is applied which has a reflectivity of less than 95 per cent, the net effect will be to reduce the reflectivity, which is the case at 1.6 μ . It is not until the top coat reaches a value of 3.10 oz./yd.² that its original value is restored. The reflectivity values shown in Figs. 9 and 10 for a nitrocellulose medium are also typical for an alkyd or epoxy/amine adduct medium.

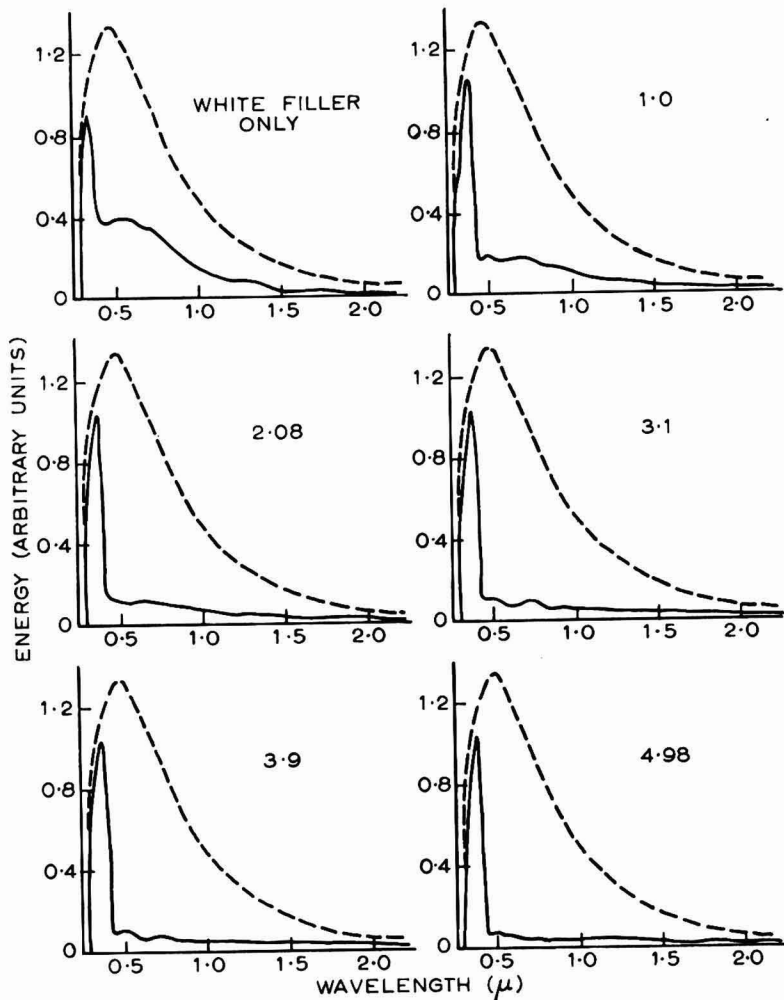


FIG. 12. ABSORPTION OF RUTILE TITANIUM DIOXIDE PIGMENTED NITROCELLULOSE FINISH ON A WHITE NITROCELLULOSE FILLER.

(The figures on the graphs indicate the finish weight in oz./yds.²)

————— Absorption curve of paint scheme
 - - - - - Solar spectrum

The integrated absorption curves against a solar source for the nitrocellulose scheme on a black or white filler are shown in Figs. 11 and 12; from these can be estimated the reflective (or absorptive) efficiency as described earlier. The results of these estimates and those for the other two media have been included in Tables III, IV and V and are shown graphically in Figs. 13, 14 and 16, against the weight of top coat; Fig. 16 also includes calculated values, details of which will be discussed later.

Referring to the integrated absorption curves in a nitrocellulose medium (Figs. 11 and 12), as the maximum efficiency is achieved in all cases the absorption in the ultra-violet region (0.3 to 0.4 μ) dominates the rest. Nitrocellulose does not absorb in this region and the absorption must therefore be due to the titanium dioxide. The alkyd and epoxy media absorb in the ultra-violet region, but the absorption of the titanium dioxide predominates. Improved reflectivity can only be achieved by a reduction of the absorption in the ultra-violet region; this would suggest the use of anatase titanium dioxide which has less absorption than rutile in that region. However, anatase has a lower refractive index than rutile so that its reflectivity in the visible and infra-red region is inferior and when considered against a solar source the gain in the ultra-violet region is just offset by the loss in the other region, so there is an equal choice in this respect. Chalk resistance considerations lead to a choice of rutile, but the inclusion of some anatase to promote a self-cleaning surface is not detrimental to the reflective efficiency.

MECHANISM OF REFLECTANCE BY PIGMENTS

General

When light passes through a medium containing pigment particles of different refractive index, then a portion of the light is reflected at each interface according to the classical formula

$$R = \left(\frac{n_1 - n_2}{n_1 + n_2} \right)^2$$

where R is the proportion reflected, and n_1 and n_2 are the refractive indexes of the pigment and medium respectively.

In a pigmented paint the reflected and transmitted light are each further reflected, transmitted, and possibly refracted until they escape from the surface of the paint. If no absorption occurs, a white pigmented paint of infinite thickness should therefore have an expected reflectivity of 100 per cent, though this is only approached in practice at the optimum wavelength by high refractive index pigments, such as rutile titanium dioxide, where the number of reflection/transmission processes for any specified efficiency is kept to a minimum. At wavelengths away from the optimum, reflectivity being less, integrated efficiency against any source must fall well below this ideal value. When absorption occurs in the pigment or medium then there is a significant loss in reflectivity. In practice, however, reflectivity can be increased by the use of a pigment, the size of which gives maximum scattering, further reducing the number of reflective processes. Fortunately, pigments which are made to give high visual scattering produce maximum reflectivity at a wavelength which corresponds closely to the maximum energy of the solar spectrum. In such

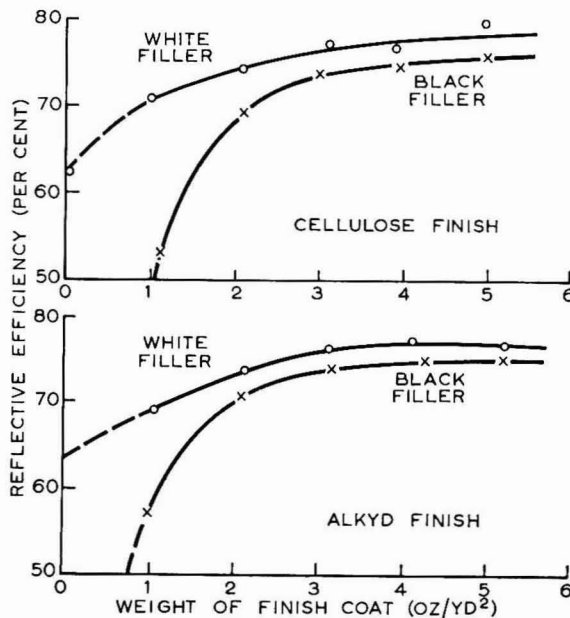
cases, however, the pigment is less efficient at other wavelengths, particularly towards the infra-red.

It is not practical to apply a strict mathematical treatment to the amount of light which is reflected from a white pigmented paint for a great number of reasons, of which the foremost are:

- (a) variable shape and orientation of pigment particles,
- (b) pigment size or size distribution,
- (c) change of refractive index of pigment with change of wavelength,
- (d) distance between pigment particles (*i.e.* pigment/vehicle concentration), if this is of the order of less than half the wavelength of radiation, when the next particle so placed will not reflect, and
- (e) absorption of pigment or medium, particularly, if this is slight and difficult to isolate from other effects, or selective.

Model of Reflectance Process

If, however, a paint has a constant ratio of absorbed to incident radiation over a definite waveband, a simple model may be devised relating the thickness of the paint to its reflectivity. Consider the paint film as a number of equal thin layers or units parallel to the face of the film. Each unit will reflect, absorb and transmit some of the radiant energy it receives. The transmitted energy from one unit becomes the incident energy for the following unit, whilst the reflected energy will travel in the opposite direction, and will again be subjected to multiple reflection. The reflection at each unit is considered to pass out of the paint film affected only by absorption and loss by internal reflection. The internal reflection which is of the order of a few per cent of the reflected ray is neglected.



REFLECTIVE EFFICIENCY OF RUTILE TITANIUM DIOXIDE PIGMENTED NITROCELLULOSE FINISH (FIG. 13) AND ALKYD FINISH (FIG. 14) ON DIFFERENT COLOURED FILLERS

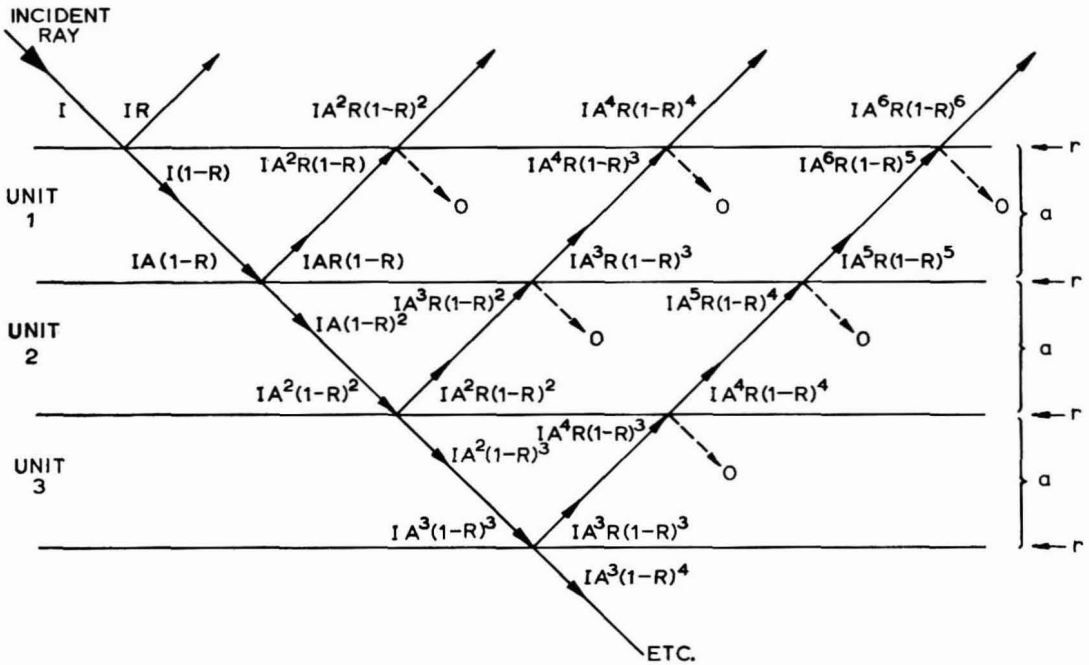


FIG. 15. MODEL OF REFLECTIVE PROCESS
(r—reflector, a—absorber)

Let the incident radiation be I and the reflectivity for each layer be R . If the absorption is such that a fraction A is transmitted by each layer then the effect of each layer based on this simple model is illustrated in Fig. 15.

The total emergent radiation from the paint film (R_t) for n layers, if the substrate is totally absorbing, is therefore given by

$$R_t = IR + IRA^2(1-R)^2 + IRA^4(1-R)^4 + IRA^6(1-R)^6 + \dots + IR \left\{ A^2(1-R)^2 \right\}^{n-1}.$$

It is convenient to replace $A^2(1-R)^2$ by a parameter y so that

$$R_t = IR(1+y+y^2+\dots+y^{n-1}) = IR \frac{1-y^n}{1-y}$$

and the efficiency E is given by

$$E = \frac{R_t}{I} = R \frac{1-y^n}{1-y} \dots \dots \dots (1)$$

However, if the substrate is a filler coat of reflectivity e , its contribution to the total reflection will be $IeA^{2n}(1-R)^{2n}$, and the total efficiency will be

$$E = R \frac{1-y^n}{1-y} + ey^n = R \frac{1 - [A^2(1-R)^2]^n}{1 - A^2(1-R)^2} + e [A^2(1-R)^2]^n (2)$$

and since A and R are each less than 1, y is less than 1. With infinite thickness, when y^n approaches zero, the maximum efficiency is therefore given by

$$E_{n \rightarrow \infty} = \frac{R}{1-y} = \frac{R}{1 - A^2(1-R)^2} \dots \dots \dots (3)$$

If this model is applied strictly to a narrow waveband, values of A and R and the corresponding E will apply which are peculiar to that wavelength. If all the values of E were known over the solar spectrum, a sum of these in proportion to the energy at these wavelengths would give the overall efficiency. It is therefore legitimate to equate the measured integrated solar efficiency to this model formula and obtain an overall value for A and R . These are not strict absorption or reflective coefficients, but are integrated to the energy of the spectrum which is reflected, and the variable absorption and reflection over that spectrum.

Experimental Check of Reflectance Process

This model formula has been applied to the epoxy/amine adduct system (Scheme C), the results of which have been summarised in Table V. If in Equation 1, which applies to a completely absorbing substrate, values of the reflective efficiency are known for top coats, the ratio of thicknesses of which

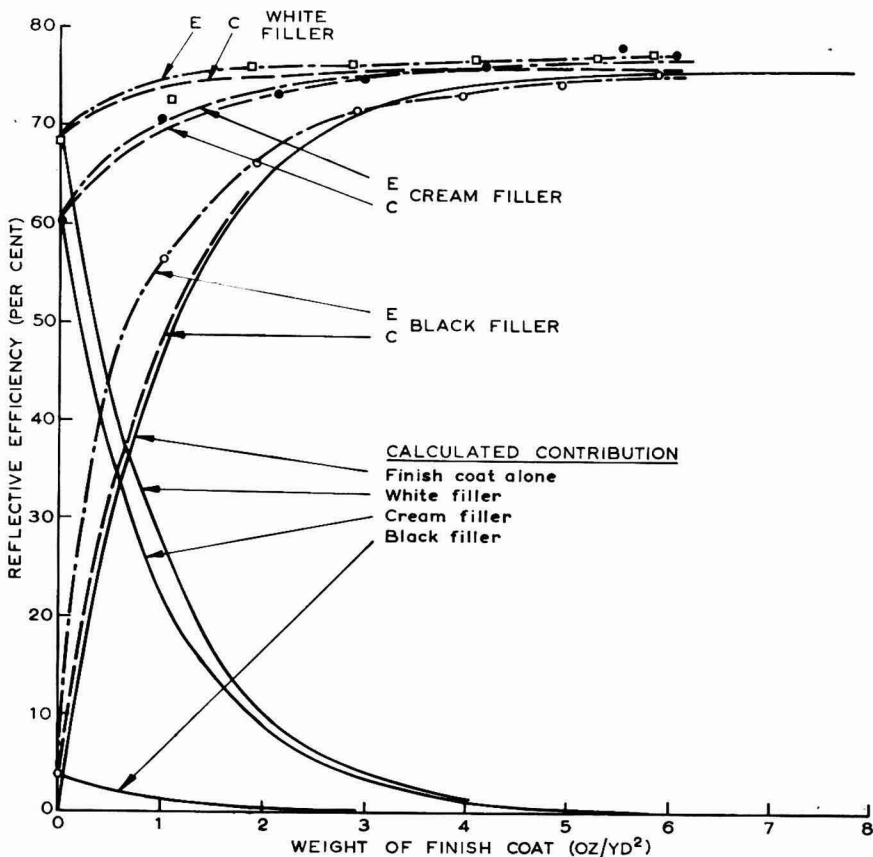


FIG. 16. THEORETICAL AND EXPERIMENTAL VALUES OF REFLECTIVE EFFICIENCY FOR RUTILE TITANIUM DIOXIDE PIGMENTED EPOXY/AMINE ADDUCT FINISH ON DIFFERENT COLOURED FILLERS (E—experimental, C—calculated)

is 2 : 1, then by choosing units of thickness such that $n=1$ and 2 and applying these to Equation 1,

$$E_2 = R \frac{1 - y^2}{1 - y} \text{ and } E_1 = R \frac{1 - y}{1 - y} = R$$

and therefore

$$\frac{E_2}{E_1} = \frac{1 - y^2}{1 - y} = 1 + y = 1 + A^2(1 - R)^2$$

The values of A and R can be obtained and applied to other values of n .

However, in the practical case chosen the substrate has some reflectivity at low film weights, and its contribution to the overall reflectivity can therefore be calculated from its known reflective efficiency (e) and the calculated values of A and R from the term ey^n in Equation 2. If allowance is made for the substrate reflection in the original values of reflectivity, corrected values of A and R can be obtained. Corrected values of A and R have been used to calculate the reflective efficiencies in Table V. These are shown, together with the experimental values, in Fig. 16 as the relation between reflective efficiency and thickness of coating.

Reasonable agreement exists between the experimental and calculated results for acceptance of the reflective model proposed. In all cases, however, the experimental values are slightly greater than the theoretical values which may be for a variety of causes. The model chosen requires at each interface the transmitted ray, which has had its energy reduced by only a few per cent, to travel away from the surface, and to suffer similar small reductions of intensity. If, however, in the practical case with pigments of irregular shape, some rays are refracted towards the surface, particularly in the upper layers, its intensity will be greater than the value permitted by the model for the same number of reflective operations. This premature emergence of the ray could easily account for the difference in the practical and theoretical values and would suggest that by suitable choice the pigment shape and orientation near the surface could materially improve the reflectivity.

In these calculations n has been expressed, for convenience, in arbitrary values proportional to the thickness. In the case of titanium dioxide in a medium of refractive index about 1.5, R must have a value of about 0.07. If R is made equal to 0.07 in the formula, A can be calculated and when applied to the practical values for a top coat of 5.89 oz./yd.², n has a value of 59. This value must be considered to be only approximate, for the model has been shown to be not strictly true. The value chosen for R in this case is not accurate, because the corresponding value of A is slightly greater than 1, instead of equal to or less than 1 by definition.

In Equation 1 when $R \rightarrow 0$ (when the refractive index of the pigment and medium approach one another), $E \rightarrow 0$, irrespective of the value of A , *i.e.* nothing is reflected because the ray entering the film is either transmitted or absorbed. If R has a small but finite value, which is typical of white paints, and there is no absorption ($A=1$), E will be high. If A falls slightly below 1, *i.e.* there is slight absorption, and the term $[A^2(1-R)^2]^n$ has a high value of n (which it must have if maximum reflectivity is to be achieved), its value will approach zero and E will be comparatively small; in the limiting case when $A=0$, E will equal R . The model, therefore, confirms what is found

TABLE V

REFLECTIVE EFFICIENCY OF RUTILE TITANIUM DIOXIDE IN EPOXY/AMINE ADDUCT MEDIUM (Scheme C)

EXPERIMENTAL AND CALCULATED RESULTS
(Nominal weight of etch primer 0.5 oz./yd.²)

Filler		White epoxy/ amine adduct finish (oz./yd. ²)	Reflective efficiency (%)			
Colour	(oz./yd. ²)		Experi- mental	Calculated		
				Filler	Finish	Total
Black	2.43	—	3.8	(3.8)	(0)	—
„	1.93	1.02	56.5	1.4	47.0	48.4
„	2.35	1.91	66.1 ₅	0.7	63.0	63.7
„	2.26	2.91	71.4*	0.2	71.2	71.4
„	2.10	3.94	73.2	0.08	75.0	75.1
„	2.12	4.93	74.5	0.03	75.2	75.2
„	2.38	5.89	75.6 ₅ *	0.01	75.6	75.6
Cream	2.14	—	60.2	(60.2)	(0)	—
„	2.09	0.99	70.6	23.0	46.3	69.3
„	2.14	2.12	73.2	8.0	65.2	73.2
„	2.11	2.98	74.9	3.5	71.7	75.2
„	2.06	4.17	76.1	1.0	74.7	75.7
„	2.68	5.52	77.2	0.2	75.5	75.7
„	2.54	6.05	76.6	0.1	75.6	75.7
White	2.42	—	68.3	(68.3)	(0)	—
„	1.87	1.05	72.6	26.4	47.8	74.2
„	1.49	1.85	76.0	11.6	62.9	74.5
„	2.11	2.86	76.2	4.8	71.1	75.9
„	2.16	4.05	76.9	1.4	74.4	75.8
„	2.28	5.26	76.9	0.5	75.5	76.0
„	2.53	5.80	77.6	0.3	75.6	75.9
„	0	∞	—	(0)	76.0	76.0

*Results on which values of A and R were calculated.

in practice that with multiple reflections any absorption in the pigment or medium produces a significant loss in reflectivity.

HIGH EFFICIENCY SURFACES

Theory

The solar efficiency of titanium dioxide paints could be improved by increasing the difference between the refractive index of pigment and medium or by reducing the absorption in the ultra-violet region. The former can be achieved only by reducing the refractive index of the medium, but the gain is not very significant. If corrosion protection is not essential, a titanium dioxide "white-wash" can give increased reflectivity (82.6 per cent) at the expense of losing it easily by soiling.

The absorption in the ultra-violet region is basic to titanium dioxide and significant in both the rutile and anatase forms. However, with titanium dioxide of good optical scattering power, which is ideal for solar radiation, it has been previously argued, from the increase in difference between reflectivity on a white or black filler with increase in wavelength, that increase in wavelength leads to a greater penetration of energy. It can therefore be concluded that in the visible region, where the reflectivity is high, a large proportion of the reflected energy comes from near the surface. This being so it is evident that, provided there is no absorption, high reflectivity in the ultra-violet region must be confined to a still smaller layer of the surface. Improvement therefore, could, theoretically be made to a titanium dioxide pigmented paint (which has high efficiency at the peak of the solar spectrum) by replacing a portion of the surface with an efficient ultra-violet reflective coating; the radiation of longer wavelength, being less efficiently reflected by this coating, will thus penetrate, and be reflected by, the efficient titanium dioxide paint.

Experimental

If a special ultra-violet reflective top coat is a practical proposition then Figs. 7 and 8 would suggest that anatase titanium dioxide or basic white lead

TABLE VI
EFFECT OF NON-ABSORBING TOP COATS ON RUTILE PIGMENTED EPOXY/AMINE ADDUCT MEDIUM

Top coat	Undercoat	Reflective efficiency (%)
Anatase in acrylic medium	None	72.5
Basic white lead in acrylic medium	None	80.6
None	Rutile in epoxy/amine adduct (5.4 oz./yd. ²)	77.1
Basic white lead in nitrocellulose 1.0 oz./yd. ² ..	„	81.2
Basic white lead in nitrocellulose 2.2 oz./yd. ² ..	„	82.1
Basic white lead in nitrocellulose 4.3 oz./yd. ² ..	„	82.6
Rutile pigment alone	None	82.6

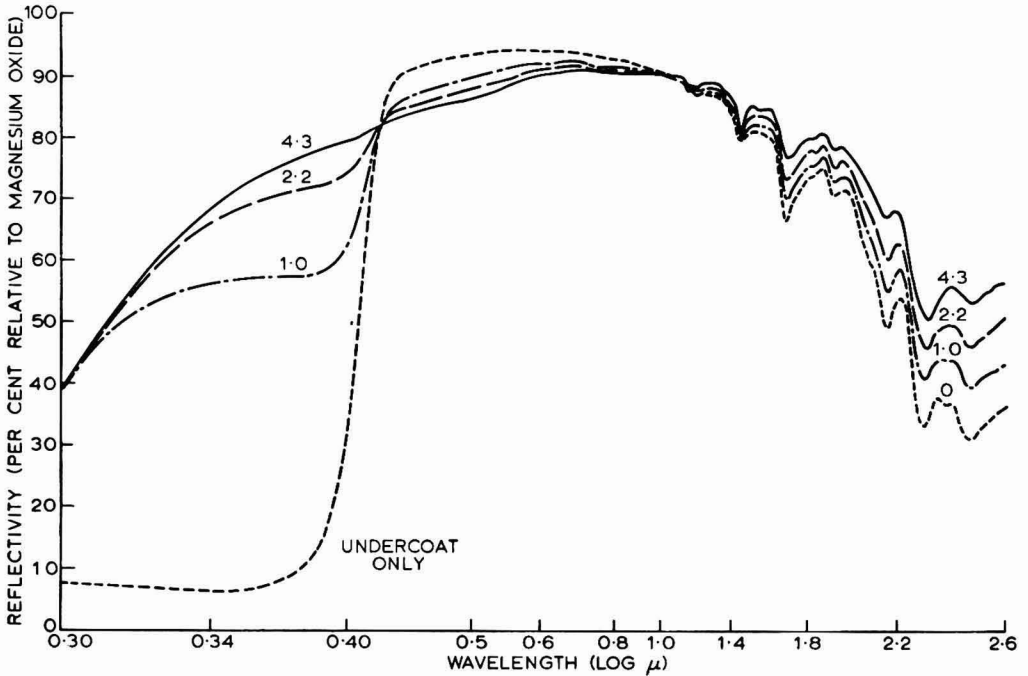
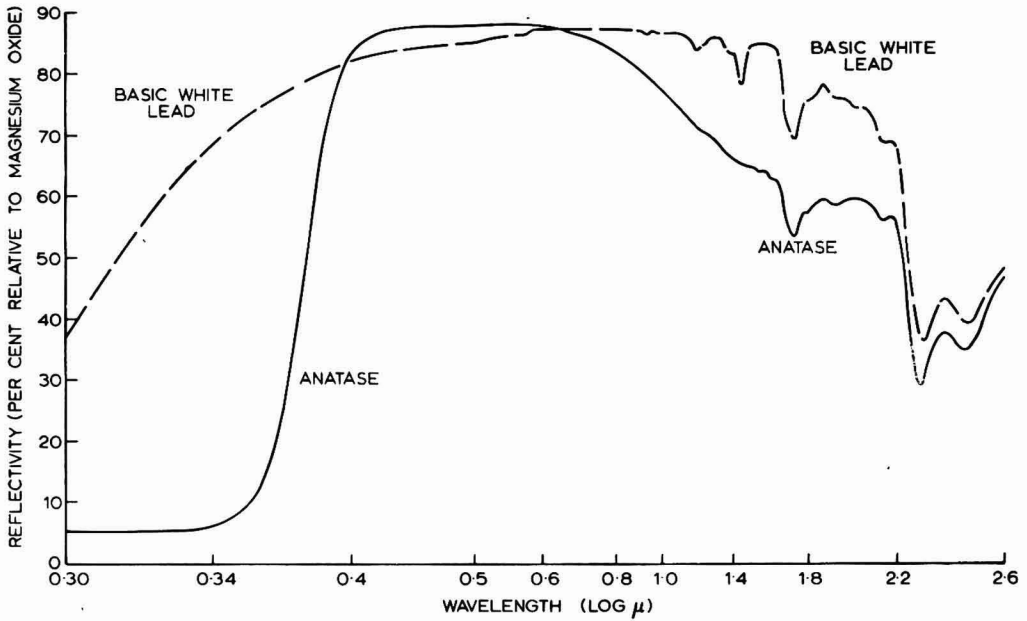


FIG. 17. (*upper*) REFLECTIVITY OF ANATASE TITANIUM DIOXIDE AND BASIC WHITE LEAD IN AN ACRYLIC MEDIUM

FIG. 18. (*lower*) REFLECTIVITY OF BASIC WHITE LEAD PIGMENTED NITROCELLULOSE TOP COATS OF DIFFERENT THICKNESS ON A RUTILE TITANIUM DIOXIDE PIGMENTED NITROCELLULOSE UNDERCOAT

(The figures on the curves indicate the weight of top coat in oz./yd.²)

might be worth considering. The reflectivity of these pigments in a non-absorbing acrylic medium is given in Fig. 17. The superiority of basic white lead is clearly demonstrated and, with a solar reflectivity of 80.6 per cent in its own right, it is superior to rutile (or anatase) titanium dioxide in a non-absorbing medium. The effect on reflectivity of applying coatings of different thickness of basic white lead in a cellulose medium on a rutile pigmented epoxy/amine adduct is given in Fig. 18. Details of these coatings and their solar reflective efficiency are given in Table VI.

The special effect of a non-absorbing top coat on rutile has been established for, with 1.0 oz./yd.² on top of a maximum efficiency rutile pigmented epoxy/amine adduct, the net effect is greater than the separate components.

CONCLUSIONS

Considered from the point of view of thermal efficiency, the best practical single pigment to use against solar radiation is rutile titanium dioxide with a particle size equivalent to that used for maximum visual reflectance. Rutile titanium dioxide can be used in any normal medium provided that it does not absorb in the region 0.4 to 2.2 μ . Between 0.3 and 0.4 μ rutile absorbs strongly and masks the absorption which occurs with an alkyd in this region. The process of reflectivity can be explained quantitatively by a simple model. Using typical pigment size associated with maximum visual scattering it has been shown that the ultra-violet radiation is mostly reflected on the surface of the paint, while the longer wavelengths progressively penetrate the paint film and reach the substrate. The ultra-violet absorption of rutile can be minimised by overcoating with a non-absorbing pigment, such as basic white lead, in a non-absorbing medium. A solar reflective efficiency of 77.0 per cent can be obtained with rutile alone and 82.6 per cent with a special non-absorptive top coat.

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[Received 25 January 1961]

DISCUSSION

MR. T. R. BULLETT congratulated Dr. Strain on a first-class, and very useful, piece of practical work. However, he had some reservations concerning the theoretical sections of the paper. With regard to the rise in temperature of surfaces exposed to solar radiation, he did not agree with the author that in the general case of a body heated by solar radiation re-radiation could be neglected as a controlling factor in the temperature rise. He recalled that in earlier work at the Paint Research Station, a simple assumption had been made, following the heat transmission data given by Fishenden and Saunders, that heat losses for a black body by convection and radiation transfer were equal. This had led to a simple heat balance equation of the following form:

$$\text{Heat intake} \propto \text{solar absorption coefficient } (A_s),$$

$$\text{where } A_s = (1 - \text{reflective efficiency})$$

$$\text{Heat loss} \propto [1 + \text{emissivity } (E_t)] \times \text{excess temperature.}$$

$$\text{Therefore the heat rise} \propto \frac{A_s}{1 + E_t}$$

The emissivity at the temperature of the surface, E_t , was about 0.88-0.95 for all non-metallic paints at normal air temperatures, but only about 0.5 for aluminium paints, and less than 0.1 for polished metals. Practical measurements of the heating of painted surfaces under solar radiation confirmed the validity of the above equation. As predicted, it was found that aluminium painted surfaces heated considerably more than non-metallic paints of the same reflective efficiency.

Concerning the role of pigments in controlling reflective efficiency, Mr. Bullett said that the controlling factor was not simply the scattering power of the pigment particle, but the ratio of this property to the absorption coefficient of the pigmented layer. Thus a low refractive index pigment with no absorption bands in the appropriate wavelength range, such as blanc fixe, if used in a non-absorbing medium, could give a very high reflective efficiency. He asked if the author had examined the possibilities of such a pigment for the ultra-violet reflecting top coat mentioned in the paper, and also whether he had considered antimony oxide for this purpose, because it was a good ultra-violet reflector and would be less subject than white lead to staining in polluted atmospheres.

Mr. Bullett considered that the model developed to explain the reflection properties of multilayers was defective in several respects. One important fact that had been overlooked was that the internal reflection at the air/paint interface was by no means negligible, being over 50 per cent and not 5 per cent as for the reflection at the external surface, the difference being due to total internal reflection. The whole treatment appeared to be unsound and the questioner wondered whether the author would have done better to base his theoretical calculations on the two constant theory of Kubelka and Munk.

DR. STRAIN, referring to re-radiation, replied that the example which Mr. Bullett had taken was quite correct, but he himself was not thinking of a modest temperature rise, something around the ambient. However, if the rise were 150°C, which was very high, there would be considerable convection. With regard to reflective power in relation to absorption, he agreed that blanc fixe had a low refractive index and therefore it achieved reflective efficiency by a large number of operations. Reflective power could be obtained provided that there was no absorption by the medium. If absorption were significant a higher refractive index and a minimum number of operations would be required.

In reply to the comment questioning the validity of the model, Dr. Strain felt that the internal reflection was fairly small, because it was the second reflective ray.

MR. P. FINK-JENSEN, however, thought that the internal reflection of about 50 per cent mentioned by Mr. Bullett was correct and that it certainly could not be neglected without further consideration. The top surface of the film was the vehicle, with a refractive index of about 1.5, while the air outside had a refractive index of 1.0. Light penetrating the top surface from outside would be diffused by pigment particles and on arrival at the film-air interface a large proportion (say 50 per cent) would be totally reflected back into the film. Accordingly, the distances which the light had to travel would differ markedly from those visualised in the paper.

In a written contribution, Mr. Fink-Jensen added that in the theory given in the paper secondary reflections were ignored entirely, which was not permissible, as shown by Equation (3). For $A=1$ (no absorption) and small but finite values of R , the maximum efficiency became 0.5, but in fact all the light would be reflected under these conditions, so that $E=1$.

MR. T. R. BULLETT thought that Dr. Strain had assumed that the pigment was concentrated at the surface, whereas the usual view of the optics of a paint film was that there was an interface between the air and the paint medium, and that the pigment particles could be considered as being in the paint at a lower level.

DR. STRAIN said that if it was considered that the light was scattered, it looked as though the secondary reflections did not contribute in any way. It was true that the light was refracted and that a certain amount came out. However, there was only a difference of 1 per cent between the calculated and the experimental results.

DR. J. A. W. VAN LAAR said there was a distinct ray and specular reflection only on the outside of the film, and not at the interface, as Fig. 15 suggested. The possibility of total reflection from the top interface (Fig. 15) from the bottom (as mentioned by Mr. Fink-Jensen) was not important, for it was probable statistically that the diffuse rays within the angle of total reflection after this reflection and re-scattering would to a great extent approach the interface at an angle favourable to escaping the film.

DR. STRAIN replied that the model was descriptive of one ray only. The reflectivity in this case was the total integrated reflectivity, being a summation of all the rays into one.

Some Aspects of the Physical Assessment of Emulsion Polymer Films

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Summary

Some methods used to investigate the properties of externally and internally plasticised p.v.a. emulsions with regard to their use as surface coating media are described.

The two major methods employed are the determination of the tensile strength, elongation at break and modulus of elasticity of films, and the determination of the adhesion of emulsion paint films to gloss alkyd painted surfaces.

The object of the work carried out was to examine methods of obtaining the best performance from a p.v.a. emulsion paint film, in particular with regard to the amounts of external or internal plasticisers used and their influence on the resistance to scrubbing and the dirt retention of the paint film. Loss of plasticiser, particularly at elevated temperatures, has also been studied and shown not to be a drawback to the use of homopolymers. In fact, this property could usefully be employed in obtaining a tough film with good scrub resistance and low dirt retention from both homo and copolymers.

INTRODUCTION

The scrub resistance and dirt retention of an emulsion paint film are considered to be complementary and closely related to certain physical characteristics of the emulsion polymer used in the paint. A closer study of the physical properties of polymer films deposited by emulsions should therefore reveal ways and means of improving the performance of paints made from them. Too often the choice of an emulsion from which a paint is made has been arbitrary, particularly with regard to the amount and type of internal or external plasticisers incorporated with the polymer. One important property which an emulsion paint must possess is the ability to form a well integrated film at the lowest likely temperature of application. The film must be tough, resistant to cleaning at ambient temperatures and must collect as little dirt as possible. Consideration of the tensile strength, elongation at break and modulus of elasticity of the polymer films can throw much light on this subject.

It should be borne in mind that characteristics such as molecular weight, chain branching and the particle size distribution of any one chemical type of polymer emulsion can greatly affect its minimum film forming temperature and other physical properties. The work described in this paper on homopolymers of vinyl acetate and copolymers of vinyl acetate with 2-ethyl hexyl acrylate should, therefore, serve only as a guide to the properties expected from related materials.

A method of assessing the adhesion of emulsion paint films to a substrate is described, with particular reference to paints made from plasticised vinyl acetate homopolymers and a copolymer of vinyl acetate with 2-ethyl hexyl acrylate. This method was adapted from that described by Phillips¹. Perhaps one of the outstanding features of this method is the speed at which a paint film subjected to widely varying weathering conditions can be tested; in this work films of high moisture content were examined before they had dried out.

EXPERIMENTAL

Tensile Strength Measurement

One of the great practical difficulties in examining the physical characteristics of surface coating materials is that of preparing films of sufficiently even thickness, free from brush marks and other surface imperfections. This difficulty is even more pronounced in the case of dispersions than of solution polymers. This is because of the poorer flow exhibited by dispersions during the evaporation of the continuous phase and the coalescing of the dispersion particles to form a continuous film.

A number of ways of obtaining even thickness films of p.v.a. emulsions were explored, including the use of film applicators and spraying techniques. The former method often gave films with surface defects so that the latter method was preferred, although care had to be exercised in spraying some emulsions which tended to clog the spray-gun. The emulsions were sprayed on to specially prepared $\frac{1}{4}$ in. thick high density polyethylene sheets to give films 0.005-0.007 in. thick after drying at 25°C and 66 per cent R. H. for three days. The films were carefully removed from the sheets and dumb-bell shaped specimens 2.25 in. long by 0.187 in. wide at the narrow section were stamped out with the aid of a cutter. The thickness of each specimen was measured with a dial micrometer. During testing the specimens were often found to break at the jaws of the Tensometer and it was found expedient to tape the wide ends of each specimen with *Sellotape* on both sides as shown in Fig. 1. It was found much easier to handle the test pieces joined together in this way rather than separately, and the tapes could be cut as each piece was ready to be tested.

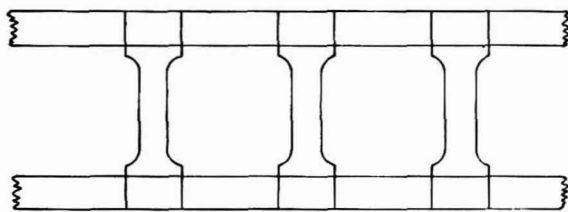


FIG. 1. TENSILE STRENGTH SPECIMENS TAPED WITH *Sellotape*

The tensile strength of each specimen was measured on an *Instron* Servo-Controlled Tensile Strength Tester, using an initial jaw separation of 1.5 in. and a rate of pulling of 2 in. per minute. This rate was found the most convenient for emulsion films of the type under investigation. The Tensometer was equipped with a variety of load cells and a recording chart, the speed of which could be varied within wide limits. One particular load cell and one chart speed were selected throughout the work described.

Films of the following homopolymer and copolymer emulsions were examined, the tensile measurements being determined at 23°C.

- (i) A plasticised vinyl acetate (VA) homopolymer containing 5, 10, 15 and 20 per cent of dibutyl phthalate (DBP), butyl *Carbitol* acetate (BCA) and *Carbitol* acetate (CA).
- (ii) VA copolymerised with 5, 7.5, 10, 12.5, 15, 17.5, 20, 22.5 and 25 per cent of 2-ethyl hexyl acrylate (2-EHA). These copolymers were then plasticised with 0, 5 and 10 per cent of DBP.
- (iii) A VA homopolymer containing 13 per cent DBP and VA, copolymerised with 17.5 per cent 2-EHA, after exposure to relative humidities ranging from 0 to 100 per cent.

Plasticiser Loss

Films of a VA homopolymer plasticised with 13 per cent DBP, and a VA/2-EHA copolymer containing 17.5 per cent 2-EHA copolymerised with the VA were heated in a ventilated oven fitted with a fan at 60°C. The loss in weight of each film was followed during a total heating period of 286 hours. In this case the films had been sprayed on to microscope slides in order to support them during the experiment.

Adhesion Measurements

The heads of $\frac{5}{16}$ in. stainless steel bolts were turned to $\frac{1}{2}$ in. diameter on a lathe and then ground flat at right angles to the stems. The heads were finally polished with a fine metal polish on a perfectly flat glass plate and cleaned in solvent. Black gloss alkyd paint was applied to the flat heads of half the bolts, allowed to dry and then artificially aged (3 weeks at 40°C, 1 hour at 100°C). These surfaces were then scrubbed, washed and allowed to dry. Two VA homopolymers A and B plasticised with 13 per cent DBP and one copolymer of VA with 17.5 per cent 2-EHA were made into paints. The pigment to binder ratio of each paint was 2.45 to 1 by weight, methyl cellulose was the thickener, and the pigment composition used, giving a Pigment Volume Concentration (PVC) of 42.8 per cent, is shown in Table I.

Pigment	Parts by weight
<i>Rutiox CR</i>	11
Paris White	10
<i>Micafine P</i>	1.1

The alkyd coated bolt heads were sprayed with the emulsion paints and allowed to dry for three days at 50 per cent R.H. at room temperature. They were then exposed for three days in desiccators at relative humidities ranging from 0 to 95 per cent before the painted surfaces were tested. At least five bolts were prepared for each experiment involving a change in the paint or exposure conditions.

Adhesion of the emulsion paint to the gloss paint was determined by sticking another bolt with a polished head to the painted bolt by means of an adhesive,

Eastman Kodak 910. This adhesive has the unusual property of forming a good bond between many different surfaces within a few minutes. Full bond strength, of course, took somewhat longer to develop, but in most cases where the adhesion of the emulsion paint to the gloss paint was not of a high order the bond formed by the adhesive was sufficiently strong within two minutes for the test to continue. The duplet formed by the two bolts was placed in special jaws in a *Hounsfield* Tensometer and, using a $\frac{1}{2}$ ton beam and a rate of jaw separation of 2 in. per minute, the force necessary to separate the bolts was measured.

Water Absorption

Ten paints were prepared using homopolymer A as the binder and titanium dioxide, *Rutiox CR*, as the sole pigment, varying the pigment volume concentration of the paint from 0 to 63.5 per cent. The paints were sprayed on to weighed microscope slides 3×1 in. and allowed to dry overnight. They were placed in a desiccator over phosphorus pentoxide until constant weight was achieved and then in a desiccator over water at 25°C , again until constant

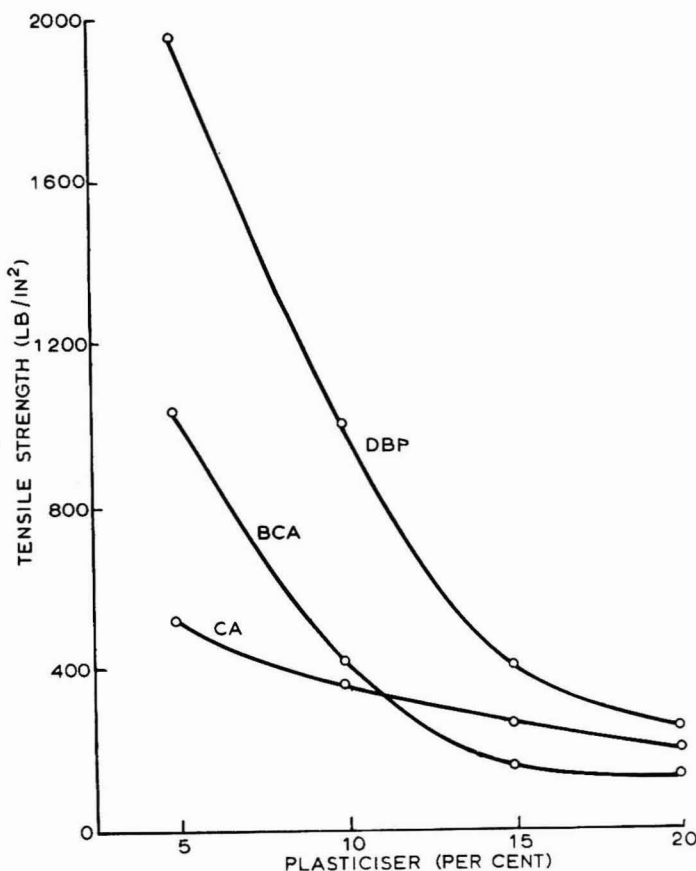


FIG. 2. TENSILE STRENGTH AT 23°C OF VA HOMOPOLYMER PLASTICISED WITH DBP, BCA AND CA

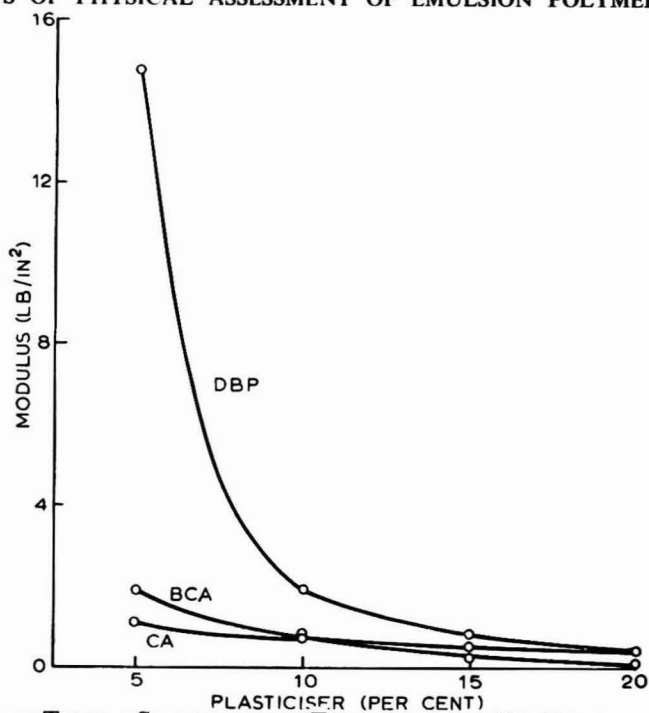


FIG. 3. MODULUS OF TENSILE STRENGTH AND ELONGATION OF VA HOMOPOLYMER PLASTICISED WITH DBP, BCA AND CA

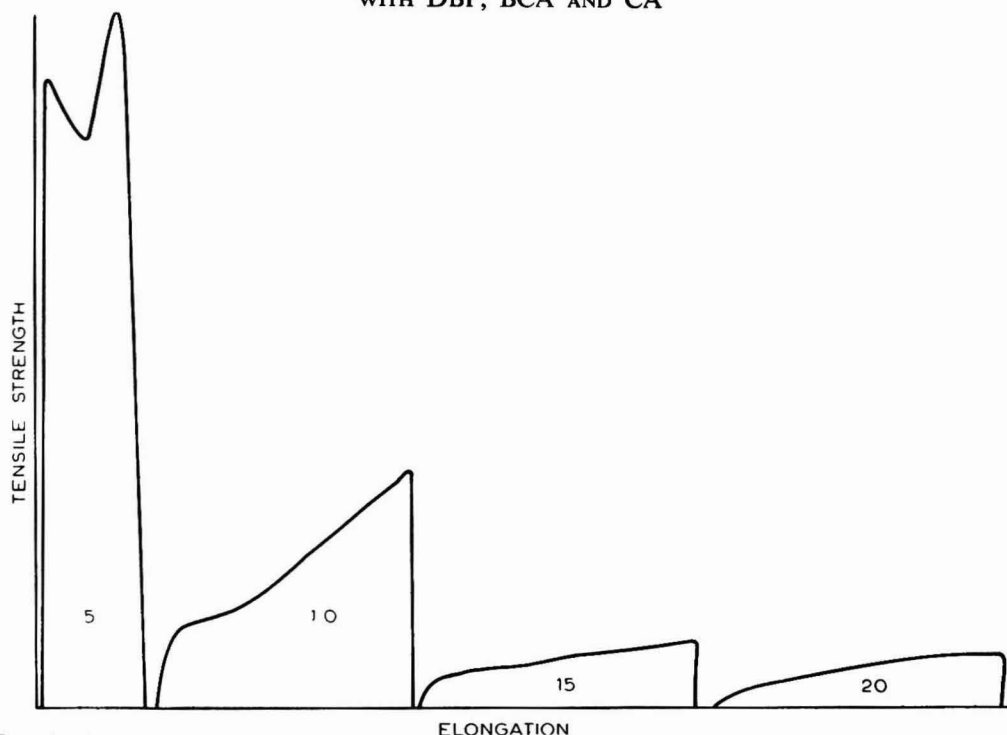


FIG. 4. Instron PLOT OF TENSILE STRENGTH AND ELONGATION OF VA HOMOPOLYMER AT 23°C (Figures indicate percentage of DBP)

weight had been achieved. The weight of water absorbed was recorded and related to the weight of paint film in each case.

RESULTS

Tensile Strength and Modulus of Elasticity—VA Homopolymer

The results obtained at 23°C for the tensile strength and modulus of elasticity of films of a VA homopolymer are shown in Figs. 2 and 3. The polymer was plasticised with three separate plasticisers, DBP, BCA and CA, in amounts varying from 5 to 20 per cent by weight of the plasticised films. The effect of a plasticiser can, of course, be judged from the tensile strength and modulus plots. A small amount of CA, for example, reduces the modulus figure more quickly than does the same amount of DBP, and thus it is considered to be more efficient in action but not necessarily more permanent or suitable.

The figures show that CA and BCA are far more efficient plasticisers than DBP, particularly at low levels. At high levels of plasticiser content, of the order of 15 per cent and upwards, the tensile strength and modulus values decrease for each type of plasticised film and approach parity. How the actual plot of tensile strength and elongation to break appeared on the chart of the *Instron* Tensometer for films plasticised with 5, 10, 15 and 20 per cent of DBP is shown in Fig. 4. Elongation, represented on the abscissa, and tensile strength, represented on the ordinate, can be compared approximately for each film since the rate of jaw separation of the instrument and the speed of rotation

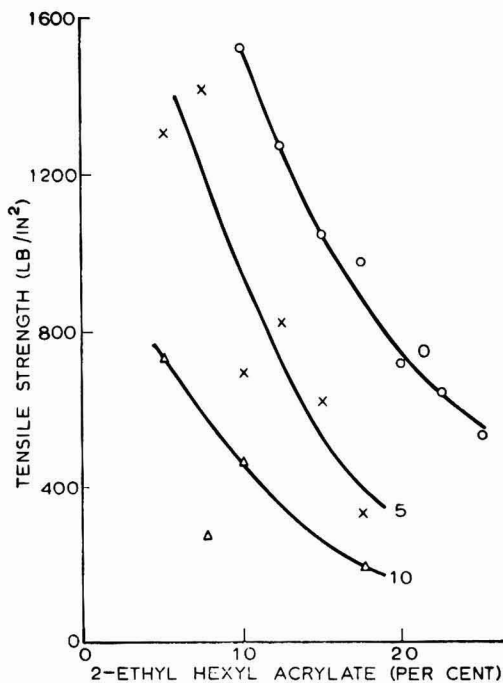


FIG. 5. TENSILE STRENGTH AT 23°C OF VA/2-EHA COPOLYMER WITH ADDED DBP (Figures on curves indicate percentage addition)

of the chart were the same in each case. However, the films varied in thickness and this was taken into account in obtaining Figs. 2 and 3. There is a peculiar first peak exhibited by the tensile strength plot of the film containing 5 per cent DBP which is at present unexplained. It seems to have occurred also to a small extent in the case of the film containing 10 per cent DBP.

Copolymers

The tensile strength and modulus at 23°C, wherever possible, of films of nine copolymers of VA with 2-EHA, varying in composition from 5 to 25 per cent of 2-EHA in the copolymer, are shown in Figs. 5 and 6. Determinations were also carried out on the same copolymers to which external plasticiser (DBP) had been added. Film formation of the above copolymers without DBP at a few degrees above 0°C did not occur readily until the amount of copolymerised 2-EHA was increased to about 17.5 per cent of the copolymer. At this level the film, when brought to room temperature, was considered to be soft and, moreover, likely to be dirt retentive, particularly at temperatures experienced on hot days.

Additions of 0, 5 and 10 per cent DBP were made to each copolymer in the series and the tensile strengths of their films determined. With 5 per cent of DBP present it was found that film formation just above 0°C could be obtained with a copolymer containing only 10 per cent 2-EHA. Reference to Figs. 5 and 6 shows that the film from such a system gave values for tensile strength and modulus comparable with those obtained for the copolymer containing 17.5 per cent 2-EHA and no DBP.

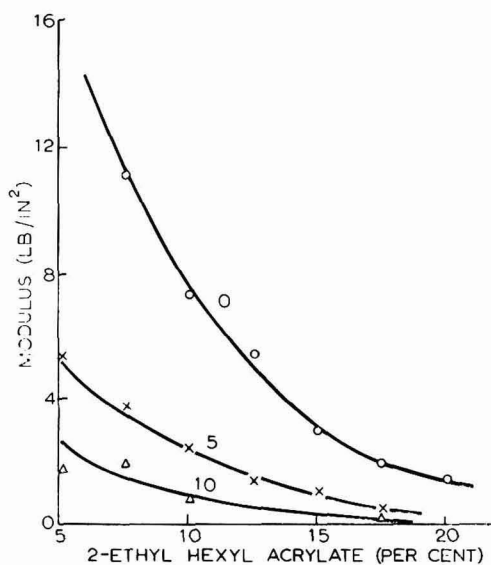


FIG. 6. MODULUS AT 23°C OF VA/2-EH

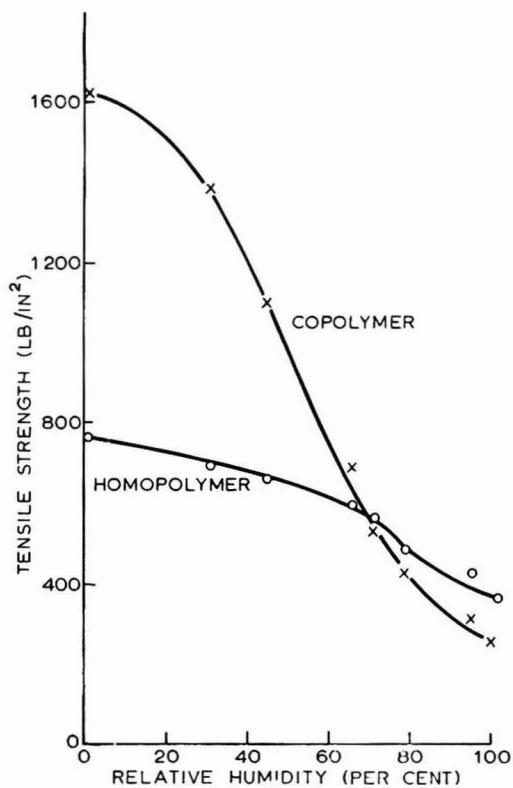


FIG. 7

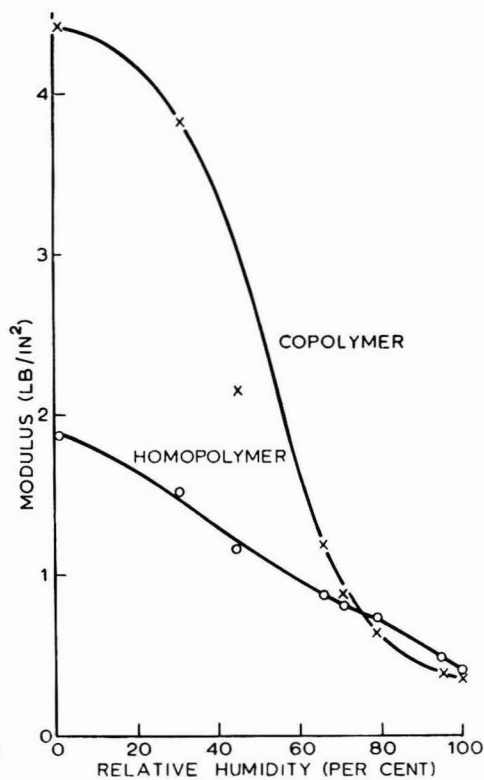


FIG. 8

EFFECT OF HUMIDITY ON TENSILE STRENGTH (FIG. 7) AND MODULUS (FIG. 8) AT 23°C OF VA HOMOPOLYMER AND VA/2-EHA COPOLYMER

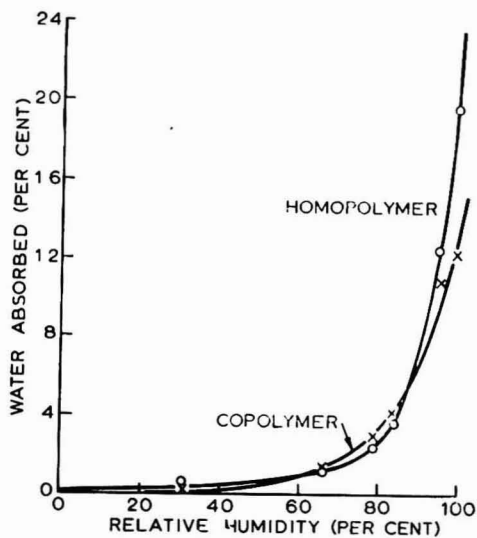


FIG. 9

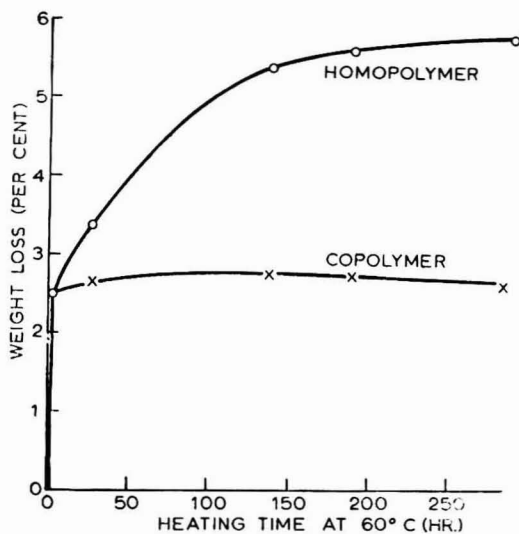


FIG. 10

WATER ABSORPTION AT 25°C (FIG. 9) AND LOSS IN WEIGHT AT 60°C OF VA HOMOPOLYMER AND VA/2-EHA COPOLYMER

Humid Conditions

The change in tensile strength and modulus at 23°C of films of two emulsions, a VA homopolymer plasticised with 13 per cent DBP and a copolymer containing 17.5 per cent of 2-EHA copolymerised with VA, on exposure to humid conditions ranging from 0 to 100 per cent relative humidity at 25°C, is shown in Figs. 7 and 8. The amount of water absorbed by the two films at 25°C, whilst exposed to the same conditions of humidity, is shown in Fig. 9. From these graphs it is apparent that the amount of water absorbed in both cases is small up to approximately 70 per cent R.H. and thereafter rises quite sharply. This coincides with a change in the tensile strength and modulus graphs at the same relative humidity.

Plasticiser Loss

The weight losses at 60°C of films of a homopolymer plasticised with 13 per cent DBP and a copolymer of VA with 17.5 per cent 2-EHA are shown in Fig. 10. Within the first three hours of heating both films lost about 2.5 per cent in weight. The homopolymer continued to lose weight up to an amount of 5.5 to 6.0 per cent within 150 hours and thereafter lost little more. The copolymer, on the other hand, lost virtually no weight at 60°C after the initial loss of 2.5 per cent in three hours.

Adhesion

The results found for the adhesion of the three emulsion paints to the prepared gloss alkyd surfaces, in conditions of relative humidity ranging from 0 to 95 per cent, are given in Figs. 11, 12 and 13. The paint from homopolymer A showed a slightly higher value for adhesion than homopolymer B, over practically the whole range of humidities. In order to obtain each point on the graphs at least five duplets were examined and the mean adhesion value plotted. Examination of the bolts showed that in virtually all cases the bond of the emulsion paint to the gloss alkyd paint had failed after pulling the bolts apart.

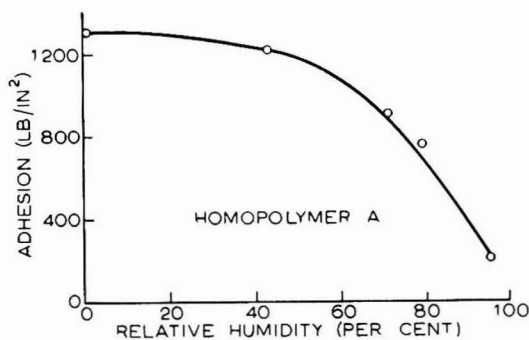


FIG. 11. ADHESION OF PAINT BASED ON HOMOPOLYMER A AT VARIOUS RELATIVE HUMIDITIES

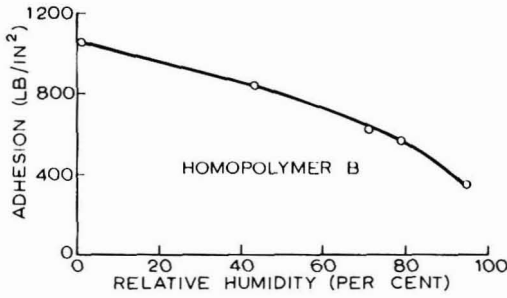


FIG. 12

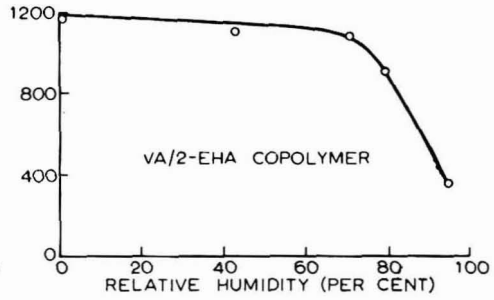


FIG. 13

ADHESION OF PAINT BASED ON HOMOPOLYMER B (FIG. 12) AND VA/2-EHA COPOLYMER (FIG. 13) AT VARIOUS RELATIVE HUMIDITIES

Water Absorption

The water absorption at 25°C and 100 per cent R.H. of paint films made from homopolymer A is given in Fig. 14. In this experiment paints were made with pigment volume concentrations ranging up to 63.5 per cent using *Rutiox CR* as the only pigment. It can be seen that at 50 per cent PVC the water absorption

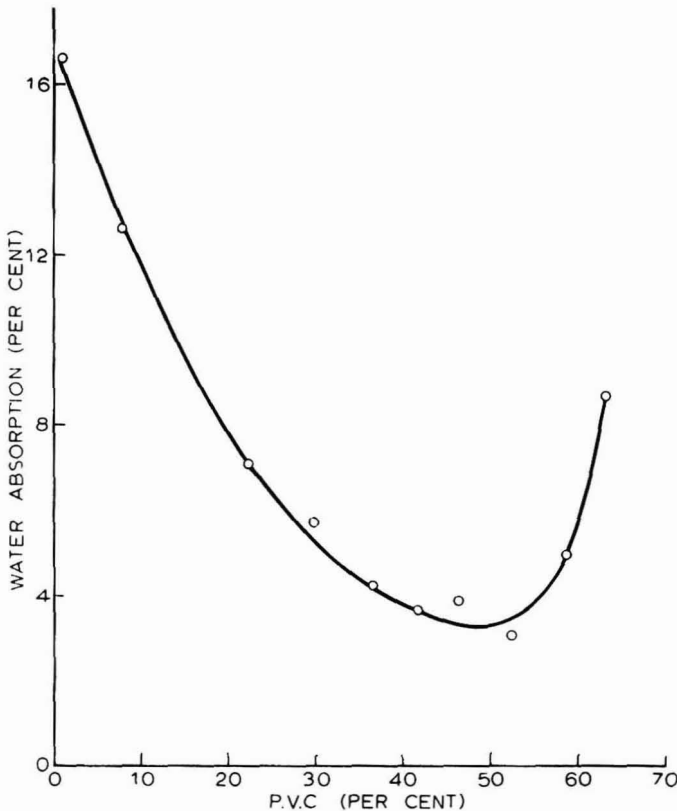


FIG. 14. WATER ABSORPTION AT 25°C AND 100 PER CENT R.H. OF PAINTS BASED ON VA HOMOPOLYMER A

of the paint film is at a minimum. It is safe, therefore, to deduce that the critical pigment volume concentration for this system occurs at about 50 per cent PVC. The paints prepared for the adhesion tests were thus made below their CPVC's. This is important since it has been pointed out by Phillips¹ that the adhesion to a substrate of a paint prepared above the CPVC can fall off quite appreciably.

DISCUSSION

Examination of the films obtained from the VA homopolymer plasticised with DBP showed that a minimum of about 10 per cent of DBP was required for room temperature film formation. Further amounts of DBP lead to a considerable reduction in tensile strength without much increase in the elongation of the film. It should, of course, be emphasised that this result applies only to the particular vinyl acetate homopolymer examined. Other VA homopolymers can be expected to differ if only because of the different molecular weights, particle sizes, soaps and colloids which may be present. Reference to Figs. 2 and 3 shows that, of the three plasticisers examined, CA is the most efficient, followed closely by BCA, and finally by DBP. It is generally accepted that DBP is the most permanent plasticiser of the three; this alone, therefore, is sufficient argument for its selection as the main plasticiser. However, since CA and BCA are more efficient, it is suggested that good use could be made of a mixture of DBP with either CA or BCA in the preparation of a VA homopolymer designed as the pigment binder in emulsion paints. Sufficient plasticiser must be incorporated in the polymer in order that film formation can take place at low temperatures, as well as at 25°C as in the case above.

It is well known that good film integration is a pre-requisite of an emulsion paint, from which properties such as good resistance to scrubbing and low dirt retention are required. The former property is obtained from films of high tensile strength and the latter from films which are not too soft or sticky, even when exposed to elevated temperatures. However, good film integration is best achieved with plenty of plasticiser, whether this is a temporary plasticiser, such as BCA or CA, and thus only a coalescing agent, or a more permanent one. A judicious balance of plasticiser and coalescing agent is therefore advocated for VA homopolymers destined for use in paint, the coalescing agent aiding in film formation at low temperatures and, on volatilising, leaving the film in a tough, non-tacky state.

Protzman and Brown² have pointed out the importance of the minimum filming temperature of polymer emulsions used in coatings. They also consider it undesirable to employ polymers of extremely low minimum filming temperature, for these polymers would exhibit softness and stickiness at temperatures of use. Control of the filming temperature is thus desirable and they have put forward a method for its determination. This method, taken into account with the tensile strength and modulus of elasticity of the film at normal temperatures, should ensure that the best combination of properties for such films is obtained. The above remarks apply equally well to films of copolymers and, whereas in the work to date only DBP has been used with the copolymer, other more transient materials, such as CA or BCA, could

usefully be employed instead of DBP. This would assure a more rapid attainment of a tough non-sticky film having adequate flexibility at temperatures of use.

With regard to plasticiser loss, Fig. 10 clearly shows the difference between a homopolymer externally plasticised and an internally plasticised copolymer. What is equally interesting is the fact that after 300 hours heating at 60°C the rate of loss in weight of the homopolymer had become extremely small. The initial loss in weight, amounting to about 2.5 per cent in each case, after heating at 60°C for the first three hours, could be regarded as being due to the loss of moisture from the films together with the loss of some volatile matter normally present in emulsion films. Subsequent losses in the case of the homopolymer would primarily be due to loss of DBP. The homopolymer initially contained 13 per cent DBP, and therefore in 300 hours at 60°C it first lost 2.5 per cent water and volatile matter, followed by a loss in weight of 3.25 per cent of the remaining film. At this point it was calculated that the film still contained about 10 per cent plasticiser. This suggests that even at 60°C a reasonable quantity of DBP is quite firmly held in the polymer film.

The effect of imbibed water in the paint film is clearly shown to result in loss in adhesion to the substrate. The relative humidity at which this loss becomes appreciable is demonstrated in Figs. 11, 12 and 13. Comparison of paints of the two homopolymers A and B in Figs. 11 and 12 shows that the loss in adhesion becomes appreciable after a relative humidity of 70 per cent has been reached. The percentage loss of initial adhesion of the three paints tested is shown in Table II.

TABLE II
PERCENTAGE LOSS OF ADHESION ON ABSORPTION OF WATER

Relative humidity (%)	Loss of initial adhesion (%)		
	Homopolymer A	Homopolymer B	Copolymer
0	0	0	0
43	5.8	21.0	6.8
71	31.6	41.5	5.0
79	41.6	46.8	18.0
95	85.2	67.0	69.5

The loss in adhesion of the copolymer is not nearly so marked as in the case of the two homopolymers. This was borne out by the results obtained from scrubbing tests carried out on the above three paints on various substrates, including old gloss alkyd surfaces. Scrub resistance was considerably upgraded when about 1 to 2 per cent of a coalescing agent, such as butyl *Carbitol* acetate, was added to the paint before application. The improvement was more marked after the painted surfaces had been aged for seven days than if the test were carried out after only one day's drying.

CONCLUSIONS

The use of plasticisers less permanent than DBP such as *Carbitol* acetate or butyl *Carbitol* acetate is recommended for both homopolymers and copolymers. In the former case they should, of course, be used in conjunction with dibutyl phthalate or a similar plasticiser. Improved scrub resistance and less dirt retention of the paint film can be obtained in this way. A method is described for determining the adhesion of emulsion paint films to substrates after such films have been exposed to humid conditions. By the use of such a method it is hoped that factors affecting the adhesion of emulsion paints under adverse conditions can eventually be assessed more reliably than by present day scrubbing tests.

Measurement of the tensile strength of polymer and paint films is thought to be a most useful way of determining the effect of various changes on the films. Provided good specimens can be made, the test itself can give reasonably accurate results. The method is applicable to a number of other investigations on surface coating materials, e.g. the determination of curing times, influence of pigments and extenders on the paints and the correlation of the effects of weathering trials on films.

ACKNOWLEDGEMENTS

The author wishes to express his thanks to the directors of British Resin Products Ltd. for granting permission to present this paper, to Mr. P. A. M. Abate, Mr. B. Curtis and Mr. B. Parton for the practical work involved, and to Mr. G. Phillips for helpful discussions.

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DISCUSSION

MR. G. W. MACK said that the adhesion figures obtained by the author and other contributors, even under adverse conditions, were often about 50 or 100 lb./in.² Some of the forces causing blistering, such as osmotic pressure, were much higher, but in many cases the operative forces, e.g. the pressure of trapped air, water vapour or solvent, appeared to be too weak to overcome adhesions of this order.

In an attempt to understand the apparent discrepancy several sustained load tests were made at the Building Research Station. They showed that some distempers and emulsion paints on wetted brick might show adhesion failure in a few hours or days, even when the load was as low as 5 lb/in.². They did not obtain a very good correlation between the results of rapid loading and sustained loading. He thought that this might depend on whether a film was plastic or elastic.

Mr. Mack felt that there was need to state the time factor in adhesion testing and to consider the figures obtained in relation to the probable demands on adhesion. He would like to see a balance sheet of the adhesions against in the various types of pressure encountered.

MR. WILLIAMS replied that the first thing to be considered was that the adhesion figures obtained were not absolute figures; they must bear relation to the way in

which the tests were made, and in particular, to the rate of separating the films from the substrate. The figures that he and his colleagues had obtained were possibly a little high. However, it was necessary to have some figures in order to compare one film with another. The general level of the values obtained gave good accuracy and made comparison of similar films easier. By employing a jaw separation rate of 2 in./min. it was possible to measure the adhesion of all materials under test using only one beam, *viz.* a $\frac{1}{2}$ ton beam, on the Housfield Tensiometer. It should also be noted that the method did not involve "peel strength", since a direct pull was used.

DR. H. W. TALEN felt that it might be of some relevance to describe a method used at the Paint Research Institute T.N.O., in Holland, for producing detached films. A piece of tin foil, very smoothly rolled to give a mirror-like surface, was placed on a smooth glass surface—a circular disc of glass was often used, but that was not essential—and a layer of paint was applied to the tin foil. The paint was often applied when the disc was being spun, but other methods could be used. Test pieces could be cut from the coated tin foil and were floated on a reservoir of mercury. In that way an excellent film was obtained. When paint was applied to ordinary tin plate a very smooth surface was never obtained and the amalgamation was not so good. He recommended the tin foil method very strongly.

Dr. Talen mentioned the reference, on page 715 of the paper, to the pigment volume concentration of a pigmented emulsion paint. He thought that the behaviour of such a paint was dependent on the manner of pigmentation; he asked how the author had pigmented the emulsion paint, because it was often done by dispersing the pigment in the water phase and the polymer emulsion was added afterwards. After drying, the pigment was distributed between the polymer particles, but was not dispersed homogeneously in the polymer. In this case the meaning of P.V.C. was not clear.

MR. WILLIAMS thanked Dr. Talen for his remarks concerning film preparation. He said that his company, in making experiments, had used tin plate to carry films, subsequently amalgamating by the use of mercury. The use of tin foil on a sheet of glass was certainly well worth considering. Concerning pigment dispersion in an emulsion paint, he appreciated the difficulty. However, they had not been able with emulsion paints to disperse the pigment particles in the polymer particles so that each pigment particle was coated with polymer. This would be an advantage, provided that such a system could be made stable in storage. At present, coating of the pigment with the polymer occurred after brushing out the emulsion paint, during the time the water was evaporating and the polymer particles were coalescing. Efficient pigment wetting by polymer at that stage was vital in order to produce good paint films.

DR. E. SUNDERLAND drew attention to Fig. 14 in the paper, dealing with water absorption, and suggested there was a much too complicated set-up in drawing the curve. Such a curve resulted from a complex of variables and tended to hide the realities of the situation. He inquired whether the author had attempted to analyse these curves according to the contributions of the different components in these non-homogeneous systems to the total water absorptions, or alternatively had attempted to synthesise this kind of curve from the determinable water absorptions of the various film components.

MR. WILLIAMS replied that the purpose of the work he had done on water absorption was to gain some knowledge of the system. He knew that the system varied in density and that there were different contributions to the total water absorption. Unfortunately, however, he could not at the moment think of any other way of determining the P.V.C. of a series of paints than by adhesion, water vapour permeability or water absorption. All these means were used and gave in this particular instance very good correlation with one another. For obtaining the P.V.C. figure, he thought that the means they were using represented an improvement on the old oil absorption method.

since, in contrast with the other method, the performance of the actual paint film, and not the separate components in different media, was being evaluated.

MR. N. R. FISK suggested that some trouble was caused and quite a lot of work wasted, because it was often not known what the co-monomers were. His point was a general one and did not relate in particular to the authors' paper, but he asked if the author would support him in urging that more information should be given about the co-monomers.

MR. WILLIAMS agreed with Mr. Fisk; with regard to composition, he said that the materials used in the work were made under closely controlled conditions, so that he and his colleagues knew the precise composition of the finished product.

MR. J. H. W. TURNER felt that there was always too much over-simplification. He recalled that Mr. Hodgson and himself had presented a paper to the Llandudno Conference, and had then postulated on very limited grounds that the film formation might involve a stage at which there was first dispersion of the colloids in a virtually continuous film of polymer and plasticiser. With plasticising, a protective colloid might separate out as another phase, forming a sort of laminate, giving almost a three-ply structure. That theory had explained some of the results they had obtained in tensile strength determinations. They had examined the strength of films in much the same way as the author had done at intervals of 24 hours, 3, 7, 14 and up to 30 days, and at intervals of six weeks and six months. Mr. Turner demonstrated the type of tensile strength/time curves obtained. He said that the curves rose initially to a peak, then tailed off and flattened out. The initial part of the curve depended on the type of plasticiser used, although the system was a copolymer system. The peak was reached at periods of between 14 and 30 days after application of load.

The weakness of the work the author had reported, so far as tensile strength determinations were concerned, was that those determinations came somewhere on the steeply rising part of the curve. The questioner wondered whether the figures the authors had obtained, showing increasing tensile strength, were double the figures he would have reported if he had made the determinations a few days later.

Mr. Turner added that the results which Mr. Hodgson and himself had obtained in those earlier experiments were not absolutely consistent, sometimes the peaks had been reached later and sometimes earlier than at other times. They did not finish the work, but Mr. Turner said that he had received a private communication concerning similar work by a team in the United States who had found exactly the same type of peak. They had obtained specific rates of increase of tensile strength which were related, not to the copolymer or homopolymer they used, but to the protective colloids. For these protective colloids the peak formation could be just as important as for the major components, the copolymer or homopolymer.

MR. WILLIAMS replied that some work which was done subsequent to the preparation of his paper had shown the same sort of thing as Mr. Turner had illustrated, but the curve was quite significantly different. The curve began nearer the top of the vertical axis, and there was a peak. The level of the curve was about 10 per cent higher than was obtained previously. In other words the figures obtained were not related to the steep part of the curve. He believed that it was influenced by the types of polymers used, and that the plasticising component was very important in this connection. It should be remembered that p.v.a. homopolymers contained a very small amount of unreacted monomer which would only take a short time to leave the film. However, some copolymers could, if not very carefully prepared, contain appreciable quantities of unreacted plasticising comonomer, and films of these resins could well show the tensile strength history mentioned by Mr. Turner, owing to the slow loss of this comonomer from the film.

DR. J. A. W. VAN LAAR, in a written contribution, also referred to Fig. 14, and he wondered whether the film weight was restored to its original value after removal from the relative humidity of 100 per cent. A R.H. of 100 per cent, he added, was not a well defined condition, for at an infinitely small reduction of temperature a liquid water phase must appear, and indeed this could easily happen. A CPVC of 50 per cent was surprisingly high compared with values found for a homogeneous distribution of pigment. In view of the heterogeneous distribution of pigment in emulsion paints, an even lower value than the usual one (20—30 per cent) would be expected. The minimum in Fig. 14 might be the result of several contrasting factors, such as swelling or leaching of vehicle, protective additives, water impurities and pigment contamination.

Information Received

(In case of difficulty regarding addresses, members and subscribers to the Journal should apply for details to the General Secretary of the Association at the address shown on the front cover.)

The *Filamatic* automatic filling machine, introduced by SHANDON SCIENTIFIC COMPANY some months ago, is now available in an air-powered version specially designed for the handling of inflammable liquids. Interchangeable pump units adapt the filler for handling liquids of varying viscosities. All parts in contact with liquid are of stainless steel or *Teflon*, and glass and plastic pump assemblies are available for metal-sensitive liquids. Also marketed by the same company are the *Sauter Fully Automatic Scales*, which enable complete formulations to be carried out quickly and accurately; no second balance is needed for the separate weighing of constituents.

It is announced that E. R. & F. TURNER LTD. are now producing a dry material batch mixing machine, the *Turner-Heesen Ultra-Rapid Square Mixer*. It is stated that the machine produces an homogeneous mix in about one-quarter of the standard mixing time. Because of their design they occupy less space than traditional mixers.

A range of powder blenders manufactured by T. P. CHEMICAL ENGINEERING CO. LTD. is designed for the rapid blending of powders of any type. It is stated that agglomeration is eliminated and that particle size can also be reduced. An outstanding increase in output is claimed over existing mixers, together with a considerable reduction in blending time.

The introduction of the *Dispersonic* has been announced by ULTRASONICS LTD., being the latest in their range of ultrasonic homogenising and dispersing units based on the principle of the liquid whistle. It has been specially designed for the high-speed dispersion of powders in liquids. It is stated that a wide range of powders, including carbon black, colloidal clay and titanium dioxide, can be dispersed rapidly.

SHELL INTERNATIONAL CHEMICAL CO. LTD. have announced the availability in pilot plant quantities of a new group of synthetic organic acids known as *Versatic*. The present samples are a blend of C₉, C₁₀ and C₁₁ monocarboxylic acids. Commercial production is planned for 1962 at a new plant being built at Pernis, near Rotterdam.

A new French company will be formed jointly by ETABLISSEMENTS KUHLMANN and DU PONT DE NEMOURS & Co. for the manufacture and sale of isocyanates; the new company will be known as DEKACHIMIE. The construction of plant near Lille will be completed in 1963.

NOVADEL LTD. have recently announced that A. J. GEMMILL & Co. LTD., of Glasgow, have been appointed as agents for Novadel products in Scotland.

It has been announced that the Hindley factory of the KEYSTONE PAINT & VARNISH CO. LTD., a member company of the BERGER, JENSON & NICHOLSON paint group, is to re-commence production, owing to increasing demands for certain products manufactured by the group.

The amalgamation of ALCHEMY LTD. with the chemical section of BURTS & HARVEY LTD. has been announced. The integrated company will operate under the name of the latter.

A technical publication received from ARMOUR HESS CHEMICALS LTD. is entitled *Stripping Agent Formulations for Protective Coatings*. It includes formulations based on their cationic surface active chemicals; of particular interest is a formula recommended for stripping epoxy resin finishes.

The ALBRIGHT & WILSON Group of chemical companies has produced an attractive, extensively illustrated brochure giving up-to-date information on the activities of the Group.

The following publications have been received and are now available: *Facts About Degreasing* and *Facts About Rust Removal and Rust Prevention* from BASOL LTD.; *Crodex* and *Cerewax Emulsifying Waxes* from CRODA LTD.; *Elastomers Notebook* from DU PONT (U.K.) LTD. *Thermex Heat Transfer Medium* from I.C.I. LTD.; *Pye Argon Chromatograph* from W. G. PYE & CO. LTD. Technical information leaflets: *Thiodipropionic Acid* from EVANS CHEMETICS INC.; *Perkadox IPP* (di-isopropyl peroxy dicarbonate) from NOVADEL LTD.

FOURTEENTH TECHNICAL EXHIBITION

26, 27, 28 February and 1 March, 1962

On the first occasion that the Exhibition has been organised by a Committee directly appointed by the Council, the Committee has allocated 101 stands, covering a record floor area of nearly 15,000 square feet, for the Fourteenth Technical Exhibition, which will take place in the Old and New Halls of the Royal Horticultural Society, London, S.W.1, on 26, 27 and 28 February and 1 March, 1962.

The Exhibition, which provides a focus for the technical display of advances made in raw materials, plant and equipment for the paint, printing ink and allied industries, will again be open on four days, as in 1961. Of the number of companies showing at the 1962 Exhibition, seventeen have never shown before at an O.C.C.A. Technical Exhibition, while nine others did not show at this year's Exhibition. There is an increase in floor area, compared with the 1961 Exhibition, of over 11 per cent.

Directly represented at the Exhibition are companies from the European Free Trade Association (Sweden), the European Common Market (France, Germany and Holland) and the United States of America. There are in addition many more overseas companies who are showing through their British associates. It is pleasing to record that this will be the first occasion on which a French company will be showing directly at an O.C.C.A. Exhibition.

A central section in the Old Hall will be devoted to a Technical Education Stand, on lines similar to that at the Thirteenth Technical Exhibition, together with the Paint Research Station. Parties of sixth form science students will once again be invited on two mornings only, when they will be given short introductory talks by members of the Association in a separate lecture hall, before visiting the Exhibition. The Technical Education Stand will be organised by representatives from both the trade organisation (Paint Manufacturers' Joint Executive Council and the Society of British Printing Ink Manu-

facturers) and technical colleges, who will show the courses available in the technology of the paint, printing ink and allied industries. It is felt that this will be of interest not only to new entrants but also to those who are concerned with the training of junior technical personnel.

A record number of nearly 10,000 people from the United Kingdom and twenty-three overseas countries attended the last Exhibition, and a further increase is expected for the 1962 Exhibition. In order to assist the increasing number of overseas visitors and exhibiting companies, there will again be interpreters in attendance.

The Exhibition Luncheon will once again be held at the Criterion Restaurant, Piccadilly, London, W.1, on 26 February, and principal officers of other scientific bodies, industrial research associations and organisations representing both suppliers and consumers will be invited to attend.

The hours of opening will be as follows:

Monday,	
26 February	.. 3 p.m. to 7 p.m.
Tuesday,	
27 February	.. 10 a.m. to 7 p.m.
Wednesday,	
28 February	.. 10 a.m. to 7 p.m.
Thursday,	
1 March	.. 10 a.m. to 7 p.m.

There will be no charge for admission to the Exhibition or for the *Official Guide*, which will be available from the Information Bureaux, or from the Association's offices prior to the Exhibition.

Buffet facilities will be available in both Halls for the convenience of visitors to the Exhibition, and a full restaurant service will be available from 12.30 p.m. to 2.30 p.m. on the second, third and fourth days.

Further information can be obtained from the General Secretary, R. H. Hamblin, M.A., F.C.I.S., F.C.C.S., Oil and Colour Chemists' Association, Wax Chandlers' Hall, Gresham Street, London, E.C.2. Tel.: MONarch 1439.

List of Exhibitors on next page

Alphabetical List of Exhibitors

- Albro-Fillers & Engineering Co. Ltd.
 Allied Colloids Ltd.
 *Amoco Chemicals Corporation
 A.P.V. Company Ltd., The
 Armour Hess Chemicals Ltd.
 Associated Lead Manufacturers Ltd.
 *Astles Chemical Co. Ltd.
 Bakelite Ltd.
 Baldwin Industrial Controls
 *Barter Trading Corporation Ltd. (Kemisk
 Vaerk Koge, A/S)
 Beck, Koller & Co. (England) Ltd.
 Berk, F. W., & Co. Ltd.
 B.I.P. Chemicals Ltd.
 Boehm, Frederick, Ltd.
 Boulton, William, Ltd.
 British Celanese Ltd.
 British Oil & Cake Mills Ltd.
 British Oxygen Chemicals Ltd.
 British Resin Products Ltd.
 British Titan Products Co. Ltd.
 *Buhler Brothers (England) Ltd.
 Burts & Harvey Ltd.
 *Bush, Beach & Segner Bayley Ltd.
 Campbell, Rex, & Co. Ltd.
 Carless, Capel & Leonard Ltd.
 *Chemische Werke Huel A.G.
 Ciba (A.R.L.) Ltd.
 Ciba Clayton Ltd.
 Cornelius Chemical Co. Ltd.
 *Coulter Electronics Ltd.
 Cray Valley Products Ltd.
 Crosfield, Joseph, & Sons Ltd.
 Croxton & Garry Ltd.
 Distillers Co. Ltd.
 *Dow Chemical Co. (U.K.) Ltd.
 Dunlop Rubber Co. Ltd.
 Durham Raw Materials Ltd.
 Elliott, H. J., Ltd.
 Esso Petroleum Co. Ltd.
 Evans Electro selenium Ltd.
 Farbenfabriken Bayer Aktiengesellschaft
 Ferranti Ltd.
 Geigy Co. Ltd.
 Golden Valley Colours Ltd.
 Goodyear Tyre & Rubber Co. (G.B.) Ltd.
 Hardman & Holden Ltd.
 Hercules Powder Co. Ltd.
 Imperial Chemical Industries Ltd.
 Johnson, Matthey & Co. Ltd.
 *Joyce, Loebel & Co. Ltd.
 Kek Ltd.
 *Kingsley & Keith (Chemicals) Ltd.
 Kunstharsfabriek Synthese, N/V
 Laporte Titanium Ltd.
 *Lennig, Chas., & Co. (G.B.) Ltd.
 Marchant Brothers Ltd.
 Metal Propellers Ltd.
 Micafine Ltd.
 Mitchell, L. A., & Co. Ltd.
 Monsanto Chemicals Ltd.
 *Morris & Ingram (London) Ltd.
 National Adhesives Ltd.
 *Noss Mayo Dispersion Products Ltd.
 Novadel Ltd.
Paint Manufacture
Paint, Oil & Colour Journal
 Paint Research Station, The
Paint Technology
 Pascall Engineering Co. Ltd.
 *Pechiney-Saint-Gobain
 Plastanol Ltd.
 Premier Colloid Mills Ltd.
 Price's (Bromborough) Ltd.
 Research Equipment (London) Ltd.
 Resinous Chemicals Ltd.
 *Runnymede Dispersions Ltd.
 *Scado-Archer Daniels, N.V.
 Schenectady-Midland Ltd.
 Scott Bader & Co. Ltd.
 Shawinigan Ltd.
 Sheen Instruments (Sales) Ltd.
 Shell Chemical Co. Ltd.
 Silverson Machines (Sales) Ltd.
 Spelthorne Metals Ltd.
 Surface Coating Synthetics Ltd.
 Svenska Oljeslageri Aktiebolaget
 Swada Ltd. & H. Haeffner & Co. Ltd.
 Tin Research Institute
 Trade & Industrial Press Ltd.
The Paint Journal
 *Translation & Technical Information
 Service
 Union Carbide Ltd.
 Universal Oil Co. Ltd., Premier Oil &
 Cake Mills Ltd. & J. L. Seaton & Co.
 Ltd.
 Vinyl Products Ltd.
 Winkworth Machinery Ltd.
 Wolf, Victor, Ltd.
 Younghusband Stephens & Co. Ltd.
 Technical Education Stand
 O.C.C.A. Information Bureaux

*Denotes exhibitors who have not shown at previous Exhibitions

Register of Members

The following elections to membership have been approved by the Council. The Sections to which the new members have been attached are given in italics.

Ordinary Members

- BRAITHWAITE, ALAN, B.S.C., 2 Hawthorn Avenue, Holcombe Brook, Nr. Bury, Lancs. (*Manchester*)
- BRAND, GEORGE DONALD, A.M.INST.F., 70 Kinross Road, Totton, Hants. (*London*)
- BULT, REINT, Kastanjelaan 10 Ryswyk (Z.H.), Holland. (*Overseas*)
- CUTLER, DAVID HOLLAND, M.A., Laporte Industries Ltd., Hanover House, Hanover Square, London, W.1. (*London*)
- DONALDSON, ALBERT EDWARD, B.S.C., 43 Wingate Street, East Bentleigh, S.E.15, Victoria, Australia. (*Victorian*)
- MANZ, WILMAR, B.S.C., PH.D., A.R.I.C., c/o E. I. Du Pont de Nemours Inc., Bush House, Aldwych, London, W.C.2. (*London*)
- PRICE, DEWI, B.S.C., A.R.I.C., M.INST.P., "Cartref", Bridle Lane, Loudwater, Rickmansworth, Herts. (*London*)
- SHIELDS, BRYAN DOUGLAS CLIVE, B.A., Shell Chemical Co. Ltd., 15/17 Great Marlborough Street, London, W.1. (*London*)
- SIMMONS, PAUL, B.A., B.S.C., 22 Aylesford Way, Stapleford, Cambs. (*London*)
- SNELL, ROBERTON ERNEST, B.S.C., 37 Upper Brownhill Road, Maybush, Southampton, Hants. (*London*)
- WILLSMER, LEICESTER CHARLES, Little Wood, Kings Road, Sunninghill, Berks. (*London*)

Associate Member

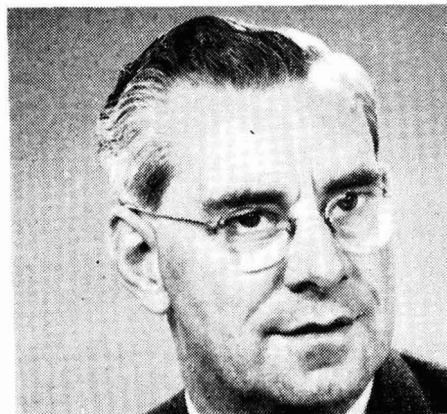
- BUCHANAN, JAMES MITCHELL, 121 Brookton Road, Roleystone, Western Australia. (*South Australian*)

Junior Members

- CARTER, TERENCE JOHN, 33 Victoria Street, Oak Park, W.6, Victoria, Australia. (*Victorian*)
- CRAINE, IVOR BARRY, 6A Elm Grove, Armadale, Melbourne, S.E.3, Victoria, Australia. (*Victorian*)
- GOUGH, HARVEY JOHN, Lot 35, Meryl Street, East Doncaster, Victoria, Australia. (*Victorian*)
- PITFIELD, PETER JOHN IXER, 26 Melrose Road, Wandsworth, London, S.W.18. (*London*)

NEWS OF MEMBERS

Mr. E. S. Lower, an Ordinary Member, attached to the Hull Section, is Managing Director (Research) of Croda Ltd. and a member of the main board of the Croda Organisation Ltd. He has recently returned from a three-week stay in the United States, where he visited Croda Inc. and many industrial users of their products.



Mr. R. F. York, an Ordinary Member attached to the London Section, has been appointed Home Sales Manager of Howards of Ilford Ltd.

Mr. L. Bottomley, an Ordinary Member attached to the London Section, has been appointed to the Board of Croxton & Garry Ltd. as Technical Director. Mr. Bottomley joined the company in 1958.



L. BOTTOMLEY

Mr. G. E. Westwood, an Ordinary Member attached to the London Section, has been elected to Fellowship of the Royal Institute of Chemistry.

3RD SLF CONFERENCE

The Third Conference of the Federation of Scandinavian Paint and Varnish Technicians, which will take place in Oslo from 12-14 October, will be opened by Mr. E. Lund, President of the Federation. The programme includes eight lectures (mainly in Scandinavian languages), a panel discussion, and visits to the research centre at Blindern and several manufacturing companies.

ADHESION

The Annual Lecture of the Technical Training Board for the Printing Ink and Roller Making Industry will be given by Dr. A. W. Hendersen on "Adhesion".

The lecture will take place at the Royal Institution, 21 Albemarle Street, London, W.1, at 7 p.m. Those interested other than students following courses in printing ink and roller technology are welcome to attend and they are requested to apply for tickets to the Secretary of the Board, Burley House, Theobalds Road, London, W.C.1.

KEYNOTE ADDRESS

The Federation of Societies for Paint Technology have announced that Dr. Carl F. Prutton, Director-Consultant of Food Machinery and Chemical Corp., will present the Keynote Address at the 39th Annual Meeting of the Federation, which will be held at the Shoreham Hotel in Washington, D.C., U.S.A., on 2-4 November.

C.I.T.A.

A new committee known as the Committee for International Technical Assistance (C.I.T.A.) has been established by the Federation of Societies for Paint Technology. The function of this committee will be to render on request technical assistance to countries with an infant paint industry, or countries about to start such an industry. The technical co-operation to be rendered by the committee will be accomplished by (a) correspondence, (b) sending experts (preferably retired) from the ranks of Federation members to countries requesting assistance, and (c) assisting foreign technicians who visit the United States.

B.S. 1014 : 1961

The following British Standard has recently been issued:

B.S. 1014 : 1961 Pigments for cement, magnesium oxychloride and concrete.

This publication has been prepared under the supervision of the Pigments, Paints and Varnishes Industry Standards Committee on which the Association is represented. Copies are obtainable from the British Standards Institution, British Standards House, 2 Park Street, London, W.1, price 7s. 6d.

Forthcoming Events

(Note: Details are given of meetings arranged in the U.K. up to the 15th of the month following publication, and in South Africa and the Commonwealth up to the 15th of the second month after publication.)

Monday, 2 October

Victorian Section. "The Paint Approvals Scheme", by J. Rischbieth, B.Sc., Secretary of the Commonwealth Paint Committee.

Tuesday, 3 October

Bristol Section. "Management Succession as a Field for Operational Research", by Lord Halsbury. Joint Meeting arranged by the Bristol and Bath Productivity Association, to be held in the Engineering Lecture Theatre of Bristol University.

Wednesday, 4 October

Association Council Meeting at 2.30 p.m.; *Past Presidents' Dinner* at 6.30 p.m. for 7 p.m.; at Wax Chandlers' Hall, Gresham Street, London, E.C.2.

Thursday, 5 October

Newcastle Section. "Consideration of Different Types of Film Formation", by Dr. H. W. Talen, at the Royal Turks Head Hotel, Newcastle upon Tyne, at 6.30 p.m.

Friday, 6 October

Hull Section. Dinner and Dance at the New York Hotel, Anlaby Road, Hull. Reception 7 p.m.

Newcastle Section Junior Group. "Solvents, Driers and Additives", by C. N. Finlay, at the Rutherford College of Technology, Northumberland Road, Newcastle upon Tyne, at 3 p.m.

Scottish Section. "Consideration of Different Types of Film Formation", by Dr. H. W. Talen at Edinburgh (details are to be announced later).

Tuesday, 10 October

West Riding Section. "Public Service Vehicle Finishing", by A. Gellman, of the London Transport Executive, at the Hotel Metropole, Leeds, at 7.30 p.m.

London Section. "Consideration of Different Types of Film Formation", by Dr. H. W. Talen (Director of Paint

Research Institute, T.N.O., Delft), at the Chemical Society, Burlington House, London, W.1, at 7 p.m.

Thursday, 12 October

New South Wales Section. Factory Inspection — Shell Chemicals Epoxy Resin Plant and Laboratory.

Scottish Section. "The Microbiology of Paints", by D. J. Cowley and Miss Brown, in the Grand Hotel, Charing Cross, Glasgow, at 7.30 p.m.

Technical Training Board for the Printing and Roller Making Industry. "Adhesion", by Dr. A. W. Hendersen (The Plessey Co. Ltd.). Annual Lecture at the Royal Institution, 21 Albemarle Street, London, W.1, at 7 p.m.

Friday, 13 October

Victorian Section. Ladies' Night at Ciro's, Collins Place, Melbourne, Victoria.

Manchester Section. "Modern Chemistry of Organic Pigments", by Dr. H. Gartner, at the Literary and Philosophical Society, 36 George Street, Manchester, at 6.30 p.m.

Saturday, 14 October

Scottish Section Student Group. "Carpet Manufacture", illustrated with coloured slides, by D. Macphee, at More's Hotel, 18 India Street, Glasgow, C.2, at 10 a.m.

Wednesday, 18 October

London Section. "Solvents in the Surface Coating Industry", by L. M. Barakan, PH.D. (The Distillers Co. Ltd.), in the Lecture Theatre of the Royal Society of Tropical Medicine and Hygiene, Manson House, 26 Portland Place, London, W.1, at 7 p.m.

Friday, 20 October

Irish Branch of the Bristol Section. Speaker from Irish Oil and Cake Mills Ltd.

Midlands Section. "Printing Processes for Paper", by K. D. C. Bruce, and "Paper Preparation and Treatment", by K. Wilkinson, B.Sc., at Mander Bros., St. John's Street, Wolverhampton.

Wednesday, 25 October

Western Australian Branch. "Artists' Colours", by Mr. Philpott, at the Builders Exchange, Havelock Street, West Perth.

Friday, 27 October

Manchester Section. Annual Dinner and Dance at the Alexandra Suite, Midland Hotel, Manchester, at 7.15 p.m.

Bristol Section. New Chairman's Address by A. Aitkenhead, at the Royal Hotel, College Green, Bristol, 1, at 7.15 p.m.

Sunday, 29 October

Victorian Section. Car Rally. Destination unknown (but on the map).

Thursday, 2 November

Newcastle Section. A paper on Metal Finishing (title to be decided), by R. M. C. Logan, at the Royal Turks Head Hotel, Newcastle upon Tyne, at 6.30 p.m.

Friday, 3 November

Newcastle Section Junior Group. "Formulation", by K. F. Baxter, in the Rutherford College of Technology, Northumberland Road, Newcastle upon Tyne, at 3 p.m.

Thursday, 9 November

New South Wales Section. "Thermosetting Acrylics", by Dr. B. James, at the Theatre, M.C.L. Building, Miller Street, North Sydney, at 6.15 p.m.

Scottish Section. "Solvent Selection for Modern Finishes", by J. E. Loible, in the Grand Hotel, Charing Cross, Glasgow, at 7.30 p.m.

London Section. "Applications of Microwave Spectroscopy", by R. S. Tebble, B.Sc., Ph.D., F.Inst.P. (Department of Physics, Sheffield University), in the Lecture Theatre, Royal Society of Tropical Medicine and Hygiene, Manson House, 26 Portland Place, London, W.1, at 7 p.m.

Friday, 10 November

Manchester Section. "Television — A Persuasive Medium", by J. P. Phoenix, M.I.P.R., to be given at the Liverpool Constitutional Club, Tithebarn Street, Liverpool, at 6.30 p.m.

Monday, 13 November

Victorian Section. "Some Recent Developments in Latex Paints", by B. McGuinness, Jordan National Pty. Ltd.

Tuesday, 14 November

West Riding Section. "Advertising to and by Paint Firms", by N. R. Fisk, at the Hotel Metropole, Leeds, at 7.30 p.m.

Wednesday, 22 November

Western Australian Branch. Meeting—details to be announced.

Wednesday, 6 December

Victorian Section. Annual Dinner, Main Buffet, Melbourne University. Guest speaker to be arranged.

Thursday, 7 December

New South Wales Section. Annual Dinner at the "Amory".



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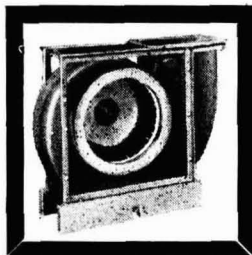
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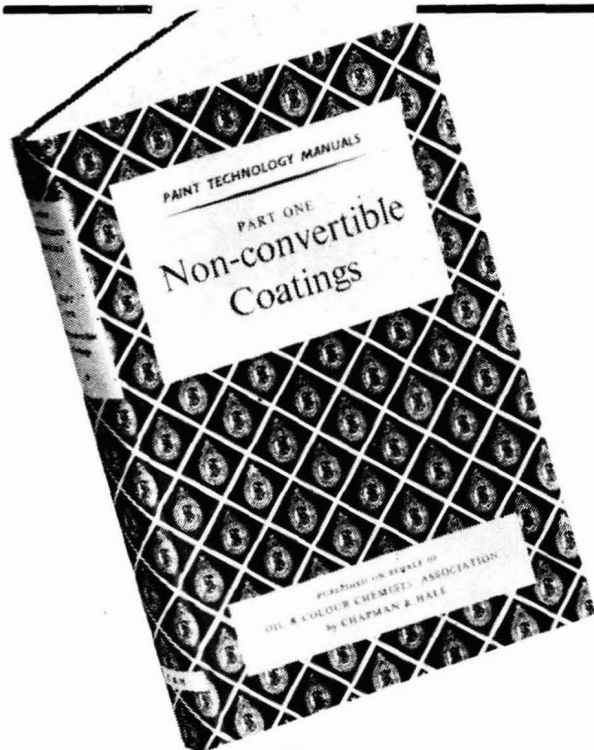
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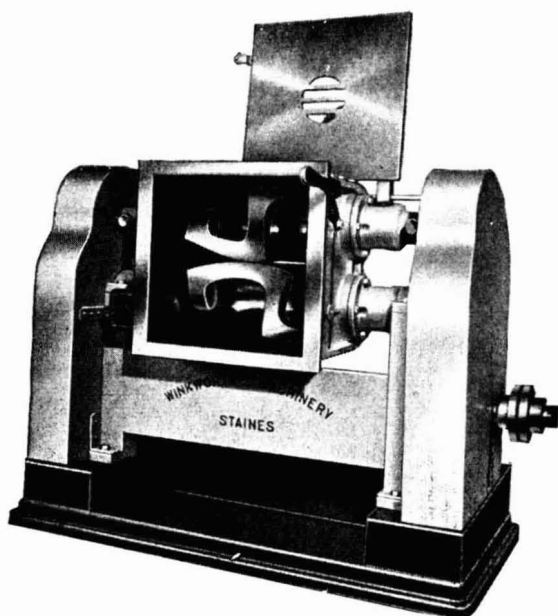
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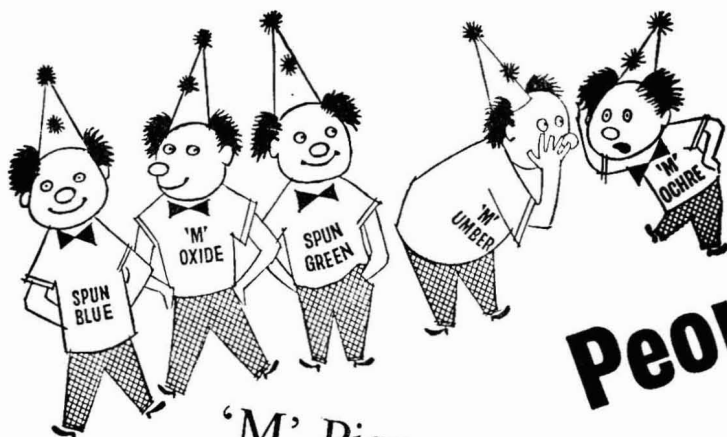
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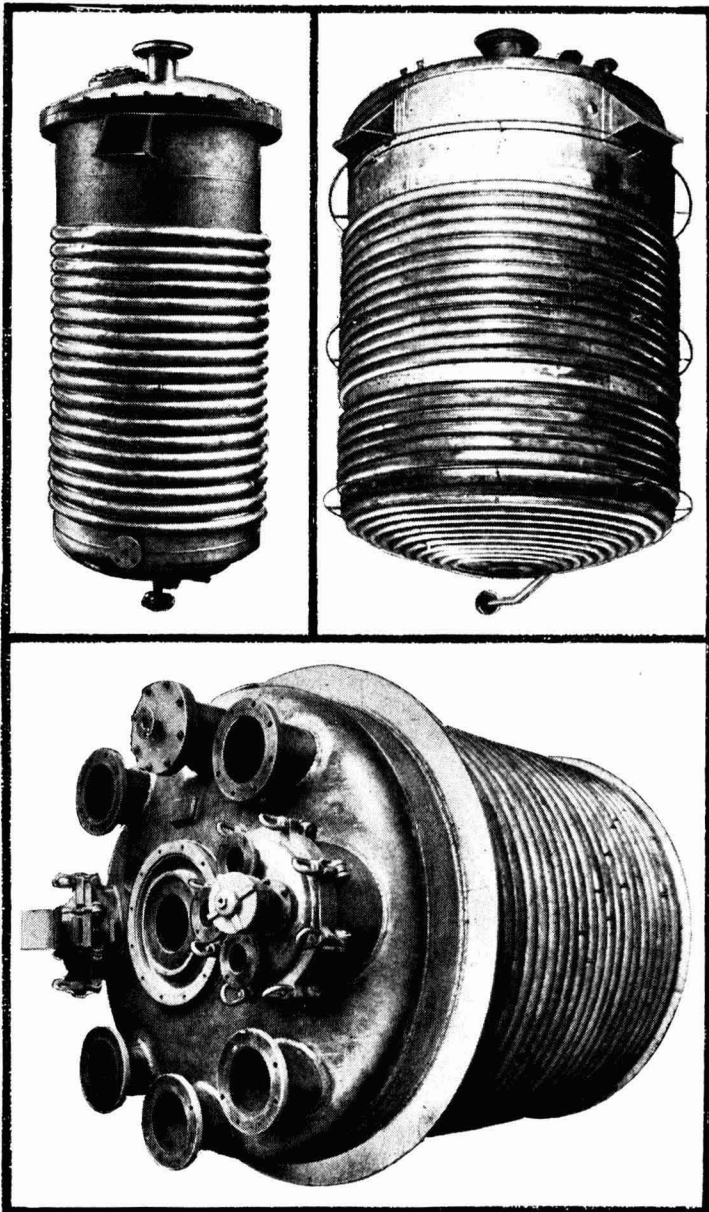
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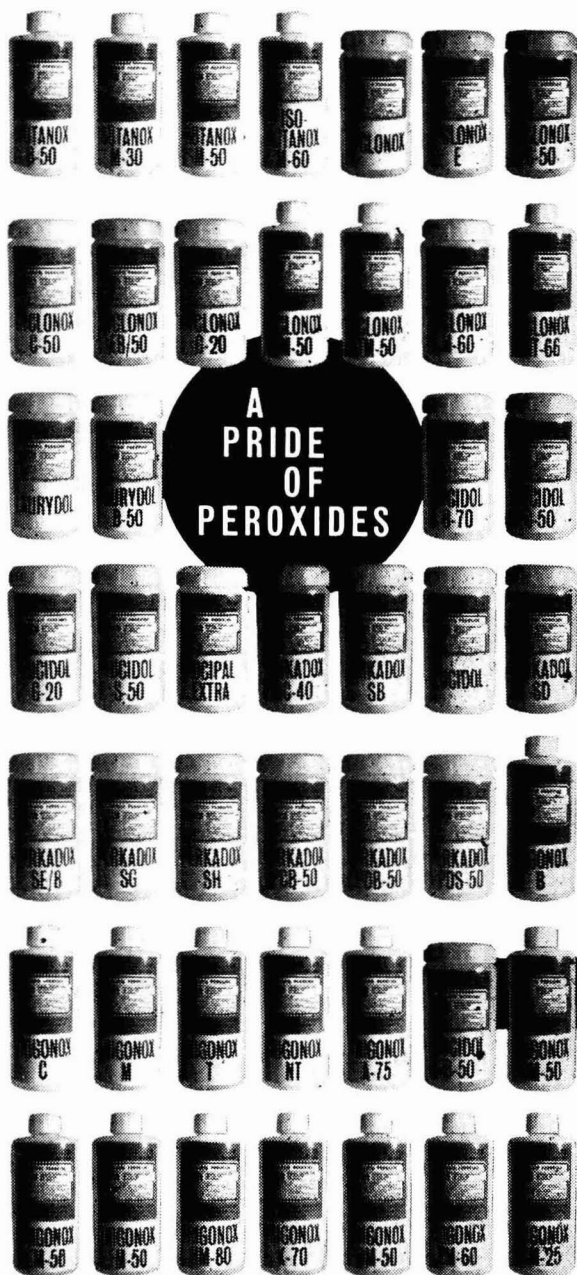


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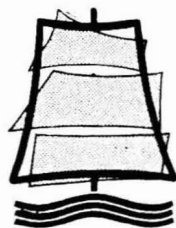
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