

JOURNAL OF THE OIL AND COLOUR CHEMISTS' ASSOCIATION



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September 1971

Papers from the Torquay Conference

Keynote address: The changing face of paint

T. R. Bullett

Appearance and performance factors in coatings for buildings

P. Whiteley and G. W. Rothwell

Colour specification by visual means

K. McLaren

Gas chromatographic study of the interaction forces between binder and solvent in paint films

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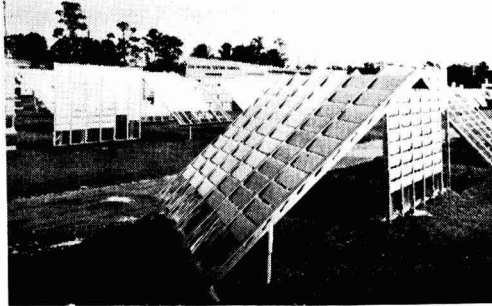
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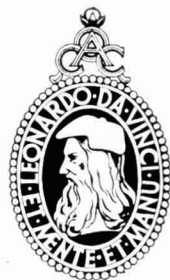
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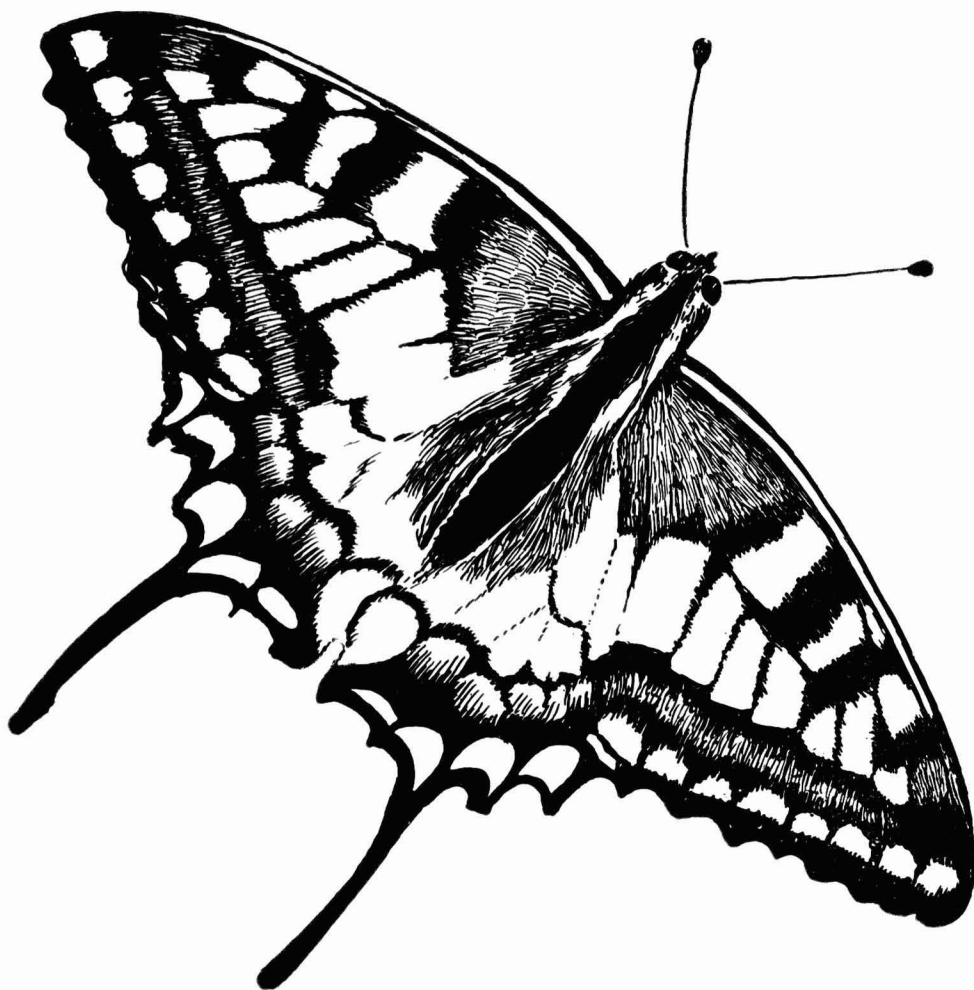
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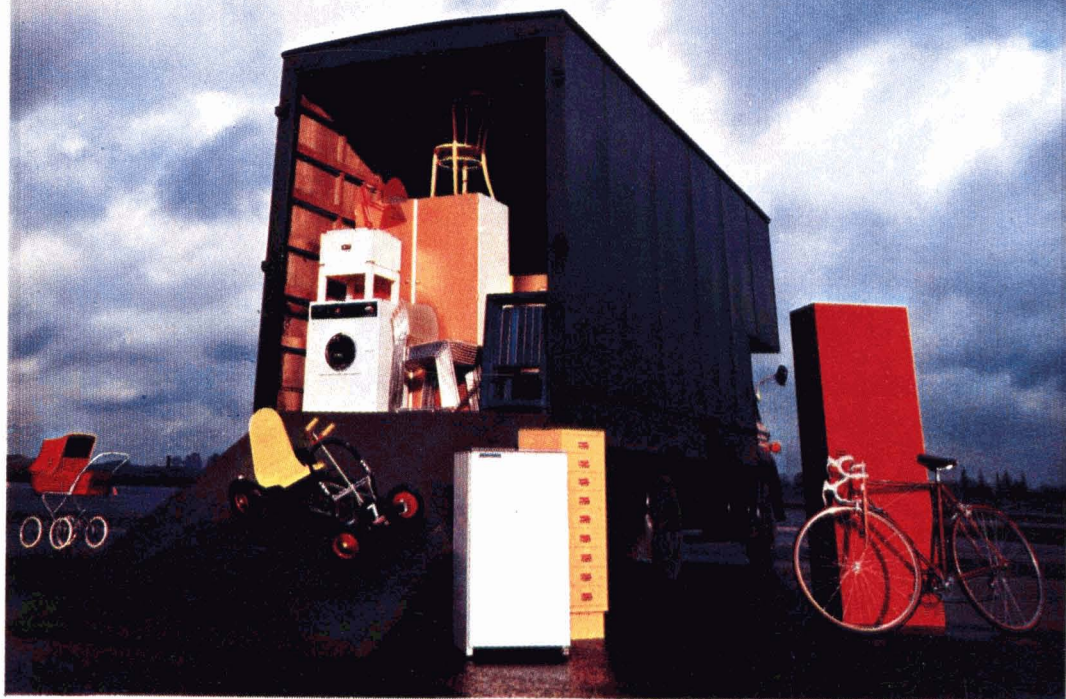
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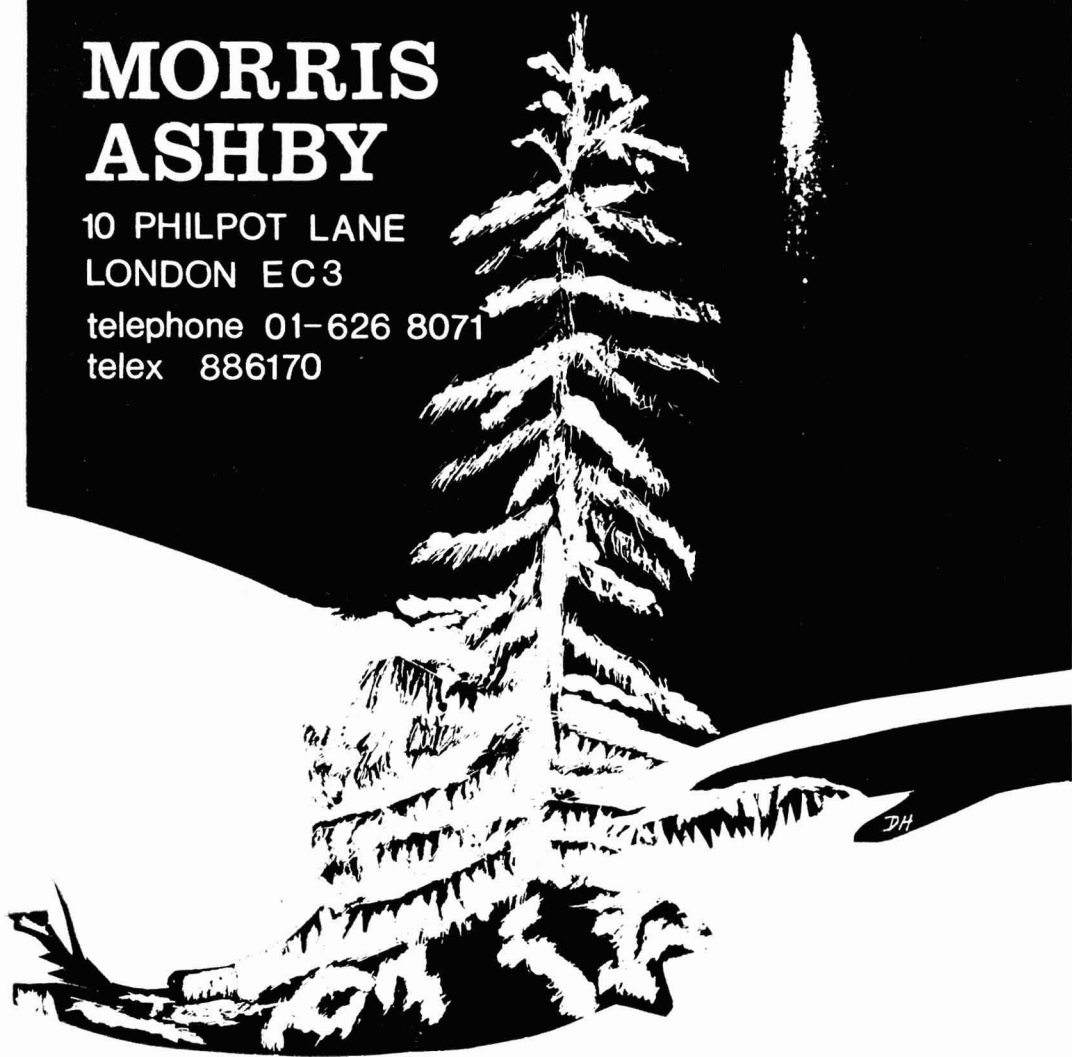


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Transactions and Communications

Keynote address: The changing face of paint*

By T. R. Bullett

The Paint Research Association, Waldegrave Road, Teddington, Middx.

Summary

Modern paints are, intrinsically, amongst the most durable of building materials; erosion rates, expressed as loss in thickness per year of unpainted steel, wood, brick and concrete, in urban atmospheres are all many times that of a good average paint film. New coating materials of even higher durability are now coming into use and, when applied under controlled conditions to properly prepared surfaces to ensure lasting adhesion, may be expected to maintain integrity for 20, 30 or more years. But integrity is not enough; the primary function of paint is decoration, so that the useful life of a coating is only the period for which it maintains a satisfactory appearance. "Loss of face" will increasingly become the main reason for repainting.

The paper reviews the ways in which a coating can change in appearance. There are external factors, such as retention of dirt, yellowing by tobacco smoke, the effects of other atmospheric pollutants, contamination by fungi and algae. Susceptibility to such factors and, perhaps more important, ease of cleaning is a function of paint composition. Building design can do much to reduce these troubles.

There are also internal changes in coatings leading to alterations in surfaces, such as the rearrangement of material around pigment particles that results in hazing, irregular shrinkage when retained solvent is finally lost and shrinkage due to weathering effects which normally precedes chalking. Finally, colour change in relation to pigment, pigment/medium interaction, exposure to light and ultraviolet radiation will be considered. An important problem, not yet entirely resolved, concerns assessment of colour changes. The new PRA fibre optics colorimeter will greatly facilitate measurement, but further study of methods for expressing colour differences in units correlating with visual judgment is desirable.

Keywords

Properties, characteristics and conditions primarily associated with dried or cured films

bloom
chalking
dirt retention
discoloration
fading
staining
yellowing

Equipment primarily associated with analysis measurement and testing

colorimeter
glass fibre optics

Discours Thème: Les aspects changeants de peinture

Résumé

Les peintures actuelles sont, intrinsèquement, parmi les matériaux de construction les plus durables; les taux d'érosion dans une atmosphère urbaine, exprimés comme la perte de l'épaisseur annuelle, de l'acier, du bois, de la brique, et du béton, sont tous maintes fois plus

*Presented to the Torquay Conference on 5 May 1971.

grandes que celle d'un feuil de peinture de bonne qualité moyenne. De nouveaux revêtements d'une durabilité même plus élevée viennent d'être utilisés, et quand ils sont appliqués sous des conditions réglementées aux surfaces préparées afin d'assurer une adhérence permanente, on doit s'attendre qu'ils retiendront leur intégrité pendant 20, 30 années ou de plus. D'ailleurs, l'intégrité elle-même ne suffit pas; la fonction primaire de la peinture est décorative, de sorte que la vie utile d'un revêtement soit seulement cette période où il maintient une apparence satisfaisante. A l'avenir la perte d'apparence deviendra de plus en plus la raison principale pour refaire la peinture.

La communication passe en revue la manière par laquelle un revêtement peut changer d'apparence. Il y a des facteurs externes tels que l'écrasement, le jaunissement par la fumée de tabac, les effets d'autres polluants atmosphériques, la contamination par moisissures ou algues. La susceptibilité à tels facteurs, et peut-être de plus importance, la facilité de mettre au net, est une fonction de la composition des peintures. On peut diminuer ces ennuis en respectant les exigences d'une bonne construction.

Il y a également des altérations dedans les revêtements qui provoquent des changements de leurs surfaces, tels qu'un remaniement de la matière autour des particules pigmentaires de sorte que mis en évidence sont un brillant voilé, une contraction irrégulière lors de la perte ultime du solvant retenu, et une contraction à cause de ces effets de vieillissement qui précèdent le commencement de farinage. Enfin on considère le changement du couleur pigmentaire, l'interaction pigment/milieu, l'exposition à la lumière et à la radiation ultra-violet. Un problème important, qui n'est pas tout à fait résolu, c'est l'appréciation des changements de couleur. Le nouveau colorimètre PRA à l'optique des fibres rendra plus facile la mesure de tels changements. D'ailleurs on doit prévoir une autre étude sur des méthodes pour exprimer les différences en couleur par les unités qui s'accordent avec l'appréciation visuelle.

Einleitende Ansprache: Das sich Ändernde Gesicht der Lackindustrie

Zusammenfassung

Moderne Anstrichmittel gehören tatsächlich zu den dauerhaftesten Baumaterialien. Erosionsgeschwindigkeiten von ungeschichtem Eisen, Holz, Ziegel und Beton in ländlicher Umgebung, ausgedrückt als jährlicher Schichtdickenverlust, sind allesamt mehrmals so gross wie die eines guten Anstrichfilms durchschnittlicher Qualität. Neue Anstrichmittel von noch höherer Dauerhaftigkeit werden jetzt in Verwendung genommen. Wenn diese unter kontrollierten Bedingungen auf ordnungsgemäss vorbereitete Oberflächen aufgetragen werden, so dass sie ihr Haftungsvermögen auf die Dauer beibehalten, kann man erwarten, dass sie 20, 30 Jahre oder noch länger unversehrt bleiben. Unversehrtheit genügt jedoch nicht; der Zweck eines Anstriches ist in erster Linie zu schmücken, sodass die nutzbare Lebensdauer eines Anstriches lediglich die Zeit umfasst, innerhalb welcher sein Aussehen genügt. "Verlust des Gesichts" wird mehr und mehr der Hauptanlass für den Neuanstrich.

Der Vortrag gibt eine Übersicht der Entwicklungen, welche zu Veränderungen des Aussehens eines Anstriches führen können: Dazu gehören äussere Einflüsse, wie z.B. Schmutzhaftung, Vergilbung durch Tabakrauch, die Auswirkungen anderer Verunreinigungen in der Luft, Verschmutzung durch Schimmelpilze und Algen. Empfindlichkeit gegenüber solchen Faktoren und, vielleicht noch wichtiger, die Leichtigkeit des Reinigens ist eine Funktion der Lackzusammensetzung. Der Bauentwurf kann viel dazu beitragen, diese Störungen zu verringern.

Ausserdem gibt es interne Veränderungen in Anstrichmitteln, die zu Veränderungen in Oberflächen führen, z.B. der Neuordnung der die Pigmentteilchen umgebenden Substanzen, welche zur Schleierbildung, nach Verlust zurückgehaltenen Lösungsmittels zu unregelmässigem Schrumpfen und als Auswirkung der Bewitterung zur Schrumpfung, die normaler Weise dem Abkreiden vorausgeht, führen. Schliesslich werden auch auf Pigment zurückzuführende Farbveränderungen, Reaktionen zwischen Pigment und Bindemittel, Lichteinflüsse, und ultraviolette Bestrahlung in Betracht gezogen werden. Ein wichtiges, noch nicht völlig gelöstes Problem betrifft die Bewertung von Farbänderungen. Das neue Faseroptik Kolorimeter der Paint Research Association wird solche Messungen wesentlich erleichtern; weitere Suche nach Methoden, mit deren Hilfe Farbunterschiede in Einheiten ausgedrückt werden können, welche mit visueller Beurteilung übereinstimmen, ist erforderlich.

Меняющийся облик красок

Резюме

Современные краски по существу принадлежат к классу одних из самых устойчивых строительных материалов; скорость эрозии выраженная как годовая потеря в толщине в неокрашенной стали, дереве, кирпиче и бетоне в городской атмосфере, намного превышает потери в хорошей красочной пленке среднего качества. Новые материалы покрытий более высокого качества входят в потребление в настоящее время и применяя их в контрольных условиях на надлежащим образом приготовленных поверхностях для обеспечения продолжительной адгезии, можно ожидать что они сохранят целостность на 20, 30 и более лет. Но целостность недостаточна; первой функцией краски является украшение и поэтому полезная жизнь покрытия определяется только периодом в течение которого оно сохраняет удовлетворительный облик. Потеря облика становится все более главным поводом для перекрашивания.

Статья описывает условия при которых покрытие может изменить свой вид. Существуют внешние факторы, как например накопление грязи, пожелтение табачным дымом, влияние прочих атмосферных загрязняющих веществ и загрязнение плесенью и водорослями. Чувствительность к этим факторам и что более важно, легкость очистки, является функцией структуры краски. Планы и расчет строений могут намного помочь в устранении этих трудностей.

Существуют также внутренние изменения в покрытиях, которые приводят к изменению поверхностей, как например перераспределение материала вокруг пигментных частиц, что ведет к помутнению, неравномерной морщинистости, когда удержанный растворитель окончательно утерян, и к сморщиванию за счет влияний выветривания, которые обычно предшествуют известкованию. Наконец обсуждается изменение цвета по отношению к пигменту, взаимодействию между пигментом и средой и облучению светом и ультрафиолетовым излучением. Важная задача, еще не вполне решенная, касается вопроса оценки изменений цвета. Новый волоконистый оптический колориметр PRA (Общества для Исследования Красок) намного облегчит измерения, но желательно дальнейшее изучение методов описания разниц цвета в единицах более согласованных с визуальной оценкой.

Introduction

In choosing Leonardo as its spiritual patron, OCCA implicitly recognised the dual function of paint, to enhance appearance and to fill a practical engineering purpose, for Leonardo was both an artist and an engineer. Throughout history and in different climates and situations, sometimes one function has dominated and sometimes the other. Thus, paint in the prehistoric caves of Lascaux and Altamira was purely decorative, being used to convey an ephemeral visual message somewhat as a political extremist might write on the wall today; yet strangely these cave paintings have survived for over ten thousand years. Resistance in a virtually unchanged condition for hundreds or even thousands of years requires not only paint that is intrinsically very stable but also a climate that ensures that the substrate shall not be attacked. To take another example, the fresco cave paintings of Ajanta, in India, dating from 200 BC to 600 AD, which were executed on mud plaster, have survived in a remarkably fine condition (as can be seen from the reference given) because of the low humidity and equable conditions of the environment¹. On the other hand, in adverse conditions where protection of the substrate has been the main aim, as on ship bottoms or steel bridges, the effective paint life has usually been less than ten years. But a new situation is now arising where both protection and decoration are demanded over almost indefinite periods, for example on the fascias of high rise buildings.

The purpose of this Conference is to explore the factors controlling the establishment and maintenance of the desired appearance in paint films. One reason for concentrating on this side of paint's function is that there is evidence that the mechanical durability of modern paints has improved faster than the durability of their appearance. It has been said many times that repainting is more often occasioned by "loss of face" than by loss of integrity. A main purpose of this paper is to explore what is meant by loss of face and to suggest ways by which the process can be controlled. Paradoxically, every new success in controlling deterioration of paint surfaces focuses public attention on the remaining defects. Thus, at one time it was almost axiomatic that paints in industrial areas would become almost black within a year or two, so that colour fastness of pigments was unimportant. Today, following clean air legislation and the elimination of pigments subject to sulphide staining, even in the worst areas large colour changes, particularly in hue, are totally unacceptable so that much more careful selection of pigments is necessary. Again, when high initial gloss became generally available in building paints in the immediate post-war period, complaints began to arise about hazing, crystalline bloom, and other gloss defects that developed in the first few weeks after painting, but had scarcely been noticed in earlier years.

Modern paints are already amongst the most durable of materials weight for weight, whilst some of the newer coatings just coming into use, such as the fluorinated vinyl polymers, show even greater resistance to breakdown. Table 1 compares erosion rates for some typical paints with corrosion figures

Table 1
Durability of materials

Corrosion rates of metals (in $\times 10^{-3}/\text{yr}$)

Mild steel ³		
Khartoum	(very dry)	0.1
Savoy	(mountain air)	0.5
UK	(rural)	1
UK	(industrial)	5
Nigeria	(surf beach)	37
Zinc (galvanised steel)		
UK	(rural)	0.1
UK	(Industrial)	1

Erosion rates of paint coatings (in $\times 10^{-3}/\text{yr}$)^{2*}

Florida exposure		
Oil base house paint	0.2
Acrylic emulsion house paint	0.15
Amine—alkyd coil coating	0.05
Vinyl organosol coil coating	0.025
Fluorocarbon organosol coil coating		0.0025

*Calculated by approximation from published data.

for metals. The superiority of the best paints over iron and zinc even in clean rural areas is evident; in industrial areas the balance swings even more in favour of paint as an intrinsically durable material. On the basis of the erosion

figures quoted for the fluorinated polymers, a life to half loss of a 20 micron film can be conservatively estimated at over 50 years. But this long life will only be achieved if the coating continues to adhere to its substrate and if the appearance remains acceptable; integrity of the film is not enough, both the interface and the surface must also be maintained.

Causes of surface change

The appearance of a paint film can be affected by external factors and by those arising internally. An obvious external factor is soiling by airborne dirt, others include contamination by fungi and algae and discoloration by atmospheric pollutants. The phenomenon of crystalline bloom, which has been closely studied by the Paint Research Association, involves condensation of the elements of ammonium sulphate from the atmosphere on the paint surface.

Dirt collection

Dirt collection on paint films is a perennial problem which has not yet received the scientific study it merits. Despite many suggestions in the literature that airborne particles are deposited on paint films by electrostatic processes, there seems little sound evidence for this view except for special situations near high voltage equipment. There is evidence, from the distribution of soiling and from controlled experiments, that a more likely process is thermal drift, that is, the bias in the random movements of small airborne particles towards cooler areas. Movement by the thermal drift process is very slow, being only 0.01mm sec^{-1} for a $1\mu\text{m}$ particle in a gradient of 1°mm^{-1} at 300°K ⁴ but it can, nevertheless, lead to serious soiling in a few weeks where a warm current of dusty air rises across a cooler surface, such as a wall over a lamp or radiator. Clearly, the movement of particles is unlikely to be affected by paint composition, except in so far as thermal conductivity of the film influences surface temperature. Moreover, it is doubtful if the accretion of particles on the surface by this process is a function of the composition, because the weight of each particle is so small that it can be held, even on a vertical surface, by extremely small forces. However, ease of removal of the soil does depend very largely on other film properties. Thus some gloss paints, for several weeks after application, remain soft and slightly tacky, particularly at elevated temperatures or under high humidity, and tend to fix dirt on their surfaces. Emulsion paints, when based on some of the softer polymers or heavily plasticised, show similar characteristics. For these paints the position of the glass transition temperature relative to the operating conditions is a useful index of likely soiling; paints based on polymers with a T_g below 0°C can be expected to retain dirt unless pigmented to a freely chalking condition.

Work by the Paint Research Association has shown that the retentivity of soils depends considerably on their nature. The tarry or greasy particles encountered in industrial areas penetrate more readily into organic films and are more difficult to remove than the more hydrophilic particles deposited in rural areas. Additions of small proportions of silicone oils to paints reduced dirt retention during weathering in rural areas but had little effect in industrial areas.

Dirt collection on paint films can be tolerated to a surprising degree if it is uniform. Uneven deposition caused by uneven surface temperatures quickly

becomes apparent, especially on surfaces of uniform solid colour. Much can be done in building design to reduce temperature differentials, particularly now that cheap foam insulation is available. From the paint side, there has been widespread development of multicolour finishes which, by breaking up the uniformity of surface appearance, effectively camouflage small amounts of dirt. Experience has shown that many users are convinced that the multicolour finishes actually collect less dirt, but no conclusive evidence or convincing theoretical reason for such a difference has been advanced. Nevertheless, with the present labour costs, the resulting ability to preserve an acceptable appearance for longer intervals before washing down is valuable.

Chemical discoloration

Colour changes of paints due to the effects of chemical pollution are many and various. Both pigment and media may be affected.

Media normally yellow, either temporarily or permanently. The problem of reversible yellowing of paints based on oily media has existed at least since the time of Rubens, who instructed his agent to put his paintings out in the sun at regular intervals to restore the pristine colours.

This type of yellowing is shown by all media containing unsaturated aliphatic chains, including oils, oleoresinous varnishes, alkyds and polyamides, its severity increasing generally with degree of unsaturation and being especially severe for oils with triene unsaturation. Work by the Paint Research Association⁵ showed that the yellowing was associated with changes during oxidative drying but that it was greatly intensified in the presence of ammonia or other nitrogen bases. Characteristic infra-red absorption spectra showed that the yellowing was associated with infra-red absorption in the region 6.1-6.7 μm and that these absorptions decreased on bleaching. It was postulated that the yellowing was due to chromophoric polymers formed from substituted pyrroles which in turn were formed by reaction between ammonia and 1,4-diketones. Bleaching by sunlight may be due to photo-oxidation of double bonds. In later work, it was found that although near ultra-violet radiation (about 370nm) bleached yellowed films, shorter wavelength radiation (about 300nm) causes intensified yellowing of alkyd resin films.

Yellowing by tobacco smoke is a severe problem in confined areas such as buses or public house bars. The effect most probably results from chemical activity of the deposited tar owing to its organic base content.

The yellowing of oily media is largely a problem in conditions of low illumination and, as with dirt collection, the differential effects are often more troublesome than the absolute change. That is, yellowing shows when a piece of furniture is moved from a painted wall or when new touch-up paint is applied. (The latter effect has been responsible for a major interest in the problem by the Navy⁶.) The effect can be minimised by careful choice of oils but, so far, no means for eliminating it completely, e.g. by the use of stabilisers or UV absorbers, has been found.

Some other types of media yellow on exposure to sunlight, as a result of the development of high levels of conjugated unsaturation. Chlorinated rubber and certain of the vinyls and polyurethanes are examples. Often the breakdown

processes are accelerated by the presence of the breakdown products, so that stabilisers that react with and neutralise these products can be effective in inhibiting the colour change.

Staining by atmospheric pollutants

Many of the colour changes on exposure investigated by the Paint Research Association are found to be due to local pollution. Complaints of this kind are becoming rarer, because the more sensitive pigments have been identified and are usually avoided. Historically, sulphide staining of lead pigments was probably the most common problem. Hydrogen sulphide is most frequently associated with bacterial action in certain muds or rotting vegetation, but it can also arise from industrial processes. The reaction with lead in paint appears to be sensitive to precise conditions, presumably permeability and moisture content of the films. In some instances, even the content of lead driers has been sufficient to cause marked grey stains but, more generally, reaction with pigments such as white lead, calcium plumbate or lead chromate is involved. The grey sulphide stain on a white paint can often be bleached by acidified hydrogen peroxide, although the process is usually slow.

Other pigments are affected by sulphuric and sulphurous acids, which are major contaminants in industrial atmospheres, possibly the most important effect being on lead chromes. As with sulphide staining, attack on lead chrome pigments varies considerably with conditions. It has been shown that, under cold humid conditions, such as in a foggy atmosphere, freshly dried paint films absorbed and concentrated sulphur dioxide from the atmosphere, the vapour being released and reacting with the pigment when the temperature rose. Under extreme conditions a Brunswick green paint can change colour to a strong blue in a few minutes. Much progress has been made in evolving acid resistant lead chromes and chrome greens and considerable improvement is possible by incorporation in water-resistant media.

Metal contaminants represent a more localised pollution problem. Copper in extremely small concentrations can cause staining by reaction with some pigments, a source of considerable trouble in artificial weathering apparatus where as little as 1 part in 10^7 of copper in spray water has led to brown staining. The occasional spotty darkening of bright red paints based on cadmium sulpho-selenide pigments, which are highly light-fast, has been associated with traces of copper in rainwater. The dripping of copper-containing water from telephone or electricity wires or from architectural copper also causes stains on paint, ranging from green to reddish brown or black, according to the composition of the paint. Addition of a copper chelating agent, such as benzotriazole, is an effective preventative for most of these troubles. Iron in solution is another source of discoloration, particularly when basic pigments such as calcium carbonate or calcium plumbate are present to accelerate precipitation of ferrous ions from the usual acid solution. Iron staining can generally be traced to adjacent underlying corroding metal. Its incidence can be reduced by careful formulation but in so far as an iron stain is a sign of corrosion which may ultimately become structurally dangerous, susceptibility to iron staining may be advantageous. Certainly, at the opposite extreme a straight red iron oxide paint can conceal rusting, dangerously, from cursory inspection.

This review of the effects of external contaminants has by no means exhausted the possible sources of discoloration. Many peculiar cases have been investigated at the Paint Research Association. One of the strangest, reported many times over a period of years, was the inexplicable development of a light green stain, often as a surface bloom on white or light coloured paints⁷. This stain, which has the properties of an indicator, showing a marked acid/alkali colour change to bluish purple, was finally identified as the dye emeraldine, which is formed by oxidation of aniline. The source of the aniline has not always been easy to trace, but in several cases evaporation from open bottles of clothes marking ink appeared likely; certainly the stain can be reproduced by exposing freshly dried oil-containing paints to aniline vapour.

Microbiological effects

Reference has already been made to loss of appearance owing to accumulation of airborne dirt. However, contamination of painted surfaces by micro-organisms can lead to more rapid and less acceptable changes in appearance. A wide range of organisms is involved, ranging from bacteria through fungi and algae to the lichens, which are organised symbiotic growths of algae and fungi and can build up into large geometrical structures. Most of these organisms require little nutrient other than water, oxygen and carbon dioxide, so that they can thrive on almost any surface that is not actively hostile because of readily extractable poisons. In the absence of toxic materials, rates of growth are mainly conditioned by temperature, humidity and, for many species of algae, light. Whiteley⁸ has shown that, under tropical conditions, building design has major effects on microbiological contamination. Wherever rainwater splashes on, or drips down, a painted wall, algae are likely to grow. In poorly ventilated, damp buildings, fungi thrive. The architect, then, can go far towards eliminating microbiological problems from most building paints, but there is a residue of situations, particularly in industrial establishments such as breweries, where problems remain, and fungicides or algicides must be used. Unfortunately, many of the more effective biocides are toxic to man, so that their use in, for example, food or drink factories must be carefully controlled. An important project on the current Paint RA research programme involves the screening of large numbers of potential biocides to select new materials which are effective at very low concentrations and are innocuous to man. Compounds are being tested against a wide range of fungi isolated from actual discoloured paint films. It is already clear that most active compounds are specific against certain fungi and that tests based on only two or three species could be highly misleading. First screening tests are being made against 16 fungal species, the test compounds being dispersed in an agar nutrient. Laboratory test methods have also been developed, and proved by comparison with exposure trials, for assessment of paints containing compounds which pass the initial sorting test⁹.

Microbiological contamination is mainly a superficial defect, although some of the growths can develop to dimensions much greater than the paint film thickness. There is little evidence that growth over the surface breaks into the film structure, apart from some conjectures on the attack on polyurethane films, although growth on infected timber or building boards underneath a paint film can disrupt the film. However, deterioration of a highly infected film is

likely to be accelerated by the moisture held by the growth and staining may result from dyes developed by the micro-organisms.

Crystalline bloom

One type of surface deterioration that seems mainly to be a problem in Britain is the phenomenon of crystalline bloom¹⁰. Crystalline bloom is a scatter of fine crystals of ammonium sulphate which can develop rapidly on the surface of a glossy paint film forming a veiling film that can be readily wiped away. It has been shown that as little as $4\mu\text{g cm}^{-2}$ of crystals is readily visible and that exceptionally high levels of atmospheric pollution are not required for the bloom to form. The necessary reactants, ammonia and sulphur dioxide, appear to be absorbed from the atmosphere by droplets of water condensed on the paint surface, since the effect can be reproduced easily in the laboratory by exposure to these vapours under condensation conditions. Crystalline bloom is associated, especially, with high quality, water-resistant media, probably because initiation of the growth depends on the surface having a high contact angle to water so that dropwise condensation occurs. For some considerable time the bloom is superficial and can be washed or wiped away without damage to the film, but electron microscope studies have shown that the continual cycles of solution and recrystallisation occurring with change in atmospheric humidity eventually result in penetration of the salt into the surface and permanent damage to the film.

Complete elimination of crystalline bloom is almost impossible, for it occurs to some degree on all surfaces, including glass, but it can be reduced by using more water absorptive paints or by treating the paint surface with solutions of surface active agents to decrease the contact angle.

Internal changes in films

Most processes of solid film formation from liquid paints involve significant changes in position of film components. Thus, a highly pigmented film may be first applied as a uniform wet film with a glossy appearance but, as the solvent evaporates, the pigment particles settle unevenly leaving a rough surface with a matt appearance. A change of face of this type may be desired, but often slow changes in the film at later stages may give rise to undesirable loss of gloss, often termed hazing. Oxidative drying media show considerable dimensional changes during initial film formation, and these continue indefinitely at a reducing rate. Thus, film volume measurements by a mercury displacement method showed a shrinkage of 6 per cent during six weeks' ageing at 25°C in the laboratory, for unpigmented and rutile titanium dioxide pigmented alkyd films^{11, 12, 13}. When the pigment is poorly dispersed, so that concentration varies greatly in different parts of the film, a shrinkage of this order can result in a marked deterioration in gloss.

Work carried out for the International Lead Zinc Research Organisation showed, however, that, in the presence of reactive pigments such as zinc oxide, gloss changes greater than those for rutile pigmented films could occur, owing to redistribution of the film material. For zinc oxide pigmented alkyd films the overall volume change was in fact very small. It was concluded, from electron microscope studies, infra-red and analytical work, that the shrinkage, in the

absence of reactive pigment, was associated with oxidative scission, which gave rise to volatile acidic products such as formic acid. When zinc oxide pigment was present, a proportion of the acids reacted with it, forming a shell rich in zinc soaps around each particle. The net effect was shrinkage of the medium between pigment particles with a build-up around the particles. When the particles were sufficiently large, or were dispersed in clumps rather than individually, gloss deteriorated rapidly, giving the so-called zinc oxide hazing. Similar effects are shown by other basic pigments, notably red lead, but are less important in practice, because these other pigments are rarely used in finishing paints. Zinc oxide hazing can be greatly reduced if a proportion of a well dispersed fine pigment, colloidal zinc oxide, is used. Another method which proved successful in reducing loss of gloss was the addition, at the paint making stage, of 2-4 per cent of water, calculated on the pigment content. The improvement was probably due in part to improved pigment dispersion resulting from the zinc soaps formed in the wet paint, but it is also likely that the soaps formed around zinc oxide particles, from reaction with relatively high molecular weight molecules in the bulk paint, subsequently restricted reaction in the dried film.

Weathering effects

The most frequent change in paint films during weathering is the erosion of surface layers, leading to development of a loose, chalky layer of pigment. Where repainting has ultimately taken place, chalking is the most desirable form of breakdown, because removal of the loose chalk provides a good surface for recoating. But for the many situations where the initial coating can be expected to last the life of its substrate, as on motor cars, chalking must be avoided, and for most other situations it should be delayed or controlled.

The chalking process normally starts with scission of molecular fragments from the polymeric binder, resulting from photoactivation. Because the energy quanta required to initiate most molecular breakdowns are large, activation is mainly by ultraviolet radiation. In solar radiation, the effective wavelength range is 300-400nm. Resins that do not absorb at these wavelengths are virtually immune to degradation by ultraviolet radiation when unpigmented. Such materials, for example polyvinyl fluoride, form the basis for coatings with a very long life. However, negligible absorption by the resin is not the only requirement, for some oxide pigments may activate breakdown by absorbing ultraviolet energy and oxidising surrounding molecules. The interaction between radiation, pigment and medium is still imperfectly understood but, fortunately, empirical study has resulted in coating techniques for titanium dioxide pigments that are effective in inhibiting breakdown over a very long period.

Chalking of white paints does not necessarily detract from appearance other than by loss of gloss; indeed by releasing engrained dirt it may help to restore appearance. When a coloured paint chalks, however, there is usually an apparent fading, because the pigment particles released from the binder scatter more light from their surfaces than before. The colour change is greatest for mixtures of white pigments with small proportions of strongly absorbing coloured pigments and is particularly obtrusive when the chalk has been wiped

or washed from parts of the painted surface. The colour change on chalking is much reduced when pale, self-coloured pigments are used in place of a mixture of white and colour. For this reason the development of long-life coatings may well lead to a revival of interest in some of the lower colour strength inorganic pigments and the development of new ones to replace the strong organic colours that are widely used in present paints. Indeed, the pigmentation of truly long-life paints may well become more similar to that of vitreous enamels or coloured glasses than to that of present day paints and plastics.

Fading of pigments

The stability of pigments to light is another important factor in the maintenance of appearance. Most organic and some inorganic pigments undergo changes, usually through oxidation for the organics and through developments of structural defects or structure changes for the inorganics. There has been much research to develop pigments of improved light fastness and to reduce fading by formulation changes, although detailed study of fading mechanisms does not appear to have been pursued as far as might have been expected. Possibly, a degree of light fastness sufficient for present day paint lives of a few years has not been too difficult to achieve by empirical selection—except for the more saturated reds, purples and violets. When 20 or 30-year lives become more general, the properties of many existing pigments will become inadequate. Again inorganic pigments may be the answer, but the pigment industry should already be developing the new generation of materials that will be required, or investigating new ways of improving existing materials.

Assessment of appearance changes

All study of change requires scales for recording and tools for measuring. Glossmeters have been developed to high sensitivity for the assessment of changes in highly glossy surfaces. Measurement of specular reflection intensity, as in the BS 3900 and ASTM D 523 methods, suffices for most gloss paints, although more refined methods have been adopted to study the very small scale surface defects that affect the eye only as a slight blurring of images reflected in the surface. For lower gloss levels, specification in figures is more difficult, because surface structures may be complex, resulting in reflected light distributions that are complicated functions of angles of illumination and viewing. Colour change can also pose measurement problems because colour is also dependent on the geometry of illumination and viewing. This dependence is obvious for paints containing metallic pigments or paints which have "bronzed" by developing selective orientations of strongly absorbing pigment particles, but is also of importance when paints of different degrees of gloss are compared. Thus, under directional illumination a paint that has merely lost gloss without any change in the pigment may appear to have faded. Indeed, gloss and colour changes are inseparable, in some degree a conclusion which complicates measurement and tolerance specification considerably.

A new colorimeter devised at the Paint RA in the last 18 months^{14, 15, 16}, makes possible more accurate measurements of the colours of surfaces and more flexible variation of illumination and viewing geometry. In this instrument

the viewing head, containing illuminating and viewing elements, is separated from the body of the colorimeter, being connected solely by fibre optics light guides. The design of the new colorimeter allows a variety of viewing heads to be used, with areas of illumination varying from, say, 1mm², required for examination of a single brush stroke on a painting, up to 100cm², required for assessment of the colour of a multicolour paint film.

One of the biggest problems will be to assess the stability of appearance of coatings intended for very long-life applications. Clearly, natural exposure tests of more than a few years' duration are impracticable. On the other hand, accelerated tests may well miss some factor that may be significant over a very long period; for example, the small amount of solar radiation below 300nm, integrated over 20 years, could produce changes in paints that were fully stable to longer wavelength radiation. The solution already suggested² for assessment of bulk film stability, is to make very careful measurements of weight change over two or three years which can be extrapolated to much longer periods. To apply the same techniques to colour changes involves accuracy at the limit of current apparatus, because sensitivity to as little as 0.1 NBS units of colour difference is required if changes of, say, 2 NBS units are to be predicted from measurements after only one-tenth of the full life exposure.

Interpretation of colour changes

Measurement of colour changes of paint films involves problems of gloss and texture changes and arguments as to degrees of cleaning which may be acceptable before measurements are made. Before a guarantee of colour stability can be meaningful, these points must be resolved by laying down an agreed procedure for measurement. But even when measurements have been made, there remain problems of interpreting the scale of colour change. The CIE system of colour measurement was an attempt to set up an objective scale which might be correlated with visual impressions. Because the system was based on linear scales, whereas sensory impressions are more nearly logarithmic, changes in CIE tristimulus values could not be expected to correspond in relative magnitudes to visual differences. Moreover, it was soon found that the scaling of visual differences varied widely in different parts of the colour field, as mapped by CIE values.

Numerous attempts have been made to convert CIE values into a uniform chromaticity difference field by geometrical projection and algebraical transformations. The development of these approaches and their application to the setting of paint colour tolerances was reviewed in a series of articles by Hemmendinger¹⁷, Davidson¹⁸ and Johnson¹⁹. Work by Davidson and others has shown that none of the transformations is entirely satisfactory, especially when large colour differences are involved, but equally it has been shown that visual judgements of colour differences are highly erratic and only become meaningful when the opinions of a panel of observers are pooled. Indeed, given an intelligent choice of sample preparation technique and colour measuring apparatus, an instrumental colour difference measurement will be much more reliable than an individual visual estimate. But it is unwise to set blanket limits of colour difference tolerances for a wide range of colours.

Tolerance limits should be set, whenever possible, by consideration of measured figures on a large number of samples that have been graded visually by observers experienced in the appropriate field of use. In this way it is possible not only to counteract errors arising from defective measurement and calculation procedures but also to take account of technological biases. For example, a change in lightness or saturation is often more acceptable than a change in hue, because on a practical paint surface one is likely to see uncontrolled variations in lightness and saturation, due to film thickness difference or differences in intensity of illumination on different facets or areas of a surface.

Conclusion

This paper has reviewed some of the ways in which paint films may change in appearance and has suggested how some of these changes may be controlled. Clearly, the face of paint is at risk at every stage from initial film formation onwards. Some of the causes of change are inbuilt and can be controlled or completely eliminated by better formulation and better paint making. Other causes of deterioration, such as soiling and microbiological contamination, are mainly outside the control of the paint manufacturer. He can, however, develop paints that are more easily cleaned and that show fungistatic if not fungicidal qualities. Pressing the analogy of the changing face of paint to the human face, may there not also be scope for paint cosmetics, e.g. special treatments to remove dirt and other surface contaminants with minimum effort? Again, because most of the change in modern paint films is entirely superficial, might it not pay to apply very thick coatings initially and to renew the appearance, after weathering, by removal of a thin layer rather than by repainting, with all the hazards of that operation under site conditions?

Nevertheless, it is believed that, eventually, developments in polymers and pigments will enable coatings to be made of any colour, and stable for very long periods—at a price. To achieve this aim will involve not only new long-life media but almost certainly new pigments. Perhaps the most difficult problem will be to develop test methods adequate for assessing long-life materials.

How far the customer will be prepared to pay heavily for long term stability remains to be seen. Indeed in the long term is an unchanging paint face desirable? Perhaps the ability to grow old gracefully may be preferred to a constant appearance of youth. Certainly the corrosion patina on a copper roof or the weathered appearance of cedar shingles or Cotswold stone is prized above the original appearance of these surfaces. Should the paint industry be considering a new concept of controlled weathering paints for external architectural use?

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Appearance and performance factors in coatings for buildings*†

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Summary

A discussion of the appearance and performance factors in exterior surface coatings for buildings, and criteria for evaluating them, is followed by a description of exposure tests of paints for masonry surfaces, ranging from emulsion- and solution-based thin films through imitation stone paints to thick textured coatings. The appearance, durability, and control of moisture movement are assessed to give a predicted life from which a tentative cost-in-use comparison is made. The thicker films give the best overall performance but are not necessarily cheaper in the long term. Generalisations are difficult and the differing combinations of properties require a choice to be made to suit the specific needs of a building or a user.

The extensibility of the coatings, measured by two methods, is shown to affect the behaviour, but must be considered in conjunction with film thickness and permeability in any attempt to forecast performance.

Keywords

Types and classes of coating

masonry finish

Types and classes of surface

concrete

Fletton brick

masonry

Properties characteristics and conditions

primarily associated with dried or cured films

durability

extensibility

permeability

Les facteurs qui influent l'apparence et le rendement des peintures pour bâtiments

Résumé

On discute les facteurs qui influent l'apparence et le rendement des revêtements appliqués aux surfaces extérieures de bâtiments, et également les critères pour les évaluer. Ensuite on décrit des essais de vieillissement des peintures, destinées aux surfaces de maçonnerie, effectués sur des feuil minces de peintures-emulsions, de peintures conventionnelles, et aussi de peintures à l'imitation de pierre, et d'ailleurs sur des couches épaisses de revêtements texturés. On apprécie l'apparence, la durabilité et la contrainte de l'accès de l'humidité afin de donner une vie ultérieure prédite à partir de laquelle on peut faire une comparaison tentative du coût de l'emploi. Les couches plus épaisses donnent les meilleurs rendements globaux, mais elles ne sont pas nécessairement plus économique à la longue. Il est difficile de faire des généralisations, et les diverses combinaisons imposent que l'on fait un choix en accord avec les besoins spécifiques d'un bâtiment ou d'un client.

On démontre que l'extensibilité de revêtements mesurées par deux méthodes influe leur comportement mais elle doit être considérée avec l'épaisseur et la perméabilité de feuil lors des tentatifs de donner des prévisions au point de vue du rendement.

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Aussehen und Leistung Betreffende Gesichtspunkte bei Bautenfarben

Zusammenfassung

Nach einer Besprechung der das Aussehen und die Leistung betreffenden Faktoren bei Aussenanstrichmitteln für Bauten, sowie der Kriterien für deren Bewertung folgt eine Beschreibung der Bewitterungsprüfungen für Anstrichmittel von Mauerwerk. Letztere umfassen dünne Filme von Farben auf Emulsions- und Lösungsmittelbasis, Imitationssteinfarben sowie dicke texturierte Anstrichmittel. Aussehen, Wetterbeständigkeit und Kontrolle der Feuchtigkeitsbewegung werden bewertet, um eine Lebensdauervoraussage machen zu können, mit deren Hilfe ein provisorischer Kostenvergleich für praktische Anwendung möglich ist. Die dickeren Filme verhalten sich im grossen und ganzen am besten, sind aber auf lange Sicht hin nicht notwendigerweise billiger. Man kann nur mit Schwierigkeiten verallgemeinern, und die verschiedenen Kombinationen von Eigenschaften machen eine für die besonderen Erfordernisse eines Gebäudes oder Verbrauchers geeignete Wahl nötig.

Die Dehnbarkeit der Anstrichmittel, auf zweierlei Weise gemessen, beeinflusst, wie gezeigt wird, deren Verhalten; wenn man versucht die Haltbarkeit vorauszusagen, muss Dehnbarkeit aber in Verbindung mit Filmdicke und Durchlässigkeit betrachtet werden.

Наружный вид и качественные характеристики покрытий для зданий

Резюме

Обсуждение наружного вида и качественных характеристик наружных поверхностных покрытий для зданий и критериев для их оценки с последующим описанием испытаний на выветривание красок для кирпичных поверхностей—от эмульсионных и растворных тонких пленок и красок имитирующих каменную поверхность до покрытий густой текстуры. Оцениваются наружный вид, прочность и контроль движения влаги для прогноза долговечности, из которого делается предварительное сравнение стоимости эксплуатации. Более густые пленки дают наилучшие общие эксплуатационные качества, но они не являются обязательно более дешевыми в конечном расчете. Обобщения предъявляют трудности и различные комбинации свойств требуют выбора для удовлетворения специальных нужд здания или потребителя.

Растяжимость покрытий, измеренная двумя методами, влияет на поведение, но должна рассматриваться в связи с толщиной пленки и проницаемостью в прогнозе эксплуатационных качеств.

Part I—General considerations

Of the total 1967 expenditure on building of £4,500m (Ministry of Public Building and Works figures), about £1,550m was for maintenance work, contractors and public authorities accounting for £1,190m, of which £375m was for painting and decorating. There could be no clear division between decorative and protective painting, but both requirements offer scope for economies. Site painting is difficult to rationalise, but improvements in efficiency and speed have been made, notably with newer types of coatings and techniques of application. Probably a better way for future reduction of maintenance expenditure lies in the use of coatings applied in the factory to sheets and other components. These can take advantage of more durable high-polymer systems unsuitable for site work, examples being pvc and pvf coatings, stoved finishes etc. There is also bound to be an increase in the use of plastics components which need no painting, for a long period at least.

Although greater durability may be accepted in general for many new coatings, evaluation of their performance and prediction of durability are still necessary, yet are becoming more difficult to make, and some indication of possible

methods has been given in a previous paper¹. There is a further need for studies of their cost in use, based on predictions of their durability. This paper deals mainly with coatings for exterior wall finishes, including some of the less tractable types with coarse textures, which receive little attention in the literature. A comparison of costs is attempted, to demonstrate a suitable method rather than to provide a definitive yardstick.

Appearance and performance requirements

The definition of "life" is necessarily partly subjective; if the coating is mainly protective it can be defined by the eventual failure to protect, eg the entry of water or the appearance of cracks or flaking, but if decorative, the extent of colour change, chalking, erosion, dirt retention and mould growth could determine the need for repainting, and the subjective judgement of these would vary widely between observers. Any failure of integrity (cracking and flaking) must be a more definite indication of need for renewal. Until fairly recently, the general loss of appearance and integrity ran roughly parallel for outdoor exposure, and repainting was commonly needed on both counts at about the same time. With increasing durability of polymer-based coatings, permitting ten years and more before recoating, there is a greater need for retention of the original appearance. Repainting of a perfectly sound film purely for the sake of appearance is going to seem highly uneconomic. For example, some early pvc coatings on metal have shown large changes in certain colours in only two to three years' exposure or the equivalent in artificial weathering, whereas the film has remained entirely sound for very much longer.

Better light-fastness of pigments, non-yellowing media, dirt and mould resistance are thus seen to be important requirements. It is, however, somewhat curious that many traditional building materials—bricks, tiles, stone, slates, wood, copper—are expected to change in appearance on weathering, and with few exceptions the changes are thought to be pleasing or even desirable, whereas newer, "artificial" products are expected to maintain their original appearance for long periods.

Criteria for appearance factors are difficult to establish. Many publications concerned with deterioration of paint films emphasise loss of gloss, a change which can be fairly readily measured, but it is not a property which gives rise to complaints in the building field. Colour change is much more important, and definition of colour by various systems and hence of colour differences in NBS units has become a feature of certain types of coating in the USA, especially coil coatings and film laminates, for which guarantees in various forms are given. Colour differences in numerical form are useful for definition, but for their subjective appreciation need a large series of colour specimens in different hues and directions. The grey scale is not an entirely adequate substitute for the differences of hue, in which changes are probably less acceptable than even larger changes in value or chroma.

One possible approach that has been suggested by the BRS is the use of the Munsell colour atlas. This is frequently used for colour choice, especially by architects, and one could reasonably assume that if any one colour were selected, all those adjacent to it in any direction of hue, value or chroma were rejected. Hence a change by a step as large as any one in the atlas, over a stated number of

No.	Type	No. of coats (2)	Maker's rec'd or normal yd ² /gl	Coverage			Actual (dry) total g/m ²	Price (1970) Retail £/gl (or per lb)	Trade or bulk	Retail
				Actual (wet) per coat yd ² /gl	Actual (wet) total g/m ²	Actual (dry) total g/m ²				
1	Copolymer emulsion paint	2	60-80	62	237	127	3.15	1.50	12.00	
2	Acrylic emulsion paint	2	100	77	210	115	3.15	2.00	9.00	
3	Chlorinated rubber paint	2	40-60	52	349	194	4.00	2.50	18.00	
4	Vinyl-toluene acrylic solution paint	2	40-60	46	319	214	3.00	1.70	15.00	
5	Vinyl-isobutyl ether solution paint	2	40-60	39	322	149	2.95	2.20	18.00	
6	Oleo-resinous stone paint	2	7.5	21	777	578	1.4	1.10	16.00	
7	Oil emulsion stone paint	2	16	20	890	698	0.88	0.80	17.00	
8	Oleo-resinous stone paint	2	(1 per lb) 30-35	36	540	452	3.60	2.50	24.00	
9	Copolymer emulsion (sanded)	2	35	39	397	254	3.05	2.70	18.00	
10	Cement-based paint	2	1-1.5	4	—	244	0.05	0.00	2.00	
11	Exterior textured paint (9)	1	(per lb) 5.5	(per lb) 6 (3)	1000	690	—	1.00	(per lb) —	
12	Copolymer emulsion (sanded)	1	10-30	17	536	407	3.28	2.50	23.00	
13	Copolymer emulsion, mineral aggregate	2	0.5/lb	—	1950	1640	0.11	—	67.00	
				(3)			(per lb) 0.09			
14	Exterior textured paint (9)	1	0.2/lb	—	1545	1040	(per lb) —			
15	"Cocoon" type emulsion (1)	2	—	—	1500	—	—	—	51.00	

- NOTES: (1) No longer on market
 (2) Excluding sealers
 (3) Sprayed (others brushed); applied weight estimated from 300 × 300mm "coupon" sprayed similarly
 (4) By roller
 (5) Estimated if materials cost same as No. 11
 (6) London measured rate, July 1970, 2 coats emulsion paint on plaster 5.4 s/m² = 27 p/m²
 (7) " " " " " " oil " " " 6.75 " 34 "
 (8) " " " " " " rendering in cement 15.2 " 76 "
 (9) Ten year guarantee
 (10) If life is 4 years, cost becomes 91 p/m²

years, could be said to be unacceptable. The steps are not uniform throughout the atlas, but this may not be an insuperable objection. There is a need for some criterion of acceptable change, varying for different end uses, to be laid down, perhaps by a panel of observers, and this need may become as important as that for definition of colour match tolerances in the original coating.

Large but uniform changes will generally be more acceptable than patchiness. Thicker coatings are then more likely to be satisfactory, since their erosion takes longer, and factory coatings should give better results than the somewhat uneven thickness resulting from site application; on the other hand, factory production requires very close control of colour uniformity from batch to batch, particularly where large areas are to be clad with coated sheets. Variable fading is an uncommon cause of complaint from site-applied paints.

Dirt retention is an interesting question with an obvious dependence on film softness and texture, but with other factors, such as electrostatic properties, not quite so obviously defined. To some extent, it can be included in the colour change measurements, but there is a subtle subjective differentiation that makes darkening by dirt more objectionable. Whereas slow chalking can be used to keep white coatings clean, it is not really desirable, since it results in a greater erosion rate than non-chalking coatings would have, it tends to cause

Table
cost in use

Cost (Material)		Time Application only 2yd ²	Cost at 90p per hr	Total Cost (comparative)			Present value of maintenance over 50 years (life from table 2) present value at 8% interest p/m ² (retail)	From quoted cost	Total cost-in-use over 50 years (1st treatment and maintenance)
Retail	Trade or bulk			Retail	Trade	Quoted			
p/m ²	p/m ²	min sec	p/m ²	p/m ²	p/m ²	p/m ²	p/m ²		
12.2	7.3	24 05	21.5	33.7	28.8	Note (6)	126 (10)	160	
9.8	6.3	21 13	19	28.8	25.3	"	78	107	
18.6	13.5	24 19	21.5	40.1	35.0	Note (7)	66	106	
15.7	14.1	26 20	23.5	39.2	37.6	"	65	104	
18.3	16.9	25 57	23	41.3	39.9	"	69	110	
16.1	12.9	28 45	25.5	41.6	38.4	"	57	99	
17.2	15.5	24 25	21.5	38.7	37.0	"	64	103	
24.1	19.4	24 40	22	46.1	41.4	Note (11)	63	109	
18.5	16.5	23 30	21	39.5	37.5	"	54	94	
2.7	2.4	14 51	13	15.7	15.4	"	42-26	58-42	
(per lb)									
—	33.0	6 30 (3)	5.5	—	38.5	107-150	44-32	83-71*	
23.0	20.5	12 20	11	34.0	31.5	"	39	73	
67.5	—	20 10 (3)	18	85.5	—	210	71	157†	
51 (5)	—	1 40 (3)	1.5	52.5	—	Note (8)	43	96‡	
—	—	13 00 (4)	11.5	—	—	120-175	—	—	

† Quoted for high buildings 86 p/m²

‡ Total cost-in-use from quoted cost: 232-325 p/m²

§ Total cost-in-use from quoted cost: 281 p/m²

¶ Total cost-in-use from quoted cost: 220-230 p/m²

a wash-down on to other surfaces, and for tints of white it generally results in an excessive colour change². A survey of wall coatings at the BRS has shown that chalking rates vary widely, and it seems that such coatings are not formulated differently for optimum performance in coastal and urban areas. Mould and algal growth are related to dirtiness of films and can detract from otherwise satisfactory appearance; they give rise to fairly frequent complaints, particularly in the wetter districts. Algae and lichens are susceptible to sulphur dioxide and are less common in polluted industrial atmospheres. The factors inhibiting growth are complex and not satisfactorily separated. Thus susceptible ingredients can be as important as the presence of toxic materials, which often have a short effective life; hardness of the film is difficult to separate from chemical constitution, and water-permeability plays a large part, affected by both polymer and pigments. Although films with high permeability might be expected to lead to wetness in the substrate and hence high growth, the ease with which water is lost through them may equally well cause low growth rates.

The film integrity also determines appearance, even if the coating does not have to provide protection or waterproofing, although with most modern gloss coatings failure by cracking and flaking is less common than formerly. It does

Table 2
Performance in exposure test

No.	Crazing on render (South)		Flaking from bricks (North)			Mould 0-4 3 yr	Algae 0-4 3 yr	Control of water content		Appearance ¹ order of merit		Estimated ² life to repaint Render Brick Years
	0-4, 0=no failure 1 1/2 yr 3yr 4yr	0=no failure 3yr 4yr	0-4, 1 1/2 yr 2yr	0=no failure 3yr 4yr	4			Up to 1yr	After 2yr	Render	Brick	
1	1	4	1	4	0	1	nil	10=	6	3	2	
2	1	3	0	3	0	1	nil	12	5	4	3	
3	0	1	2	3	0	1	nil	4=	11=	5	2	
4	0	2	1	2	0	0	nil	7=	10	5	2	
5	1	2	3	4	0	0	nil	4=	11=	5	2	
6	0	1	1	1	2	1	poor	—	—	7	4	
7	0	2	1	3	0	1	nil	9	4	6	3-4	
8	0	2	1	3	0	1	nil	3	3	7	4	
9	0	0	0	1	1	3	fair	4=	8	7	5	
10	3	3	0	1	0	4	poor	10=	9	4-6	5 ³	
11	0	0	0	1	1	0	poor	2	2	8-10	7	
12	0	0	0	2	0	0	poor	7=	7	8	5	
13	0	1	0	1	0	0	fair	—	—	10	10	
14	0	0	0	0	1	0	good	1	1	10	10	
15	0	0	0	1	2	0	good	—	—	10	5	

NOTES (1) Excluding non-white coatings; coatings 13 and 15 satisfactory retention of appearance but rather dirty, coating 6 very patchy.
 (2) Ignoring failure on two lowest courses.
 (3) Very dependent on appearance.
 (4) Film unbroken but not adhering to some bricks.

Table 3
Extensibility

No.	Film wt gm/m ²	Direct extensibility at rate (on aluminium)									Garston exp 6 weeks			BS 3,900 Pt F3 500 hrs			Extensibility in mortar block test			
		Control			4 weeks 70°C			500 mm/min ii— 50 mm/min iii— 5 mm/min			i—500 mm/min ii— 50 mm/min iii— 5 mm/min			i				ii		
1	175	22	25	—	10	16	—	24	27	—	8	17	—	0.025						
2	168	40+	40+	—	40+	40+	—	40+	40+	—	35+	35+	—	0.025						
3	28	3	9	20	—	—	—	—	—	—	3	6	8	0.0						
4	44	40+	40+	—	—	—	—	—	—	—	18	24	31	0.01						
5	24	40+	40+	—	—	—	—	—	—	—	11	15	18	0.01						
6	110	25	35+	—	9	15	—	11	15	—	8	9	—	0.08						
7	303	1	—	—	1	—	—	3	6	—	1	4	—	0.0						
8	245	3	7	9	1	3	4	40+	40+	cracked before test	35+	35+	5	0.05						
9	105	40+	40+	—	35+	35+	—	—	—	—	8	—	—	0.1*						
10		Not tested—too brittle																		
11	422	8	10	—	2	7	—	3	7	—	3	5	—	—						
12	298	40+	40+	—	40+	40+	—	40+	40+	—	35+	35+	—	0.25						
13	707	35+	35+	—	29	35	—	30	35+	—	18	22	—	—						
14	390	11	13	—	7	10	—	8	12	—	3	5	—	—						

*After 4 weeks exposure 0.05

however still affect many exterior wall coatings.

Coatings for concrete and masonry

The function of coatings for concrete, renderings, and masonry tends to be more for decorative purposes than protective, but many coatings are claimed to have a waterproofing action and a few, such as bitumen coatings, are purely protective. The life of such coatings varies from 2 or 3 years for some emulsion paints to over ten years for some very thick textured coatings, and film thickness plays as important a part as composition. Concrete, apart from its alkalinity, is regarded as stable, but loss of adhesion to it is common; renderings are generally less stable and develop hair cracks or crazing, the result of shrinkage caused by initial drying or subsequent carbonation, or of movement of the background. The authors' comparison of different types of finish has shown that such crazing becomes visible through thin film coatings long before thicker ones, and that the more permeable or brittle the coatings, the worse this defect shows. (See Tables 1, 2, and 3). The possibility of coatings having sufficient extensibility to cope with crazing and cracking of the substrate is discussed later.

A further detrimental effect can be the development of efflorescence or lime stain. Although faint, these effects are usually patchy and lead to poor appearance, but not necessarily to film failure. Resistance to the passage of salts, especially calcium ions, and to hydrolysis, is an important function of coatings for masonry surfaces, and is generally achieved by selection of suitable polymers, but other factors such as pigment volume cannot be neglected.^{3,4} This aspect has been studied at BRS both in exposure tests and in a rapid test of the coatings on mortar blocks in the alternating humidity of the corrosion cabinet. Complete resistance of coatings to passage of salts, particularly sodium sulphate, may result in a build-up beneath the film and its eventual disruption; this effect is, however, less commonly seen externally.

Methods of test

Many of the accepted laboratory test methods cannot be applied to coarsely pigmented or textured coatings. Comparative exposure testing on a suitable wall brings out differences between coatings very strikingly, but the standards required are difficult to define, whether for a specification or for a single evaluation as in an Agrément certificate. The large numbers of available coatings make comprehensive tests expensive, and costs are increased by the colour ranges, the need for replication and for tests on other than new surfaces (weathered surfaces being almost impossible to define and reproduce). There is further difficulty in that the relative order of merit tends to change year by year or even by season depending on the chalking rate and algal growth, or even lighting conditions or wetness of the surface, so that interim judgements may be misleading. This point has to be emphasised to visitors to BRS test walls, to avoid giving a wrong impression in the early years, but after three or four years' exposure there is a fairly positive and steady difference in the general appearance of coatings of widely different types. This is a long time to wait for results, but exterior wall coatings, thick film or textured, and especially those based on thermoplastic polymers, are less easily evaluated by

artificial weathering and short term tests than are the thin gloss paint types. Many of them survive in the BS 3,900 part F3 cycle for thousands of hours, with little visible change except darkening or a deposit. To produce breakdown or loss of adhesion it seems that a different approach is needed, possibly using an unstable substrate or incorporating the effects of frost, which are probably more important in practice with this type of coatings than previous work has shown them to be in the artificial weathering of gloss paints on metal. The authors have, however, been able to find large changes of extensibility in some of these coatings after periods of natural and artificial weathering too short to produce visible failures. Such changes have to be related to actual performance on various substrates over a longer term, in order to be of use for predictive purposes. Dirt retention is difficult to simulate artificially, but mould and algal resistance can be investigated using methods developed by the Paint Research Association which correlate well with natural tests.

Part II—Evaluation of exterior wall finishes at BRS

The authors have found exposure tests on asbestos cement panels, at 45° facing south, to be unreliable, especially in respect of dirt retention, blistering and flaking. After many years of tests on miniature Fletton brick walls, which cause rapid failure of most paints, a large test wall was built in 1966 for evaluation and demonstration purposes, its dimensions being $9.144 \times 1.524 \times 0.230\text{m}$, ie $30\text{ft} \times 5\text{ft} \times 9\text{in}$. This provided an area of 0.836m^2 (1sq yd) per side for each paint. The construction was of Flettons over two courses of engineering bricks on a concrete slab (with no damp proof course), rendered on the south side with the 1:1:6 cement:lime:sand mix used as mortar, and left bare on the north face (Figs. 1 and 2). This wall is clearly not subject to some of the temperature and moisture gradients of an occupied building. Table 1 shows the paints used.

In addition to the visual behaviour of the coatings on the test wall, shown in Table 2, their extensibility was investigated both by direct tensile tests on soft metal substrates¹ and by another method which overcomes objections to the different adhesion conditions that could exist between the chromated aluminium substrates used and the surface of cement-based materials. Periods of ageing by natural weathering, artificial weathering and heat alone have been used to determine the susceptibility of the binder to various influences.

Micro-indentation tests of hardness are impossible for coatings containing coarse particles, and if applied to the medium only, without the reinforcement and protective effect of pigment and coarse particles against UV radiation, could be misleading. This technique has not, therefore, been employed for this type of coating.

Results

The behaviour of paint on the Flettons is much worse than on the rendering and would have been even worse on Flettons facing south. It is also worse on the lower courses, which stay wet longer.

The estimated life shown in Table 2 combines both appearance and integrity, which differ between the rendering and the bare brick. Failures on the two lowest courses have been ignored in making this estimate, but are an indication

of the need to take special measures close to the ground or below damp-proof course level.

Appearance changes seasonally and yearly, and although reflectance measurements have been made they are not a good guide; also, not all the coatings could be obtained in a comparable good white colour. Of the appearance factors described earlier, loss of integrity is a major feature on the bricks (Fig. 4) and failure by crazing of the rendering also assumes importance. Dirt retention is less serious than mould and algal growth, and several coatings developed a patchy appearance partly through chalking and partly as a result of lime efflorescence. Retention of a good white appearance has been given considerable weight in the assessment of appearance in Table 2.

Algal growth occurs mainly on the north side and mainly at the foot of the wall, which is wetter, and there are large differences between coatings, the

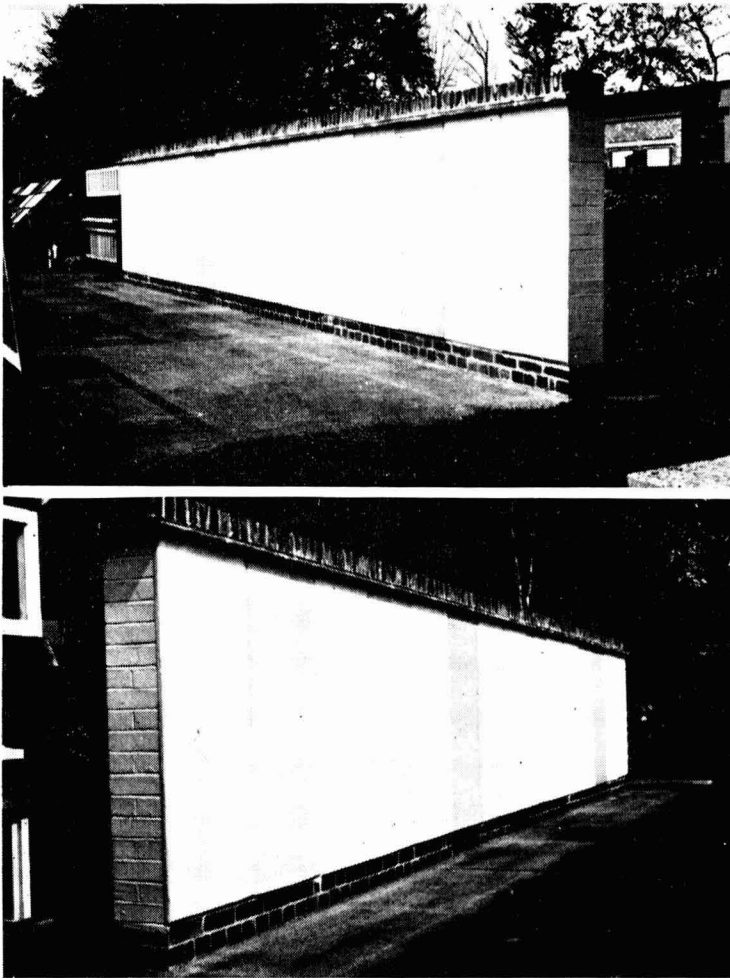


Fig. 1. Two views of the south aspect of the test wall at 2 years

cement-based paint and one emulsion textured paint being susceptible to algae, those with oil/resin binders being rather prone to mould. There was a characteristic growth of mould in a pattern of a run-down of spores from random points, with no mycelium, mainly on the south face. This growth affected the whitest and best looking finishes at one stage, but largely disappeared during the summer.

Permeability. The moisture movement in and out of a wall is an important feature of coatings, even the purely decorative ones, and many are claimed to

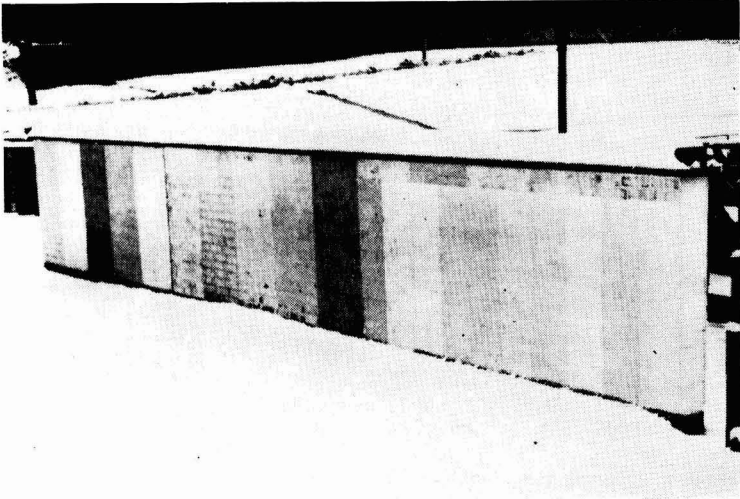


Fig. 2. Test wall, north aspect, at 3½ years

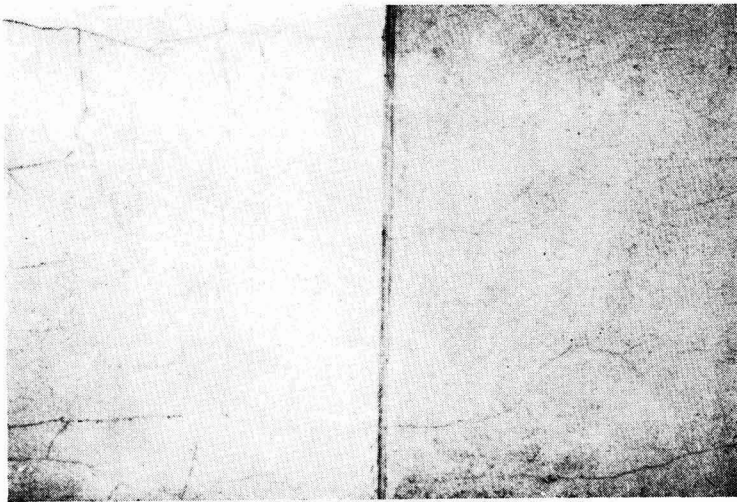


Fig. 3. Test wall, sections 1 and 2, south aspect showing crazing at 3½ years

be resistant to rain yet able to "breathe," i.e. to let vapour escape. The moisture permeability of coarse textured coatings is difficult to determine by standard methods, such as the Payne cup, and has, therefore, been determined by monitoring the water content of the free standing test wall using a microwave meter⁵ which gives a direct indication in-situ of the moisture content and hence indicates the permeability. The transmitter and receiver are placed on opposite sides of the wall and the attenuation of the signal is proportional to the mass of water between them; materials such as brick and concrete have very little effect, for which compensation can be made. Soluble salts can affect the absolute readings, but for repeated measurements at fixed points with little movement of salts, the meter can be considered fairly reliable. Even then, some erratic



readings are obtained, but the general trend is shown in Figs. 5 and 6. The foot of the wall is generally wetter than the top, and readings of top, middle, and bottom have been averaged.



Fig. 4. Test wall at 3 years, showing failure on bricks:

section 1 north aspect,
section 2 north aspect,
section 3 north aspect,
section 5 north aspect.

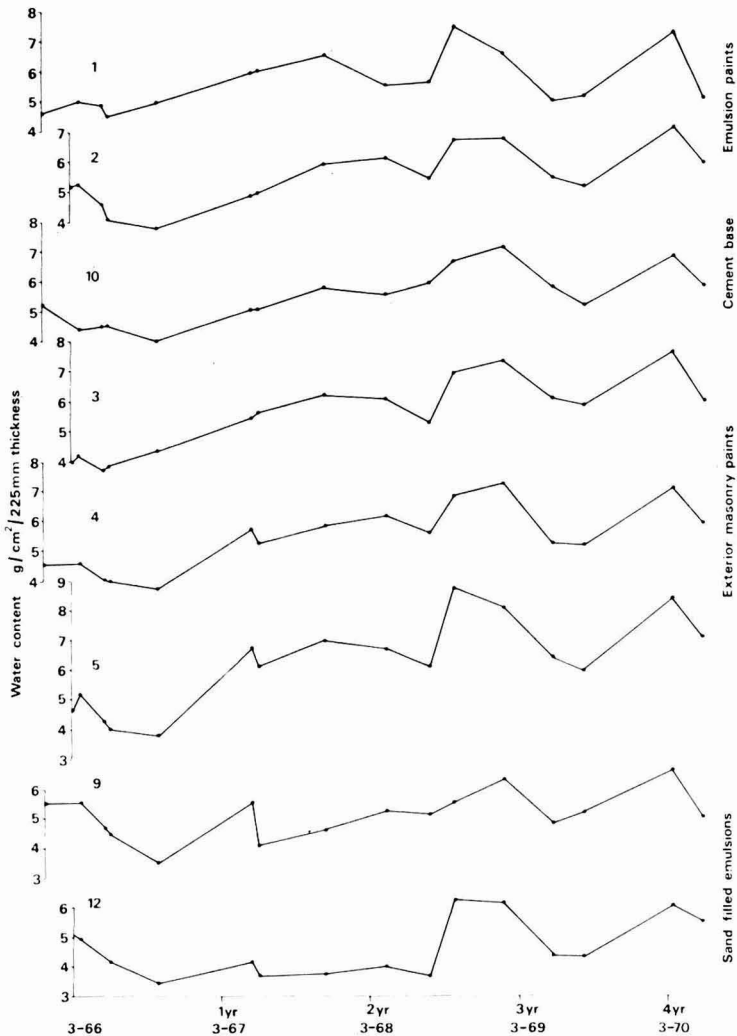


Fig. 5. Water content of test wall

There is naturally a drying-out during summer and an increase of moisture in winter, but the ability of the films to control this is a guide to their performance and an overall comparative rating is shown in Table 2. All the coatings allowed drying-out to occur during the first summer, and few had regained their original state after the first winter, but the thinner coatings deteriorated after this stage and allowed large swings following the seasonal cycles. As failures developed on the north side, some films had to be recoated to enable readings to continue. Some of the thicker coatings largely prevented water entering, but allowed vapour to escape and hence showed an overall reduction in water content over four years. Whether their ability to breathe is sufficient for them to be used safely on older walls with a source of continuing moisture such as rising damp is difficult to decide from this piece of evidence. Failures of stone

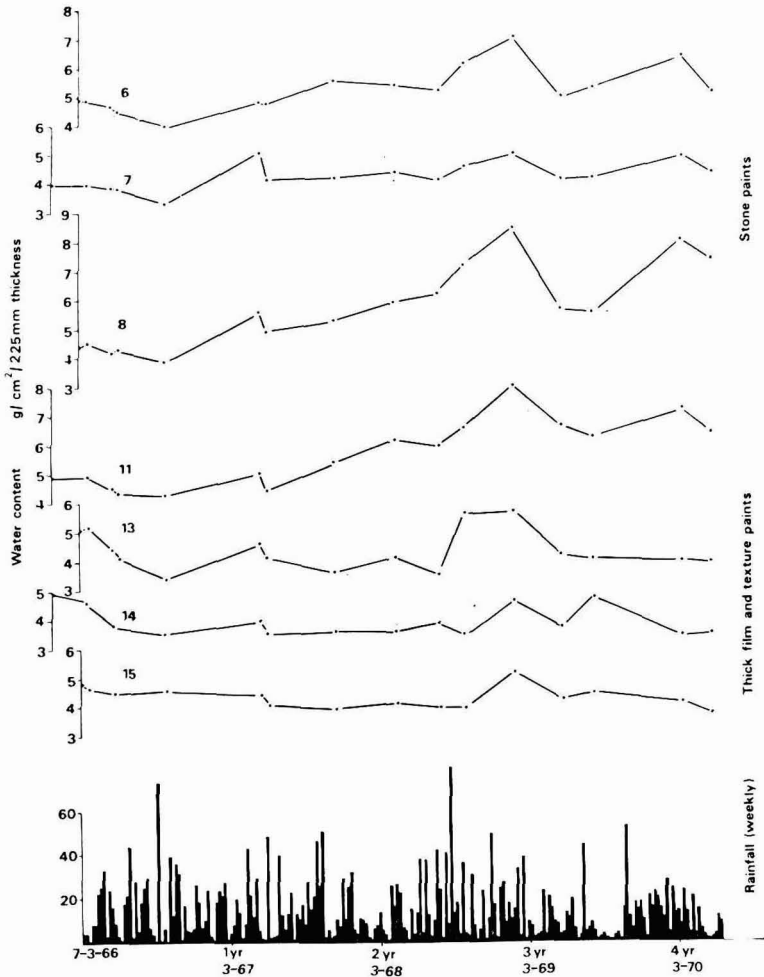


Fig. 6. Water content of test wall

textured paints on renderings, and even of bricks by spalling under the coatings, are not uncommon and it is usually impossible to say how far the paint was responsible for keeping back moisture.

Extensibility and cracking behaviour. The ability claimed for some coatings to bridge a crack or craze which forms in concrete or rendering is a debatable point; thus it has been held that the formation of the crack would require an infinite extensibility for the coating to remain intact, since two adjacent points would become separated by a finite distance. But, in practice, it would not be expected that the coating would adhere to the surface, rough and inhomogeneous as it is, at every point; or that its adhesion would be maintained at the edges of the crack under the tensile forces applied. Coatings of moderate extensibility might then be expected to bridge gaps of limited size. The present work has shown that this is possible, although the claims of high flexibility

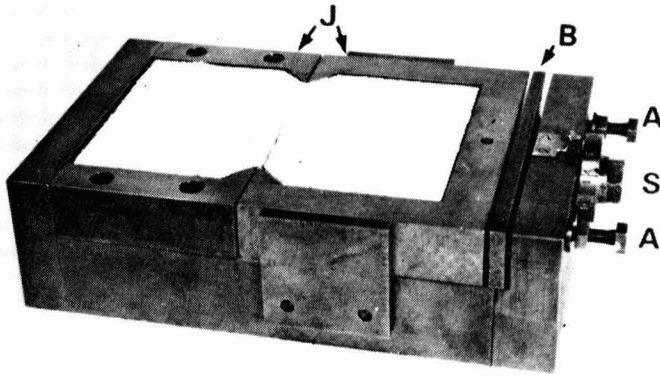


Fig. 7. Apparatus for producing a controlled crack in concrete blocks

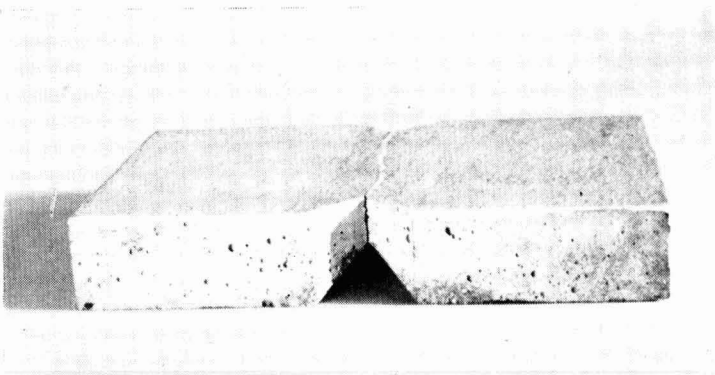


Fig. 8. Cracked mortar block showing extension of paint coating

for many commercial products seem to be excessive, some being initially brittle and others becoming so on ageing.

The apparatus for producing a crack in a mortar block and stretching the film by a controlled amount only is shown in Fig. 7. It is considered essential to produce a crack in tension in this way, since if two pieces of concrete or mortar are butted together and painted over the joint, the paint penetrates the crack sufficiently to invalidate the results. The mortar blocks (1:3 cement: sand) (Fig. 8) are cast in a mould with inserts which produce a weak section across the centre and points at the sides where the block can be gripped by jaws (*J*). The movable jaw is pulled by the fine screw (*S*) in increments of 0.02mm (determined by the screw thread used). To prevent elastic recoil in the screw when the mortar breaks, the bar (*B*) is provided and set with adjusting screws (*A*) so that the travel of the jaws is no greater than the gap between them and the bar, the distance being fixed by feeler gauges. The test procedure requires a step-wise extension and is not closely controlled in time. (The test may alternatively be carried out in a tensile test machine.) The faces of the

block in contact with the jaws are cleaned and smoothed and the block is set squarely in the apparatus. The jaws are gently tightened by hand in suitable increments until a distinct click indicates that the block has cracked; if the coating remains intact the crack can be confirmed by inspection from below. The crack formed should thus not exceed 0.02mm and may be less, but it has not been possible to control it to less than this width. The extension is then continued in suitable steps, e.g. 0.02mm or 0.1mm or more if the coating is fairly extensible, without using the feeler gauges, until the film cracks or small splits develop. The extension is expressed only as a total crack width and not by relation to the initial crack width.

The results (Table 3) are affected by film thickness, which is difficult to control on the rough surface of mortar, whether trowelled or cast. "Normal" brushed coats, conforming to the manufacturer's recommended spreading rate, if known, are applied and conditioned for at least one week including three days at 40°C. The tests may be repeated after suitable periods of natural or artificial weathering.

Brittleness in the coatings shown in the extensibility tests correlates fairly well with cracking failure on the exposure test, but some coatings of high extensibility shown in the direct extension tests do not perform well on the wall or the mortar blocks, either by reason of their low film thickness, or (on the wall) because their permeability allows considerable movement of the substrate. The mortar block test is not very discriminating for the thinner films, most of which fail in less than 0.1mm. The high initial extensibility of numbers 4 and 5 is not paralleled in the mortar block test because their penetration of the laitance increases their adhesion. The main value of this test is in its indication that ability to bridge cracks can only be developed by sufficient thickness, plus extensibility, and possibly a low degree of penetration and hence adhesion. (Large extensions in this test have only been observed with thick rubbery coatings.)

Adhesion: Cement based materials tend to have a weak surface layer which causes loss of paint adhesion, but good quality renderings rarely show this effect. Adhesion of most paint films is adequate except when wet, and measurements should therefore be made in the wet state. This is a difficult technique, as is that of securing pull-off blocks to a vertical rough surface, and adhesion tests have not been made on the exposure wall. No adhesion failures have occurred on the rendering, but the Fletton bricks on the north face have shown the rapid failure common to this surface, which has areas of high glaze and fine fissures, and which develops efflorescence. The solvent-thinned coatings tend to run away from the edges of the crazes, whereas the emulsion paints, being of a thicker consistency, are better able to fill them; even so, flaking caused all five thin-film paints to need repainting after around three years. Failures of adhesion began to show in the stone paints during the third and fourth year near the foot, and even the thicker films are beginning to show failure in the fifth year. The use of sealers is a normal recommendation, but leads to excessive reliance on them when the removal of a weak existing film is strictly necessary. The extensibility of the sealers examined (excluding emulsion-based ones) is usually even less than that of the top coat, a point which might well merit further attention. When used on the Fletton side of the test wall, the majority

of sealers improved the performance of the stone textured paints, or made no difference, but one reduced the adhesion.

Discussion and conclusions

The chosen coatings were of seven different types in three main categories, and in general the thick textured ones gave best results, and the thin films worst; the relation between thickness and durability is evident. On a cost basis, however (with the errors in predicting the life emphasised), there is a less distinct picture, and the individual variations in behaviour of different coatings call for selection according to specific needs rather than on a general basis. For example, one of the best coatings cannot be recommended in very damp situations because of its excessive tendency to growth of algae.

The three thickest coatings gave the longest projected life (11, 13, 14), the best appearance (11, 14) and the greatest control of water movement (13, 14). Their comparative cost in use, shown in Table 1, is unduly favourable, since there is a greater difference between this and the quoted cost of the job than for most of the other coatings. This cost would include more thorough surface preparation, and helps to finance a guarantee, the better appearance throughout and the reduced inconvenience of painting at longer intervals being added considerations, but the authors would conclude that their overall cost in use is not really less than the more conventional coatings. Coating 11 might well have equalled 14 on rendering if applied at equal film thickness, but its adhesion to Flettons was less satisfactory. Coating 13 contains only mineral particles and no pigment, and its totally different appearance makes comparisons difficult. However, it maintained its original appearance fairly well, subject to some dirtiness. No prices were available for the discontinued coating 15, which was claimed to produce a waterproof skin; it maintained good control of moisture up to four years, after which it cracked over the mortar joints of the bricks; it appeared to be embrittled and may have been attacked by alkali.

The poorest performance and appearance on rendering came from the cement-based paint (the cheapest initially and over 50 years) and then from the two emulsion paints, which suffered from the low film thickness produced by their ease of application. They showed severe crazing of the rendering and coating 2 presented a grey appearance throughout, as the chalking rate was low. They gave better results on Flettons than the thin solvent-type masonry paints (Fig. 4). Their high cost in use reflects the unlikelihood that a user would go on repainting with the same paint after the life has been found to be so short.

The masonry paints (3, 4, 5) maintained a better appearance than emulsion paints on the rendering but also cracked, and they failed at an early stage by flaking on the Fletton bricks. Their somewhat high cost in use reflects their lower spreading rate rather than the greater difficulty of brushing them, but (with high volatile contents) their film thicknesses are less than those of the stone textured paints, which have about the same cost in use but produce a thicker film from an even lower coverage.

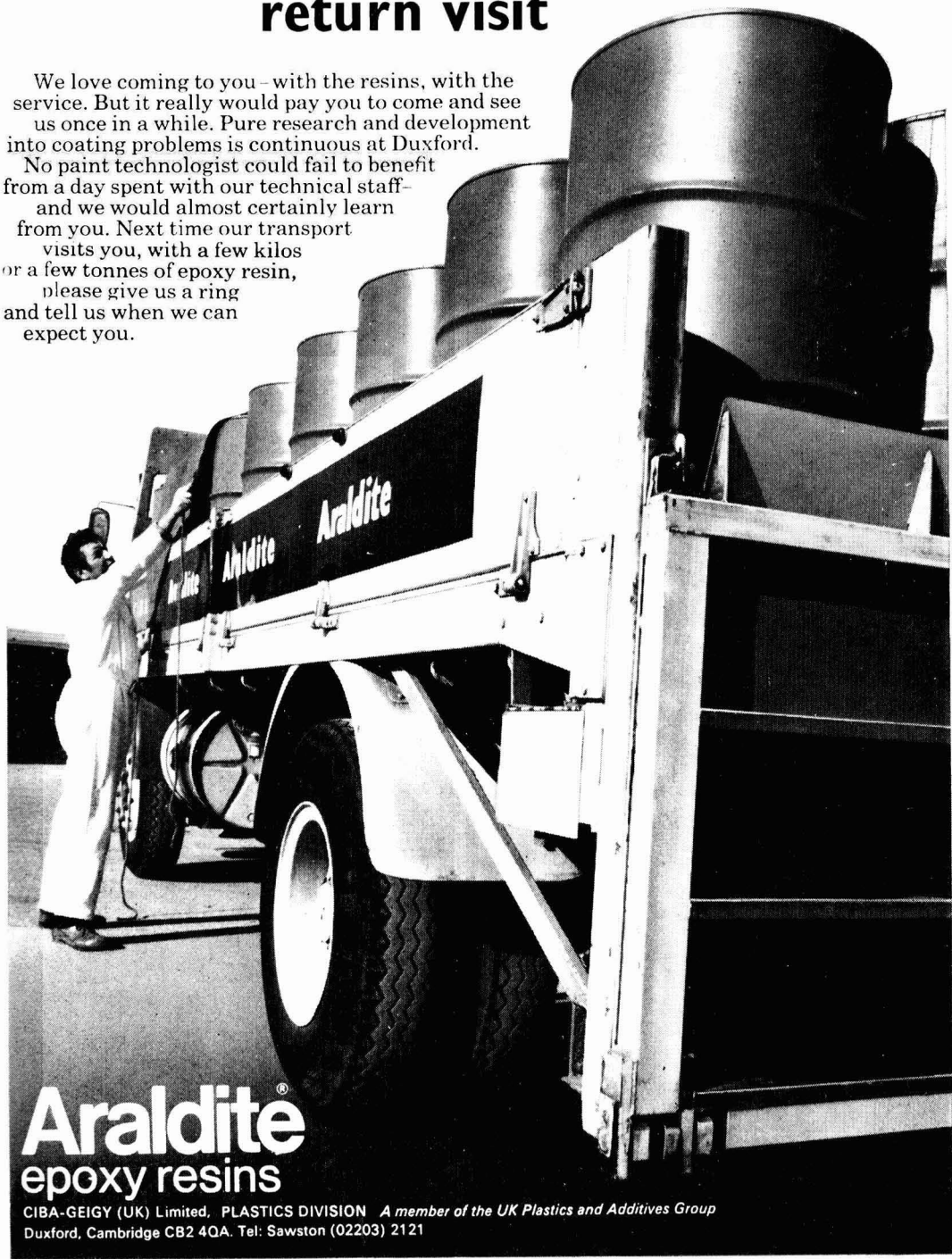
This latter group offers a useful performance on both surfaces and produces favourable costs in use (without benefit of guarantee), but there are individual differences such as unavailability of white, or susceptibility to moulds or algae, which make necessary a careful selection within the group. The texture

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methods has been given in a previous paper¹. There is a further need for studies of their cost in use, based on predictions of their durability. This paper deals mainly with coatings for exterior wall finishes, including some of the less tractable types with coarse textures, which receive little attention in the literature. A comparison of costs is attempted, to demonstrate a suitable method rather than to provide a definitive yardstick.

Appearance and performance requirements

The definition of "life" is necessarily partly subjective; if the coating is mainly protective it can be defined by the eventual failure to protect, eg the entry of water or the appearance of cracks or flaking, but if decorative, the extent of colour change, chalking, erosion, dirt retention and mould growth could determine the need for repainting, and the subjective judgement of these would vary widely between observers. Any failure of integrity (cracking and flaking) must be a more definite indication of need for renewal. Until fairly recently, the general loss of appearance and integrity ran roughly parallel for outdoor exposure, and repainting was commonly needed on both counts at about the same time. With increasing durability of polymer-based coatings, permitting ten years and more before recoating, there is a greater need for retention of the original appearance. Repainting of a perfectly sound film purely for the sake of appearance is going to seem highly uneconomic. For example, some early pvc coatings on metal have shown large changes in certain colours in only two to three years' exposure or the equivalent in artificial weathering, whereas the film has remained entirely sound for very much longer.

Better light-fastness of pigments, non-yellowing media, dirt and mould resistance are thus seen to be important requirements. It is, however, somewhat curious that many traditional building materials—bricks, tiles, stone, slates, wood, copper—are expected to change in appearance on weathering, and with few exceptions the changes are thought to be pleasing or even desirable, whereas newer, "artificial" products are expected to maintain their original appearance for long periods.

Criteria for appearance factors are difficult to establish. Many publications concerned with deterioration of paint films emphasise loss of gloss, a change which can be fairly readily measured, but it is not a property which gives rise to complaints in the building field. Colour change is much more important, and definition of colour by various systems and hence of colour differences in NBS units has become a feature of certain types of coating in the USA, especially coil coatings and film laminates, for which guarantees in various forms are given. Colour differences in numerical form are useful for definition, but for their subjective appreciation need a large series of colour specimens in different hues and directions. The grey scale is not an entirely adequate substitute for the differences of hue, in which changes are probably less acceptable than even larger changes in value or chroma.

One possible approach that has been suggested by the BRS is the use of the Munsell colour atlas. This is frequently used for colour choice, especially by architects, and one could reasonably assume that if any one colour were selected, all those adjacent to it in any direction of hue, value or chroma were rejected. Hence a change by a step as large as any one in the atlas, over a stated number of

paint stop at the junction with a double coat. Thus the formation, or at least the development, of crazes is shown to be dependent on the properties of the paint, i.e. permeability and extensibility.

In general, the results confirm that the life of coatings for masonry and concrete is as yet too short for them to satisfy the large potential market for decorative treatment of these surfaces, especially on large buildings where maintenance costs are high, and to overcome the prevailing opinion that painting will lead to continuing maintenance expenditure. A minimum life of ten years, and a reasonably certain expectation of 20, seem to be needed. For smaller houses, the best of the coatings provide an acceptable performance, but the initial cost is too high for the majority of users and the cost in use does not show attractive savings over the long term.

Fig. 10 shows a possible relation between the applied film thickness on rendering and the "life," up to the time when repainting was considered necessary for appearance. The "life" is as observed up to five years, and thereafter is estimated (broken line). Increasing film thickness up to about 1000gm/m² appears beneficial, but a life greater than about ten years does not seem feasible with the present types of coating.

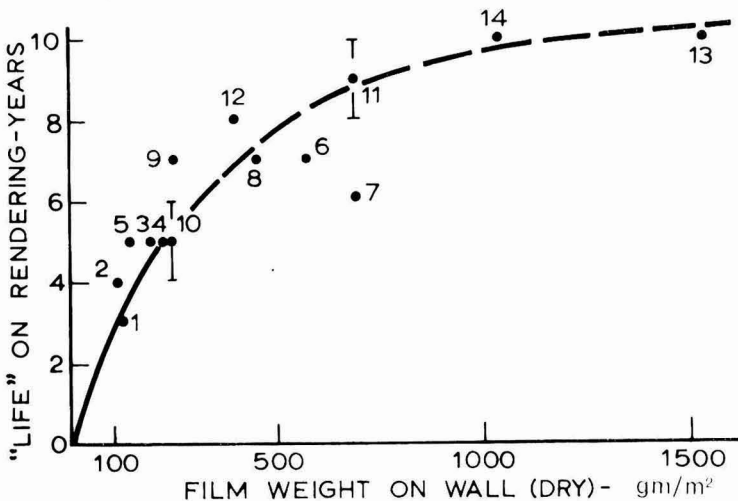


Fig. 10. Relationship between film weight and durability

It is concluded that exterior masonry coatings require a higher degree of extensibility than is normally considered appropriate or achieved with oxidising media, coupled with the ability to exclude rain but permit the escape of moisture. Some coatings of rather low extensibility survived in this test by being relatively impervious, but others of quite high extensibility are seen to be unable to prevent movement of the substrate and subsequent cracking because of their inability to control moisture movement. On this free standing test wall, water penetration is almost entirely via the film; however, there is a further consideration derived from practical experience of frost damage to renderings and brickwork caused by retention of moisture by some types of coatings. Here, when the coating was used for decorative purposes, and there was a risk of water entering the substrate through defects or unpainted points, high vapour permeability might

have been more useful, at the expense of a shorter life for the coating. The difficulty with the thicker and less permeable coatings is that, in spite of showing the lower water content in the test wall, their vapour permeability may not be sufficient to allow drying out to occur in some other situations.

Several more recently introduced coatings are already under examination, and interest centres on the ability of coarse granules, flakes or fibres to reinforce a film.

Acknowledgment

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Discussion at the Torquay Conference

MR M. HESS asked if the film thicknesses quoted in the paper were theoretical, or if they were film thicknesses that had been found in practice. In his experience, contractors often applied films that were much thinner than those specified, and this was often a cause of early breakdown of paint systems.

MR P. WHITELEY replied that, as far as possible, the film thicknesses recommended by the manufacturers had been applied. The spreading rates per gallon of the various formulations were given in Table 1. He agreed with Mr Hess that, in practice, much thinner films than specified were applied.

MR J. A. L. HAWKEY, referring to Fig. 10 of the paper, said that it could be argued that the points plotted gave rise to a straight line, and not a curve as the authors had drawn. He felt that this was symptomatic of the general malaise in both the paint and printing ink industries; there were no new directions for research appearing, and most work appeared to be variations on an established theme.

MR WHITELEY disagreed that the curve was incorrect. It was presented provocatively, for interest, but the projections made were, he felt, reasonable, and agreed with the evidence available.

MR E. L. FRENCH asked if, in the context of the change of appearance of concrete facings on exposure, the authors could comment on the value of water repellent treatments, which were claimed to solve the problem of dirt retention.

The paper had been exclusively concerned with coatings in the form of films applied to the concrete. Had the authors any information on the effectiveness of the impregnation treatments that were being introduced in the building industry?

MR WHITELEY replied that he had little information on either point. Work at the BRS on silicone water repellants had been mainly concerned with their effect on brick, and there was mixed evidence on their utility in dirt-shedding on concrete. No work had been carried out on impregnation coatings as yet, but this was being considered.

DR J. DUNDERDALE said that, in the calculation of maintenance costs, today's prices had been applied at a future date, in other words, no escalation had been allowed

for. If this was taken into account, it would have the effect of reducing the nett discount rate, which would in turn tend to favour the use of long life coatings. Would the authors like to comment?

MR WHITELEY replied that, although he was not an economist, he understood that it was valid to ignore the effect of inflation and future price increases, since costs and benefits were equally affected. Thus, if expenditure rose in the future, this would be more or less cancelled out by the relative increase in value.

The relative balance between materials and labour might change, and perhaps the effect of this on the costing should be considered, but it was a legitimate technique to use today's prices and ignore inflation.

DR DUNDERDALE said that this was quite true if all the escalated costs were to be applied at the same time, but where the maintenance costs were to be applied after different intervals in the future, the argument might not be strictly valid. Since some maintenance would be carried out in two years and some in three, the escalation might have had different effects in each case.

MR WHITELEY answered that, if there was a fairly smooth progression for the escalation, he doubted if there would be much difference in the final result. It would, however, be interesting to see calculations taking escalation into account, so that any difference between the two results could be observed. (It would be even harder to project a variable inflation rate than a fixed one.)

DR R. BULT referred to work that had been carried out at the Verfinstitut TNO on painting concrete surfaces. Various types of paints had been studied on various types of concrete to see if any particular paint was better on any particular type of concrete. However, in the course of this investigation, it appeared that the quality and structure of the surface layer of concrete was at least as important as the quality of the paint in determining the life of the coating.

MR WHITELEY agreed. The work described in the paper had not been concerned with concrete; coatings had been applied to a wood floated rendering, where there was very little laitance. Where laitance was present on the concrete, coatings tended to craze or lose adhesion; however, he had not specifically investigated this phenomenon.

MR P. L. J. MORRIS commented that the base of the wall had been stated to be wetter than the top due to water entering the footings, and this had caused failure of the film. Would a gradient of water content be expected between the top and bottom of the wall, and would this be influenced by the permeability of the paint film?

MR WHITELEY said that the wall had been of engineering brick on a concrete base. The top had a tile capping. It was considered that most of the increased water content at the base of the wall was due to water penetration through the paint surface. On the brick surface of the wall, failure occurred mainly at the foot, spreading up the wall as the water content increased. On the rendered face, failure was fairly general over the whole area, but was initially more pronounced at the foot.

MR G. W. ROTHWELL added that a lot of the wetness at the bottom of the wall was due to the poorer ventilation conditions. A great deal of care had to be exercised in respect of micro-climates in this sort of work. In this case, it was quite normal for the base of the wall to be wetter, irrespective of the coating; it was nearer the ground and did not dry out so easily.

MR A. B. LOCK remarked that exposure experiments on test walls were a great step forward from exposure or durability tests on panels. Could Mr Whiteley amplify his remarks on the correlation between exposure evidence on a test wall and performance on a building wall in practice?

MR WHITELEY said that paint on the exterior wall of a building would be maintained at a higher temperature than that on a wall exposed on both sides. It was probably

less liable to frost damage, or to water behind the coating freezing. On the other hand, if the building was of the type where moisture had to pass out through the walls, which was generally the case, then the coating could cause an accumulation of water.

Thus, it was not certain how well these test wall results correlated with performance in practice. For this reason, an experimental building was being erected which would be maintained at a high internal temperature and humidity. This should produce a better correlation and demonstrate permeability effects.

MR A. R. H. TAWN (Chairman) asked if, in general, the test wall gave better or worse results than a building with warmth and humidity inside.

MR WHITELEY said that he was loth to give an opinion, but if forced to do so, would expect slightly worse results with the wall; there was probably a slight acceleration of breakdown.

MR J. BOROKY referred to the instrument for determining extensibility by cracking a cement block. Was it intended that this would give a guide to extensibility over cracks caused by structural changes in a building, such as a sinking foundation, or cracks caused by temperature fluctuations. The latter could have particular significance in the Southern Hemisphere, where temperatures exceeding 120°F were observed during the day, falling to less than 50°F at night. In this case, the application temperature for the test block might be of importance.

On the correlation of test wall results with practical findings, he felt that, in climates with strongly fluctuating temperatures, the test wall results would be better than those in practice.

MR WHITELEY replied that coatings were not expected to stretch to cover large cracks caused by structural movement. The authors' interest had been in crazing in concrete or rendering; concrete crazes were cracks that occurred on the surface only. There was a long-standing argument that coatings could not stretch over a crack which formed beneath them, since this would require a finite extension between two points which were infinitely close. This argument did not seem valid, particularly on a non-homogeneous substrate of cement paste and sand grains, where slight adhesion failure might be expected to occur at the edge of the crack in such a way as to allow a finite width to be extended and remain unbroken by the formation of a crack or craze. The apparatus had been designed to prove this point.

The essential feature was that, if a gradually increasing pulling force was applied to a piece of mortar with a coating on its surface, strain energy was built up in the screw, causing a recoil on cracking. The device had to have a restraint incorporated to limit the width of the crack; this had been fixed by the pitch at about 20 microns, but was of the order of size of crack that might occur in practice. Many coatings cracked immediately, but some rubbery coatings would stretch much further, and might be useful in maintaining waterproofing on some buildings. This type had not been closely investigated.

MR FRENCH said that the rate of cracking was also important. Were the authors satisfied that the crack would be rapid in formation? If there were a slower rate of cracking, stress relaxation might occur. Was this likely in the type of coatings evaluated?

MR WHITELEY replied that crack formation in the substrate would be rapid. Most of the coatings evaluated were viscoelastic, and their behaviour would depend on the rate of extension and temperature. Stress relaxation at certain temperatures was possible. It would not occur, however, at temperatures below the glass transition temperature, when cracks would form very quickly. The results were dependent on temperature and rate of strain, which were not controlled in this apparatus. Although the results were perhaps not very precise, he felt that they could provide some guide lines. (*Authors' Note:* It would distinguish, for instance, the two paints in Fig. 9, of which the crack-free film was not in Table 3.)

MR ROTHWELL said that it had been found that films could be divided roughly into the categories brittle and visco-elastic. The former would crack almost instantaneously in a tensile test, as would all thin films; results for viscoelastic films of sufficient thickness would be affected by the uncontrolled rate of the cracking device.

MR H. MEYER asked if the application costs included such items as scaffolding costs for larger houses; obviously it was cheaper to paint a smaller house.

Also, had any work been carried out on concrete without laitance? Most houses in the future would be built of prefabricated, prepainted concrete, and results on this might be much superior to those from the rendered brick wall.

MR WHITELEY replied that the labour costs had included overheads, and, as the footnote showed, compared quite well with the London Measured Rates used for estimating. The costs were self consistent for this particular operation, but were not comparable with those of painting a high building. The much higher "quoted" costs in the table for specialist application would include scaffolding.

He thought that the results on the rendering were likely to be better than on concrete, particularly precast concrete, to which good adhesion was difficult to achieve. (*Authors' Note: Adhesion to concrete can also be affected by residual mould oils.*)

MR TAWN remarked that most papers advocating the use of very long life coatings, of more than twenty years, suggested the use of polymers such as polyvinylidene fluorides, justifying the increased cost by quoting the exceptionally large costs of repainting high rise buildings, as distinct from single storey properties. No calculations had appeared in the paper taking these related costs into account. Had this aspect been ignored by the authors?

MR WHITELEY replied that this type of coating required stoving and could not be used on concrete. Although pvf and similar polymers had probably the greatest durability, they had not found much application to wall surfaces, except applied to a sheet base which was fixed to the wall. In principle, a high cost coating of long life should be acceptable for high buildings, but generally these were not given decorative coatings because of doubts about performance. The authors' cost figures had not been applied to the high rise situation, when a life of over 10 years was essential.

Colour specification by visual means*

By K. McLaren

Imperial Chemical Industries Limited, Dyestuffs Division, Hexagon House, Manchester M9 3DA

Summary

By combining a series of neutral grey films with about 1,300 colour samples, a colour atlas illustrating over 20,000 shades has been produced, each colour being specified by a simple combination of letters and numbers. As a means of communication it is unrivalled.

Keywords

Properties characteristic and conditions primarily associated with materials in general
colour

Equipment primarily associated with
colour chart

Process and methods primarily associated with analysis, measurement and testing
colour matching

La spécification de couleur par moyens visuel

Résumé

On a mis au jour un atlas de couleurs, renfermant plus de 20,000 nuances, par la combinaison d'une série de feuillets gris neutre avec à peu près de 1,300 échantillons de couleur, chaque nuance de l'atlas étant définie par une simple combinaison de chiffres et de lettres. En tant qu'un moyen de communication c'est sans rival.

Farbbestimmung mit dem Auge

Zusammenfassung

Durch Kombination einer Reihe neutraler grauer Filme mit ungefähr 1300 Farbmustern wurde ein Farbatlas erzeugt, welcher über 20 000 Farbtöne illustriert. Jede Farbe wird durch eine einfache Kombination von Buchstaben und Nummern bestimmt.

Als Verständigungsmittel ist die Methode unvergleichlich.

Спецификация цветов визуальным методом

Резюме

Совмещая серию нейтральных серых пленок и приблизительно 1300 цветных образцов, получен атлас цветов, иллюстрирующий свыше 20-ти тысяч оттенков, причем каждый цвет обозначается простым сочетанием букв и чисел. В качестве средства коммуникации атлас не имеет себе равных.

Introduction

Although it is theoretically possible for the written manufacturing or purchasing specification of an article to include its colour and permitted colour tolerances, the practical difficulties involved are still so formidable that whilst each year sees some improvement, it will be a very long time before colour can be treated purely objectively once the original choice has been made. Physical samples illustrating the required colour will therefore continue to pass along the manufacturing chain, which in the textile industry can be both long and branched.

*Presented at the Torquay Conference on 5 May 1971.

One obvious way of overcoming the inconvenience this causes is to have available a number of copies of a collection of colour samples representative of the whole gamut of surface colours, and if the original subjective choice can be confined to this particular collection, the problem of communication is solved. This has proved successful in the paint industry in the UK, 101 colours being illustrated in BS 2660:1955, and even if the current attempt to double this number is successful, there is still no difficulty in providing samples, as the 101 colours themselves are, in the main, a selection of the 1,500 or so colour samples in the Munsell collection, which is universally recognised.

The situation in the textile industry is, however, quite different. Textile designers have never been restricted to any range of standard colours and would undoubtedly find even the full 1,500 Munsell range frustratingly small. They make their initial choices from literally any source and the dyer will reject the chosen colour only if it cannot be obtained satisfactorily on the fibre in question for technical reasons: rarely is a colour rejected in favour of another similar shade which was fashionable, say, five years ago and for which proven recipes on many fibres are still available.

The total number of surface colours that a trained observer can discriminate has been reliably estimated at 10 million, and whilst commercial tolerances are many times larger than the limits of visual differentiation, the dyer may still be called upon to match any one of, say, a hundred thousand shades, none of the others being close enough. These statistics clearly indicate that even the 7,000 colours in the Maerz & Paul dictionary or in the "Villalobos Atlas" cannot be used for colour specification in the textile industry. Of course, it is theoretically possible to interpolate any number of colours between adjacent Munsell colours, but this is only practicable if the colours vary in only one Munsell dimension; the great majority of colours would need interpolation in three dimensions and this is probably impossible by the unaided eye with any degree of precision.

Even if one is not aiming at the evenness of spacing which is such an important characteristic of the Munsell samples, the cost of producing a comprehensive collection by conventional means is prohibitive. In 1961, however, an unconventional approach was tried which was successful after eight years' development by a team of colour technologists. The new approach was to use one of the techniques of colour photography.

The dye-transfer process

There is a well-established technique of producing colour prints in which black and white film is exposed to, say, the red light coming from a coloured object (using a red filter), treated with a developer which also hardens the gelatin in proportion to the amount of red light falling on the film, washed in hot water to remove all the unhardened gelatin and then soaked in a solution of a magenta dye; the hardened gelatin absorbs the red dye which is then transferred to gelatin-coated paper so that the amount on the paper in each part of the image is proportional to the amount of red light originally coming from the object. Considerable experience of this technique in producing many copies of sales aids, etc., was available, and a reasonable degree of reproducibility had been achieved. The idea was not merely to use this technique to produce

colour prints of a collection of colour samples but actually to create the colour samples themselves. A method of achieving this was evolved using a step-wedge produced by gradually increasing the exposure in strips across the film. The step-wedge is processed in the hardening developer, soaked in a magenta dye and transferred to paper, giving a series of stripes increasing from white through pink to full red, the spacing being controlled, of course, by the original exposure of the step wedge. By repeating this operation with a yellow dye superimposed at right angles, a series of squares arranged in matrix form varying from white to full red, full yellow and mixture shades (orange) could be produced. The same procedure could then be repeated for the two other pairs of primaries, so that an extremely cheap method of producing a large number of evenly spaced colour samples was evolved. This process is covered by British Patent 1,115,279.

The next step was the obvious one of increasing the gamut by superimposing the third subtractive primary and the first attempt to do this was to orient the three wedges at 60° to each other (giving triangular colour samples) but it proved quite impossible to control the uptake of the three dyes closely enough. The effect of the third primary, however, can also be obtained by using the other two primaries and varying amounts of a neutral grey, but this failed because none of the suitable neutral grey dyes was homogenous; all were in fact mixtures of three primaries! For a considerable time the problem appeared to be insoluble, and the project had almost been abandoned when a solution was found: this was to put varying amounts of the neutral grey dye in transparent films, each of which could be superimposed over any colour sample; this vital step is covered by B.P. 1,160,673.

This breakthrough also solved another problem which might otherwise have proved insoluble: in order to achieve the closeness of spacing desired it would have been necessary to take every combination of each pair of primaries, split each into at least 10, and over-dye with a different amount of neutral grey. Not only would this have been very difficult to carry out, but it would also probably have been impossible to arrange the resultant colour samples in a logical manner to permit rapid identification of the one nearest to a given specimen. This is because of the fundamental characteristic of colour samples: they can only be arranged systematically in three dimensions—hence the term “colour solid.” The “Munsell Book of Colour” is not as easy to use as the “Munsell Colour Tree,” though the former is easier to handle. The physical separation of the third dimension of colour, which Munsell termed “value,” converted a two-dimensional array into a three dimensional solid without affecting its ease of handling.

The photographic colour atlas

Once this breakthrough in design had been made, all that remained was the arduous task of finding by trial and error the correct spacing of the step-wedge to give the best visual spacing of the resultant colour samples, ensuring that each replicate was identical and that each set of neutral grey filters was correctly spaced. The filters were obtained by varying the exposure of ordinary black and white photographic film.

The final version was obtained from a dye transfer step-wedge consisting of 15 steps which was used for each of three pairs of primaries, thus producing an atlas consisting of $3 \times 15 \times 15 = 675$ colour samples. Twenty neutral grey filters were produced making the total number of colours illustrated 13,500, a significant increase over existing atlases.

The usefulness of this photographic colour atlas as a means of colour specification was determined in rather an unusual manner.

A major part of the technical service provided by dyestuff manufacturers consists of matching shades submitted by dyers—one of the branches of the chain already mentioned. In 1963 this was facilitated by the introduction of IMP, Instrumental Match Prediction, the first system using a digital computer. This required the measurement of the colour of the sample to be matched by colorimetry, but unfortunately this has a number of limitations which have restricted the use made of IMP. Once the XYZ values of each of the 13,500 colours in the atlas had been determined, however, it could be used to provide input data for the IMP computer, thus overcoming the limitations of instruments. After a trial lasting several months, it was clear that the photographic atlas was reliable for this purpose and therefore reliable enough for communication between any two observers, particularly between customers and any technical service laboratory. For this purpose, the number of atlases required would be in the thousands and it would have been quite impossible to produce these photographically; moreover, the light fastness of many of the pale shades was as low as 1 on the blue wool scale and serious fading had been observed after only a few months' use. The McCorquodale colour depositing process appeared to be ideal for this purpose, and examination of a number of paint shade cards taken at random from a run of tens of thousands printed by this process showed that the desired colour constancy could be achieved. McCorquodale Colour Display Ltd. accepted the commission, which before completion was to challenge their skills to a degree not fully realised at the time of acceptance.

McCorquodale wanted one sample of each shade for matching, and this provided the opportunity to eliminate one or two weaknesses that users had detected in the photographic atlas. The most important of these arose because, although the difference between the amount of one primary in adjacent strips was constant along the length, this constant difference was combined with amounts of a second primary varying from zero to full shade. A compromise had to be struck, because if the colour difference between two adjacent strips in the absence of the second primary was correct, the difference in the presence of a large amount of the second primary was undetectable; conversely, a difference which was correct when the full amount of the second primary was present would be excessive for one primary on its own. The second defect arose from the fact that the three primaries used were not nearly as good as the standard trichromatic printing inks (dyers have never used magenta, cyan and yellow primaries) and hence the resultant gamut was not as good as one would have liked.

The first weakness was overcome by using differently spaced step-wedges for different regions—as many as six were used for the red-purple-blue section—and the second by using two blue primaries, a reddish blue for the purples

and a cyan for the greens, with mixtures of both for pure blues. These improvements more than doubled the number of colour samples submitted to McCorquodale for matching.

The production of thousands of grey filters to extremely close tolerances was also beyond ICI's capabilities, but the company was fortunate to have the co-operation of Ilford Ltd. who, like McCorquodale's, did not fully realise the magnitude of the task when they accepted it.

The final atlas

The atlas which finally emerged consisted of 1,379 colour samples printed in three sections—

red—orange—yellow
yellow—green—blue
blue—purple—red

each section spread over four pages and each sample identified by a simple code e.g. *R7Y4*, *B0R22*,—*R*, *Y* and *B* referring, of course, to the primaries originally used. The neutral grey filters were numbered *N0-N19* and it is interesting to note that *N0* indicates that no filter has been used, *N1* is a piece of the uncoated cellulose acetate film base, *N2* is a piece of film which has not been exposed, but was developed and therefore represents the fog level, and *N3-N19* were given progressively longer exposure.

The colour atlas was intended to have a life of many years, even if used continually, so it was desirable that only pigments of high light fastness were used. This would have restricted the gamut excessively, however, so it was necessary to accept a minimum of four on the blue wool scale for certain pale shades. Even with this concession, however, the gamut of very bright shades was restricted, but the effect of this was minimised by the simple expedient of using the neutral grey filters to dull the sample of the bright shade to be matched rather than the atlas colour sample. When the filter was used for this purpose, the filter number was prefixed by *X* in the identification code: a separate set of filters numbered *X1-X5* is supplied for convenience in handling.

The theoretical maximum number of shades the atlas can illustrate is $20 \times 1,379 = 27,580$, but in practice many combinations involving deep grey filters are indistinguishable; the actual number of discriminable shades has never been determined. The closeness of the spacing has been determined for every adjacent pair of the 1,379 colour samples (2,551 in all): the mean value is 4.5 AN(42) units using the SDC-recommended colour difference formula. The mean value of the difference caused by changing the neutral grey filter by one step is 4.7 units.

Although the number of shades illustrated is by far the largest of any atlas, even making a generous allowance for the unknown number of combinations not discriminable, the atlas must not be regarded as being closely spaced, as its spacing is about five times wider than what is widely accepted in America as a commercial tolerance for dyeing i.e. 1 NBS unit. For practical purposes, however, the spacing would seem to be ideal. The most serious limitation

concerns the gamut covered: some textiles can be dyed in shades that are more saturated than any in the atlas. This is possible because their surface characteristics permit light to penetrate and to be reflected back to the eye after multiple internal reflections; extreme examples are velvets and cut-pile carpets. Whilst excessive brightness can be handled by means of the X filter, there is no practical method of either increasing the saturation of an atlas colour sample or of decreasing that of pile fabrics.

[Received 9 September 1970]

Discussion at the Torquay Conference

MR S. L. DAVIDSON referred to work done by Davidson and Friede (*JOSA*, 1953, 43, 581) on carpet standards and dyeing. They had shown that the MacAdam equation was better than the Adams Nickerson equation. Would Mr McLaren comment on this work?

MR K. MCLAREN said that, until recently, the Davidson and Friede paper had been the only one in which the visual judgments of professional colourists were available for comparison with colour difference measurements. Unfortunately, the statistics used in the paper had been rather elementary; for instance, no correlation coefficient had been calculated; when he had done so and applied tests of significance, the superiority of the MacAdam formula was found to be not above chance level. The Davidson and Friede work had been included in the SDC investigation, but unpublished work by Jaeckel et al at the Hosiery and Allied Trades Research Association, using essentially the same technique as Davidson and Friede, but applying modern statistical methods, had shown that formulae of the Adams-Nickerson type correlated better with visual judgement, to a degree which was statistically significant. The evidence from Jaeckel's work was to be presented to the CIE at its Barcelona meeting later in 1971.

MR DAVIDSON asked if any problems in the use of the atlas would be caused by metamerism, for example, when customers from other countries had a sample coloured by dyes different to those in use in the UK.

MR MCLAREN replied that the light source for use with the atlas was specified as source D.65, which was universally obtainable. If there was a doubt about metameric effects, it was recommended that the sample be matched under both the standard illuminant and tungsten light, and both sets of co-ordinates forwarded. This tended to solve the problem, particularly as the colours used by McCorquodale had been chosen for illuminant stability.

MR P. FINK-JENSEN asked if the size of the sample in the atlas was great enough to allow accurate discrimination between neighbouring samples.

MR MCLAREN said that the size chosen had had to be a compromise. A 2in square sample would have been preferred, but size limitations had dictated a size of $\frac{3}{4}$ in square. Criticisms of the atlas had been asked for, but none had been received in respect of this particular aspect.

MR FINK-JENSEN remarked that uniform spacing of the samples had not been attempted. What was the greatest colour difference between samples, in terms of NBS or some other unit? It seemed that the usefulness of the atlas would be determined by the area where there was the greatest colour difference between samples.

MR MCLAREN pointed out that the atlas was intended merely as a means of communication; the deciding factor had been to make the spacing as visually uniform as possible. The average colour difference between adjacent samples was 4.7 AN 42 units, the maximum at any point was 16.5 and the minimum 0.3 units. What proportion of this was due to the non-uniformity of Adams-Nickerson colour space

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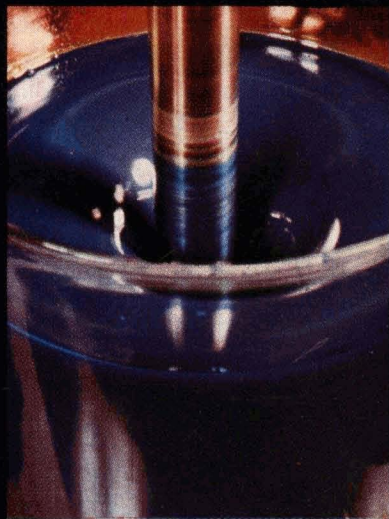
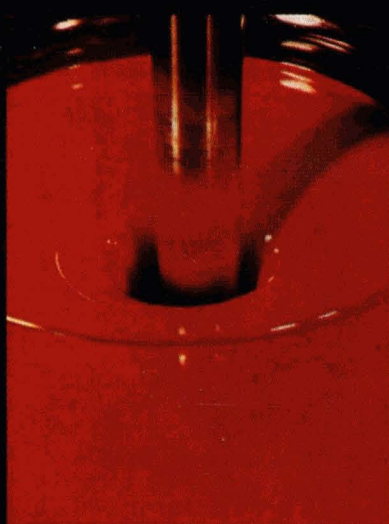
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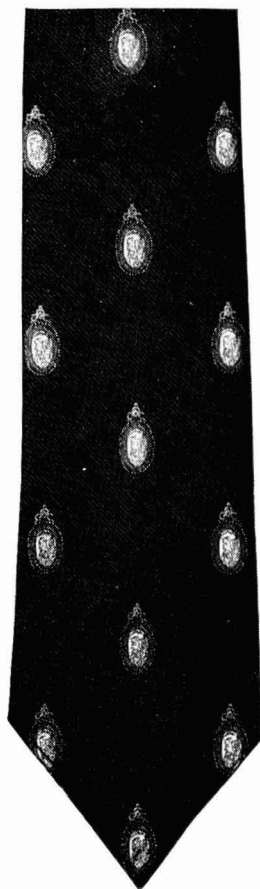
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had not been determined. The main objective of the atlas had been that a recipe for a colour could be produced, and it was felt that, if the recipe gave a shade which was not more than one square away from the target, this was good enough. Again, no complaints on this account had arisen during the period of use of the atlas.

MR FINK JENSEN said that this applied to the dyeing industry; in the paint industry, different spacing might be needed. Did Mr McLaren think that the atlas would be of use in the paint industry?

DR D. A. PLANT, who had been involved in the use of the atlas in paint formulations, replied that it had been of limited value in parts of the paint industry as a means of communication.

MR G. A. NEWELL, developing the theme that the atlas had been developed for the textile industry, pointed out that there was an essential difference in colour matching in textiles and paints. In the case of textiles, the texture of the materials, which caused internal reflections, would influence the colour. There were similar problems, of course, in matching matt and gloss paints. How did the textile matching facility tie up with the needs for paints?

MR McLAREN said that Mr Newell was quite right. In fact, in some senses the atlas would be more appropriate if designed for the paint industry, since paint films had been used for the colour samples. For textiles, matt samples would have been preferred but a degree of glossiness halfway between matt and gloss had been chosen to reduce the soiling propensities of matt films. However, in the textile field, the dyer was used to dyeing a fabric to, say, a watercolour painted by the designer, and to achieving the same shade on a variety of substrates, so that the use of a smooth-surfaced sample had not caused any problem, to his knowledge.

MR DAVIDSON said that, in 1931, the CIE had established a Standard Observer, based on work by Wright and Guild which was of doubtful accuracy. There was no system of uniform spacing based on tristimulus values. Would Mr McLaren comment?

MR McLAREN replied that Wright and Guild's work was by no means of doubtful accuracy. The errors in the 1931 CIE 2° Standard Observer data arose from the need to make the \bar{y} function identical with that of the luminous efficiency function ($V\lambda$ curve) defined by the CIE in 1924: this was known to have been out by a factor of ten at the violet end of the spectrum. In spite of this the 2° data had been remarkably successful, particularly in match prediction, but it was desirable that the 1964 CIE 10° Supplementary Standard Observer data should be used in industry because viewing conditions were nearer 10° than 2° and the 10° data did not perpetuate the $V\lambda$ errors. It was just as easy to convert spectrophotometric data into $X_{10} Y_{10} Z_{10}$ as $X_2 Y_2 Z_2$ but no colorimeters had yet been made to conform to the former.

He added that he had deliberately refrained from using the well-known term "NBS units" to describe colour differences calculated using the Adams-Nickerson formula. The reason for this was that the term obviously suggested that the NBS colour difference formula had been used, but to convert Adam-Nickerson units into true NBS units, factors varying between 11.3 and 93.1 would be needed, whereas the Adams-Nickerson formula could have only one (*Color Engineering*, 1970, (8) 21). As values of 40, 42 and 50 had been used at various times, the SDC selected 42 but recommended that the units should be termed "AN (42)" units to avoid ambiguity. There was also a danger that the term "CIE unit" would similarly lose its precision by being applied to colour differences calculated using formulae other than the 1964 CIE formula, as suggested by Mudd and Woods (*JOCCA*, 1970, 53, 852).

MR DAVIDSON agreed with the comments on the NBS unit. However, he was concerned about the references to the Munsell system. Munsell had been an artist, and his spacing had not been uniform. There was a Munsell renotation system, to which the Adams-Nickerson equation applied. Moreover, the Munsell Book of

Colour was available with both matt and glossy chips, which was perhaps more useful for fabric matching. Would it not be possible to select from the Munsell System enough samples to satisfy the needs of the dyer? He himself had helped in the production of a colour system in which mixtures of pigments were used to produce a specific colour, and this colour had then been placed in various planes of brightness. The concept of using a neutral grey filter was rather new, in effect, it was using a negative colour concept to match bright colours that didn't exist.

MR MCLAREN said that the range of colours in the Munsell System was totally inadequate to satisfy the textile designer. He could not comment on its usefulness in the paint field.

MR R. SMITH, in a separate communication, said that in his presentation Mr McLaren had mentioned that the BS 2660 colour range was selected from Munsell. In his experience this was not the position, the original colours being first selected and approximate Munsell references being given later as an aid to designers using the colour range. This fact was stated on the cards themselves, to quote: "The Munsell System of Notation has not been adopted by BSI but the above approximate Munsell references are given as an aid to designers," and was supported by the facts that:

- (a) visual comparison with the Munsell book of colour showed that the matches were often only approximate,
- (b) Munsell references were often intermediate, for instance 5Y 9.25/1, for which no visual references were available, and
- (c) value and chroma were often such as to fall outside the visual range represented.

Gas chromatographic study of the interaction forces between binder and solvent in paint films*

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Summary

Solvent retention volumes have been determined on gas chromatographic columns with polymeric binders as the stationary phases, and these are representative of the evaporation behaviour of the solvents from the binder phase. From these experiments characteristic empirical indices, and even, in some cases, absolute thermodynamic data, indicating the total interaction of solvents with binders, have been obtained. The indices for the solvents investigated on polar binders and obtained from direct measurements on concentrated binder/solvent systems increase with the increased content of polar groups in an homologous series of solvents. Likewise the indices for polar solvents on different binders increase with the increased content of polar groups in the binder, e.g. with the increased acetate content in a series of vinyl acetate polymers. Further, the order of solvents with respect to the indices indicating the interaction of solvents with binders, varies from binder to binder, which is due to the different proportions of the individual interaction forces yielding the total interaction.

Keywords

Properties, characteristics, and conditions primarily associated with coatings during application

solvent/binder interaction

Processes and methods primarily associated with analysis, measurement and testing

gas chromatography

Une étude, au moyens de la chromatographie en phase gazeuse, des forces d'interaction liant/solvant en feuillets de peintures

Résumé

Les volumes du solvant retenus ont été dosés sur des colonnes de chromatographie en phase gazeuse, en se servant des liants polymères comme les phases fixes, et ils sont typiques en ce qui concerne l'évaporation du comportement des solvants à partir du liant. On a obtenu au moyens de ces expériences des indices caractéristiques et à la fois empiriques, et même en certains cas des données thermodynamiques absolues, qui indiquent l'interaction des solvants et des liants. Ces indices, des solvants examinés sur des liants polaires, obtenus à partir des mesures directes sur des systèmes concentrés de liant/solvant augmentent au fur et à mesure de la teneur en groupements polaires dans une série homologue de solvants. Également les indices des solvants polaires sur de divers liants augmentent selon l'augmentation du groupement polaire dans le liant, par exemple, selon la teneur augmentée en acétate dans la série de polymères de l'acétate de vinyle. D'ailleurs, l'ordre des solvants au point de vue des indices de l'interaction des solvants et des liants, se varie d'un liant à l'autre, à cause de différentes proportions des forces individuelles de l'interaction qui comprennent l'interaction entière.

Gaschromatographische Untersuchung der in Lackfilmen zwischen Bindemittel und Lösungsmittel aufeinander Einwirkenden Kräfte

Zusammenfassung

An gaschromatographischen Trennsäulen mit polymeren Bindemitteln als stationäre Phase wurden Retentionsvolumina von Lösungsmitteln bestimmt, die für das Verdampfungsverhalten der Lösungsmittel aus der Bindemittelphase charakteristisch sind. Aus diesen an

*Presented at the Torquay Conference on 6 May 1971.

концентрированных Bindemittel-Lösungsmittel-Systemen bestimmten Werten konnten empirische Kenngrößen, in einigen Fällen auch absolute thermodynamische Daten für die Wechselwirkung zwischen Bindemittel und Lösungsmittel erhalten werden. Diese empirischen Größen nehmen für die untersuchten Lösungsmittel gegenüber polaren Bindemitteln mit zunehmendem Anteil der polaren Gruppen innerhalb homologer Lösungsmittelreihen und für polare Lösungsmittel mit zunehmendem polarem Anteil der Bindemittel, z.B. mit steigendem Acetatgehalt in einer Reihe von Vinylacetat-Polymeren zu. Die Skala der Lösungsmittel bezüglich der Kenngröße ihrer Wechselwirkung gegenüber Bindemitteln ändert sich ferner von Bindemittel zu Bindemittel, was auf einen unterschiedlichen Anteil der einzelnen Wechselwirkungskräfte an der Gesamtwechselwirkung zurückgeführt wird.

Газово-хроматографический анализ взаимодействующих сил между связующим веществом и растворителем в красочных пленках.

Резюме

Определялись сохраняемые объемы растворителя в газо-хроматографических колоннах с полимерными связующими веществами в качестве стационарных фаз, которые отражают поведение испарения растворителей из фазы крепителя. Из этих опытов были получены характеристические эмпирические показатели и даже в некоторых случаях абсолютные термодинамические данные, определяющие полное взаимодействие между растворителями и крепителями. Эти показатели растворителей, анализированные на полярных крепителях и полученные путем непосредственного измерения на концентрированных системах крепителей-растворителей, повышаются с увеличением количества полярных групп в гомологической серии растворителей. Точно также, показатели полярных растворителей на различных крепителях повышаются с увеличением количества полярных групп в связующем веществе, т. е. с повышением ацетатного содержания в ряде винил-ацетатных полимеров. Далее, порядок растворителей по отношению к показателям взаимодействия между растворителями и крепителями, изменяется от крепителя к крепителю, из-за различных пропорций отдельных взаимодействующих сил составляющих итог общего взаимодействия.

Introduction

In addition to polymeric binders and pigments, paint films generally contain a number of further components which strongly influence the properties of the film. The volatile components (solvents, monomers and low molecular weight polycondensation products) evaporate almost completely from the liquid paint during the process of film formation. The properties of the film, however, are greatly affected by the progress of the evaporation and by the residual volatile components remaining in the film. Both factors depend not only on the properties of the volatile components, but also, as shown by Tysall¹ and Crowley², on the interaction between the solvent and the other film components.

This has led to a great interest in finding parameters that characterise the molecular interaction and which may be determined easily by experiment. As originally defined by Hildebrand and Scott³, the solubility parameter δ is a constant for a material, but in the development of the theory only intermolecular dispersion forces were taken into consideration. It follows from this that, when the difference in the solubility parameters for two components is small, good miscibility would be expected. However, more recently, systems comprising several parameters have been used to characterise the intermolecular interactions (see e.g. Gardon⁴, Crowley²,⁵ and Hansen⁶). These systems take into account the additional influence of polar interaction forces, such as hydrogen bonds and dipole interactions. For example, Hansen subdivided Hildebrand's solubility parameter into individual expressions for the dispersion forces δ_d , the dipole-dipole interaction δ_p and the hydrogen bonding forces δ_h .

$$\delta^2 \equiv \delta_d^2 + \delta_a^2 \equiv \delta_d^2 + \delta_p^2 + \delta_h^2 \dots\dots\dots(1)$$

Although it is often possible to predict the behaviour of binder/solvent systems, with these complex solubility parameter systems, the following factors usually lead to errors in the prediction:

- entropy changes are not considered,
- the individual parameters are evaluated for the pure substances or for model systems and are therefore only characteristic quantities for the interactions between molecules in these systems,
- the parameters can, in general, be directly determined only for the volatile components. The corresponding values for the binders are mostly evaluated indirectly, for example from solubility experiments.

By measuring the gas chromatographic retention properties of solvents on columns in which the binder is used as the stationary phase, characteristic indices for the distribution equilibrium of solvents between the binder and an inert gas phase⁷, which are also dependent on the molecular interaction forces between the solvent and the binder, can be determined. The effect of these interaction forces in normal gas chromatography is to separate volatile substances, due to their different retention volumes on standard stationary phases. A volatile substance that has a strong interaction with the stationary phase requires a larger volume of the gas phase (retention volume) for its complete elution than a substance of equivalent boiling point that is inert with respect to the stationary phase.

When binders are used as the stationary phase, the logarithm of the solvent retention volume corrected for temperature and pressure, $\log V_r$, is proportional to the standard free energy of evaporation, ΔG_o^{ev} , of the solvent from the binder, under the experimental conditions.

$$\Delta G_o^{ev} = \Delta H_o^{ev} - T \Delta S_o^{ev} = RT \ln K \dots\dots\dots(2)$$

$$K = c_{1l}/c_{1g} = (m_{1l} v_G)/(m_{1g} v_L) \\ \sim v_G/v_L = (v_G \rho_L)/m_L \equiv (V_r \rho_L)/m_L \dots\dots\dots(3)$$

$$\Delta G_o^{ev} = 2.3 RT \log V_r + 2.3 RT \log (\rho_L/m_L) \dots\dots\dots(4)$$

Because the second term in equation (4) represents a column constant which depends only on the density ρ_L and mass m_L of the stationary phase, values of $\log V_r$ may be used directly to give relative values for the evaporation equilibria for various solvents, provided the same column is used. When comparing the retention volumes of various solvents on a number of different binders, reference is made to the relative retention r_S with respect to a standard solvent.

$$\log r_S = \log V_r - \log V_{rS} = \frac{\Delta(\Delta G_o^{ev})}{2.3 RT} \dots\dots\dots(5)$$

From the temperature dependence of $\log V_r$, the standard enthalpy ΔH_o^{ev} and, using equation (2), the standard entropy ΔS_o^{ev} , of the evaporation of the solvent from the binder can be determined.

$$\log V_r = \frac{\Delta H_o^{ev}}{2.3 R} \cdot \frac{1}{T} - \frac{\Delta S_o^{ev}}{2.3 R} - \log (\rho_L/m_L) \dots\dots\dots(6)$$

The process of evaporation of the solvent from the binder has been separated into two terms by Purnell⁸: the demixing of solvent and binder molecules and the vaporisation of the pure solvent. The interaction of the solvent with the molecules of the stationary phase is included in the demixing term, and is usually expressed by the activity coefficient, γ^∞ , of the solvent, extrapolated to infinite dilution in the binder phase. This activity coefficient can also be determined by gas chromatography.

$$\Delta G_o^{ev} = \Delta G_o^{demix} + \Delta G_o^v \equiv \Delta G_o^v - \Delta G_o^{mix} \dots\dots\dots(7)$$

$$\Delta G_o^{mix} = RT \ln \gamma^\infty \dots\dots\dots(8)$$

$$\gamma^\infty = \frac{RT m_L}{p^\circ M_L V_r} \dots\dots\dots(9)$$

where p° and M_L are the saturation vapour pressure of the solvent and the molecular weight of the binder, respectively.

Experimental

Gas chromatographic measurements of the retention volumes of solvents on binder columns were carried out on an apparatus (model 116 E from Perkin Elmer) having an air thermostat and a thermal conductivity detector. Helium was used as the carrier gas with flow velocities between 40 — 100 ml \times min⁻¹. Tubes 1 and 2 metres in length, having internal diameters of 4.65mm and 3mm, were used as columns. Experiments were carried out using different commercial binders. Inert silica products (see Table 1) were used as supports. When an active support was required, a commercial zinc oxide pigment was used. The percentage loading of the support with binder material was varied in different experiments.

Preparation of the columns

The supports were immersed in dilute solutions of binders, so that after evaporation the desired loading was obtained. The solvents (butyl alcohol for the urea resin, white spirit for cyclised rubber, acetone for the vinyl acetate polymers and toluene, xylene, and butyl alcohol for the alkyd/melamine resin) were evaporated by agitation at low temperature in vacuum. Before use, these binder coated supports were dried, and in some cases hardened, in a controlled manner. They were then conditioned in the gas chromatograph under the most stringent experimental conditions that would be used. These processes of drying, and hardening and conditioning, were carried out for the individual cases as shown below.

Binder	Drying conditions.		Hardening/conditioning treatment	
	(in air)		(under helium)	
urea resin	1h	140°C	8h	100°C
cyclised rubber	1 day	90°C	8h	110°C
vinyl acetate polymers	1 day	20°C	8h	110°C
alkyd/melamine resin	1h	130°C	8h	130°C

When using zinc oxide as a support, the above procedure could not be employed because of the small grain size of the pigment (ca. 1-5 μm). The loading in this case was achieved by depositing the material on a substrate in the form of a thick film from a heavily pigmented paint. After removal from the substrate, the film was pulverised, screened and then conditioned as above in the gas chromatograph.

The extent of the loading of the support with binder was determined by measuring the loss of weight on ignition. The columns were filled under vacuum, using continuous vibration to ensure even packing. The amount of loaded support was determined in each case. Table 1 summarises the binder columns used and their corresponding characteristic data.

Table 1
Characteristics of polymer binder columns (length 2m)

No.	Binder/Support	Binder loading		Column pressure, mean value (torr)
		(g)	(% w/w)	
1	Polyvinyl acetate ^a /Chromosorb A ^b	4.41	20.3	950
2	Polyvinyl acetate/Chromosorb A	2.07	10.8	931
3	Polyvinyl acetate/zinc oxide ^c	8.42	20.6	1,007
4	Polyvinyl acetate/zinc oxide	4.84	12.1	1,178
5	Vinyl chloride/vinyl acetate copolymer 1 ^d / Chromosorb A	4.06	19.1	931
6	Vinyl chloride/vinyl acetate copolymer 2 ^e / Chromosorb A	4.00	19.9	923
7	Cyclic rubber ^f /silica gel ^g ; length 1m	0.68	17.0	1,710
8	Urea resin ^h /Chromosorb G ⁱ	0.68	3.1	1,140
9	Alkyd/melamine resin ^k /Chromosorb G	0.70	3.0	1,140

(a) Mowilith 20, mol.wt.ca. 40,000 (Farbwerke Hoechst), (b) Chromosorb A, NAW, 30-40 mesh (Johns Manville), (c) Zinkweiss Grünsiegel, grain size 1-5 μm (Zinkweiss Gesellschaft), (d) Vinnol H 40/55, 40% w/w vac (Wacker-Chemie), (e) Vinylite VYHH, 15% w/w vac (Union Carbide Corp.), (f) Alplex 450J (Chemische Werke Albert), (g) Kieselgel OS 0.92, 60-100 mesh (Perkin-Elmer), (h) Polyester modified urea resin, Plastopal AW/AT (50/50) (BASF), (i) Chromosorb G/AW, DMCS, 80-100 mesh (Johns Manville), (k) Short coconut oil alkyd, Alkydal C25 (Farbenfabriken Bayer)/Maprenal NPX (Casella), 75/25.

Gas chromatographic measurement

The retention volumes, V_r , on the binder columns shown in Table 1, were measured for a series of solvents at different column temperatures. The main experiments were carried out using the following solvents: aliphatic hydrocarbons: *n*-hexane (hex), *n*-heptane (hept), *n*-octane (oct), *n*-decane (dec); aromatic hydrocarbons: benzene (bnz), toluene (tol), *p*-xylene (xyl); esters: methyl, ethyl, and *n*-propyl formates (MeF, EtF and PF, resp.) and methyl acetate (MeAc); and alcohols: methanol (MeOH), ethanol (EtOH) and *n*-propyl alcohol (POH). Because the distribution equilibrium of the solvents between binder and carrier gas is frequently dependent on the concentration of the solvent in the gas, experiments were carried out using different sample volumes, and the results extrapolated to zero sample volume. Preliminary results were given in a previous communication⁷.

Experiments on an homologous series of solvents yielded a linear relation between $\log V_r$ and the number of carbon atoms in the solvent. This represents the constant increase in the standard free energy of evaporation $\Delta(\Delta G_o^{\text{ev}})$ for a single CH_2 group in the homologous series.

The agreement of values of the logarithm of the reduced retention volume, $\log V_{\text{red}}$, for pvac columns (Table 2) having different loadings with binder and supports of different activity, indicates that a true distribution equilibrium between binder and gas phase has been established and that the observed values have not been influenced by adsorption processes at the surfaces of the binder, the support or the column wall, and that the measurements are highly reproducible and are not influenced by the column filling procedure.

Table 2
Logarithms of the reduced retention volumes, $\log V_{\text{red}}$, for solvents on polyvinyl acetate at 100°C

Solvent	pvac on Chromosorb A(1) log m = 0.64	pvac on Chromosorb A(2) log m = 0.32	pvac on zinc oxide(3) log m = 0.92	pvac on zinc oxide(4) log m = 0.68
MeF ..	1.22	(0.91)	1.17	1.18
EtF ..	1.45	1.35	1.39	1.41
nPF ..	1.70	1.63	1.63	1.66
MeAc ..	1.43	1.39	1.41	1.40
Bzn ..	1.69	1.63	1.64	1.66
Tol ..	1.99	1.92	1.88	1.93
pXyl ..	2.22	2.22	2.15	2.17
nOct ..	1.23	1.26	1.20	1.29
nDec ..	1.75	1.81	1.68	1.80

$$\log V_{\text{red}} = \log V_r - \log m$$

Temperature dependence of retention volumes

The results of these measurements confirm the linear relationship between $\log V_r$ and the reciprocal of the absolute temperature required by equation (6). This may be seen from Fig. 1, which shows values of retention for several solvents on cyclised rubber. This fact indicates that, under the experimental conditions, the gas chromatographic measurements are based on a true equilibrium. Recent investigations by Guillet and co-workers⁹, have shown, however, that, for some polymeric materials, a true distribution equilibrium exists only above the glass transition temperature, T_g , at least for poor solvents. It is thus possible to use gas chromatographic measurements to determine the glass transition temperature of polymers⁹. The influence of the glass transition temperature on the region of linearity of the relation between $\log V_r$ and temperature may be also observed in some of the results of this work. For instance, in the case of vinyl acetate polymers, where T_g is a function of the amount of vinyl acetate present in the polymer, it is found that the boundary of the region of linearity of the graph of $\log V_r$ for *n*-dodecane versus $1/T$ lies at higher temperatures the higher the T_g of the polymer (Fig. 2). With regard to the interpretation of retention volumes in terms of the molecular interactions, only values within the linear temperature region were used.

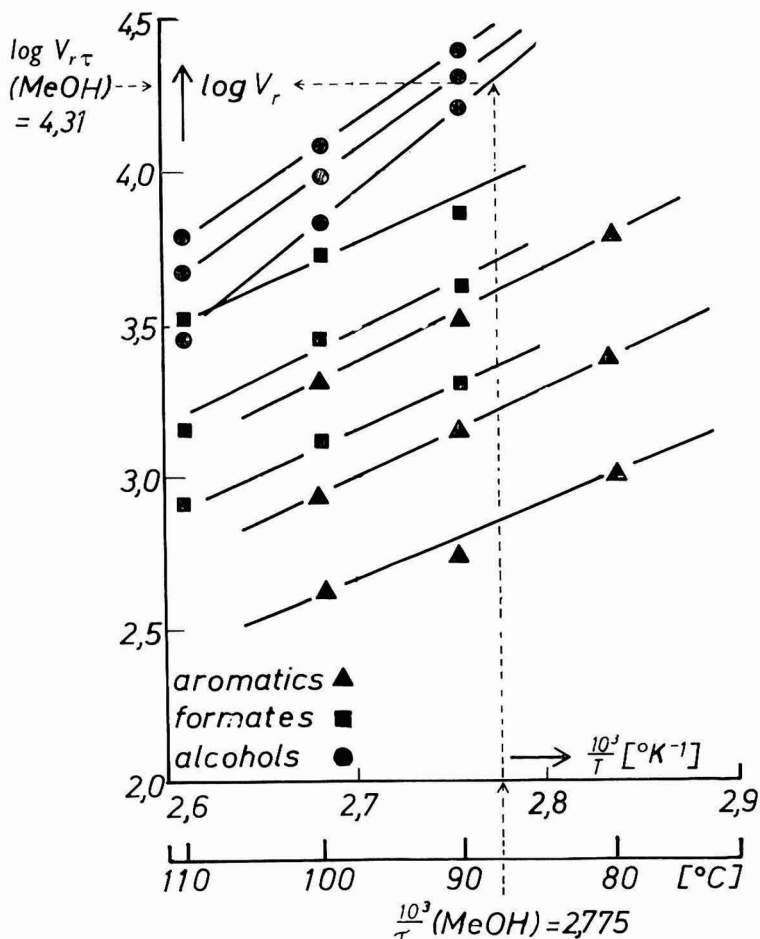


Fig. 1. Temperature dependence of logarithms of retention volumes, $\log V_r$, for solvents on 0.68 g cyclised rubber (7)

Results and discussion

As is shown from equation (4), values of $\log V_r$, or better still, of $\log V_{red}$ (Table 2), may be used to characterise the total force of interaction that must be overcome by solvent molecules during evaporation at a particular temperature from a binder/solvent system of low solvent concentration. In order to compare the total interaction forces for solvents evaporating from different binders, the logarithm of the relative retention, $\log r_s$, equation (5), should be used. Because the effects of vaporisation of the pure solvents are included in these values, some other equations dependent only on the inter-molecular forces between binder and solvent would be preferred for the prediction of properties of binder/solvent systems.

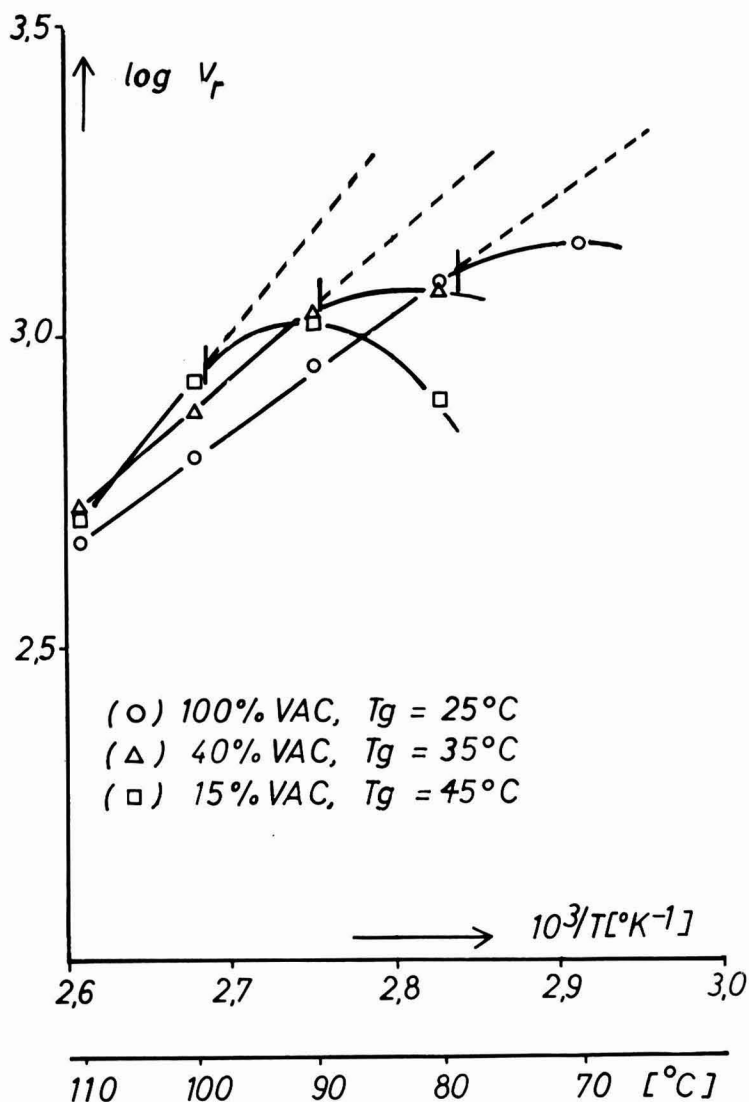


Fig. 2. Temperature dependence of logarithms of retention volumes, $\log V_r$, for n-dodecane on pvac (1), copolymers 1 (5) and 2 (6) with increasing glass transition temperatures

Characteristic indices for the (polar) interaction between binder and solvent

Kováts¹⁰, and also Kaiser¹¹, have introduced empirical parameters from gas chromatographic measurements, representing, on the basis of their definition, characteristic values for the polar interactions of volatile substances with a stationary phase.

Kováts (1959 paper) recommended the so called retention index I for the gas chromatographic retention properties of solvents on defined column materials.

I is based on the behaviour of linear aliphatic hydrocarbons having n and $n+2$ carbon atoms.

$$I = 200 \frac{\log V_r - \log V_{rn}}{\log V_{r(n+2)} - \log V_{rn}} + 100 n \dots\dots\dots(10)$$

Because volatile substances have different interactions on different stationary phases, they have correspondingly different values of the retention index I . Kováts proposed, therefore, that the retention index increment ΔI against a dipolar (p) and an apolar (a) standard phase increases proportionally with the polarity, in particular with the dipolar properties of the material investigated, providing that the same apolar phase is used.

$$\Delta I = I^p - I^a \dots\dots\dots(11)$$

From the investigation of the retention behaviour of volatile materials on a number of stationary phases in general use, Rohrschneider^{12, 13} examined the polarity scale as given by the retention index increments ΔI^{sq} of a series of materials with respect to squalane (2,6,10,15,19,23-hexamethyltetracosane) as the apolar stationary phase. He showed that the polarity scale varied from stationary phase to stationary phase and considered that the reason for this is to be found in the different proportions of the individual polar interaction components that make up the total polar interaction between the volatile material and the stationary phase.

The function F that was used by Kaiser¹¹ is built up from the difference of the retention indices for a chosen volatile material, I , and for the corresponding linear hydrocarbon, I_o , e.g. for acetone and n -propane, respectively, on the same stationary phase. F varies in a similar way as ΔI with the polar interaction forces between substances and the stationary phase.

$$F = I - I_o \dots\dots\dots(12)$$

A further empirical parameter has been used in this work for the characterisation of interaction forces between the binder and solvent. Equation (7) shows that the directly determined gas chromatographic retention volume include the vapourisation behaviour of the pure solvents, which depends on the particular experimental conditions. The logarithms of the retention volumes at a temperature τ , $\log V_{r\tau}$, where τ corresponds to the boiling point of the solvents at the mean column pressure, was determined graphically from the temperature dependence of the retention volumes. In this case ΔG_o^v in equation (7) becomes zero, so that $\Delta G_{o\tau}^{ev}$ is equal to $\Delta G_{o\tau}^{demix}$. As a first approximation to the theory, the author has used the $\log V_{r\tau}$ values, in a limited temperature range, as characteristic constants for the interaction, although, in reality, as shown in a later example (Table 5), ΔG_o^{demix} varies somewhat with temperature.

The behaviour of several solvents with cyclised rubber is represented in Table 3 by the values of $\log V_{r\tau}$. In contrast to the directly determined values of $\log V_r$, values of $\log V_{r\tau}$ increase with the increased proportion of polar to non-polar portions of the molecules accompanying a decreased number of carbon atoms in an homologous series. In just the same way, it is found that, with cyclised rubber containing polar groups resulting from the production process, $\log V_{r\tau}$ values for different classes of substances increase from the aliphatics

to the aromatics and further to the esters and alcohols. This means that $\log V_{r\tau}$ is really representative of the polar interaction between the solvent and binder. As shown in Fig. 3, these values therefore correlate in order of magnitude better with Hansen's polar solvent solubility parameter δ_a than with Hildebrand's solubility parameter δ . This is especially true for the alcohols.

Table 3
Gas chromatographic data for solvent retention on 0.68 g cyclised rubber

Solvent	Boiling temperature		$\log V_r$ (90°C)	$\log V_{r\tau}$	δ_{25}	δ_{a25} (Hansen)
	T (°C)	τ (°K)				
MeOH ..	64.7	360.5	4.20*	4.31	14.28	12.40
EtOH ..	78.3	374.0	4.34*	3.97	12.92	10.45
<i>n</i> POH ..	97.2	393.0	4.39*	3.49	11.97	9.10
MeF ..	31.5	329.5	3.30	4.08	10.05	6.66
EtF ..	54.0	353.0	3.63	3.92	9.55	6.10
<i>n</i> PF ..	81.2	383.0	3.88	3.51	9.56	6.09
Bzn ..	80.2	394.0	2.78	2.39	9.15	1.48
Tol ..	110.8	412.0	3.15	2.17	8.91	2.18
<i>p</i> Xyl ..	138.4	459.7	3.52	1.66	8.75	2.30
<i>n</i> Hex ..	69.0	372.0	2.08	1.97	7.24	(0)
<i>n</i> Hept ..	98.4	402.2	2.40	1.79	7.40	(0)
<i>n</i> Oct ..	125.8	431.7	2.75	1.68	7.60	(0)

*Extrapolated from $1/T$ plot

In Table 4, results of gas chromatographic retention experiments for solvents on polyvinyl acetate are summarised, together with Hansen's polar solvent solubility parameter δ_a and with Hildebrand's solubility parameter δ . In addition to the retention indices I_{100} , which are representative of the evaporation equilibrium at 100°C, experimental values of $\log V_{r\tau}$, of $\log r_{10\tau}$ with *n*-decane as a standard substance, and of F_{100} and ΔI_{100}^{sq} are shown; these characterise the (polar) interaction forces between the solvents and polyvinyl acetate. Again, these values increase with a decreasing number of carbon atoms in an homologous series. They indicate, also, a stronger interaction of the binder with esters and alcohols than with aromatics and aliphatics. The solvent scales with respect to the different indices, $\log V_{r\tau}$, $\log r_{10\tau}$, F_{100} and ΔI_{100}^{sq} , however, vary somewhat in the range of the alcohols. A large number of experiments is necessary to estimate which of these parameters would be the most representative of the characterisation of the molecular interaction. Further reported values of F_{100} , from Kaiser, were obtained from measurements on polyethylene glycol adipate¹¹ and are in good agreement with the author's results on the chemically quite similar polyvinyl acetate.

Gas chromatographic measurements of the retention behaviour of a number of solvents with respect to different binder materials were carried out, using three vinyl acetate polymers that have progressively larger amounts of vinyl acetate in the polymer chain. In Table 5, differences in the $\log r_{oct\tau}$ values of

solvents with the individual polymer, for copolymer 1, containing 40 per cent w/w vinyl acetate, and with copolymer 2, having a content of vinyl acetate of 15 per cent w/w, are presented, *n*-octane being used as a standard substance. It is shown that these values increase, especially for the esters, with an increased amount of vinyl acetate in the binder, indicating increased interactions of these substances with the polymer.

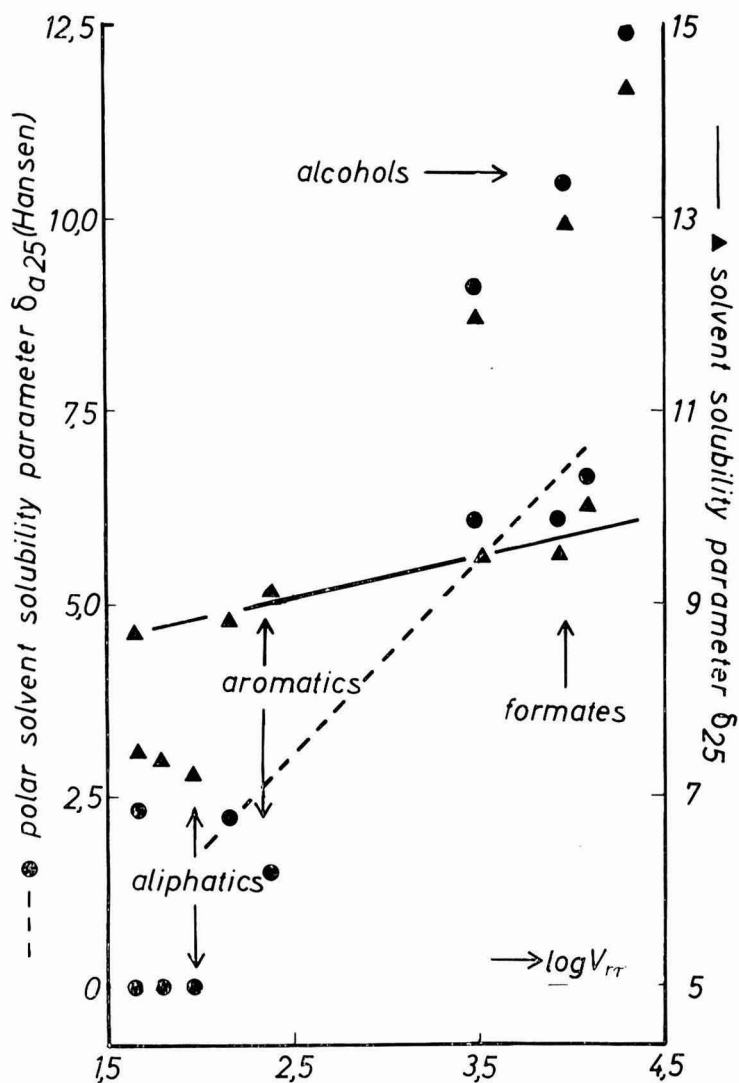


Fig. 3. Correlation of gas chromatographic data for solvent retention on cyclised rubber (7) with solvent solubility parameter, δ , and with polar solvent solubility parameter increment, δ_a

Table 4
Gas chromatographic data for solvent retention on 4.41 g polyvinyl acetate

Solvent	I_{100}	$\log V_{r\tau}$	$\log r_{10\tau}(a)$	F_{100}	$\Delta I_{100}^{sq}(b)$	$F_{100}(c)$ (Kaiser)	δ_{25}	δ_{a25} (Hansen)
MeOH ^(d)	891	2.43	+0.76	791	580	819	14.28	12.40
EtOH ^(d)	939	2.42	+0.75	739	544	767	12.92	10.45
<i>n</i> POH ^(d)	1,038	2.49	+0.81	738	527	774	11.97	9.10
MeF	799	2.67	+1.00	599	439	—	10.05	6.66
EtF	883	2.57	+0.90	583	417	584	9.55	6.10
<i>n</i> PF	979	2.48	+0.81	579	410	597	9.56	6.09
MeAc	877	2.55	+0.88	577	403	601	9.49	5.72
Bzn	978	2.47	+0.80	378	315	419	9.15	1.48
Tol	1,090	2.42	+0.75	390	317	437	8.91	2.18
<i>p</i> Xyl	1,178	2.28	+0.61	378	298	436	8.75	2.30
<i>n</i> Oct	800	1.53	-0.14	0	0	0	7.60	(0)
<i>n</i> Dec	1,000	1.67	0	0	0	0	6.60	(0)

(a) $\log r_{10\tau} = \log V_{r\tau}i - \log V_{r\tau}n\text{-decane}$

(b) using I_{100}^{sq} values, measured on a squalane column, from Kaiser¹¹

(c) values measured on polyethylene glycol adipate¹¹

(d) experimental values from measurements on a second column¹⁴

Table 5
Relative data for gas chromatographic solvent retention on polyvinyl acetate binders with different contents of vinyl acetate $R_{oct\tau}$

Solvent	Copolymer 2 15% w/w vac	Copolymer 1 40% w/w vac	Polyvinyl acetate 100% w/w vac
MeOH*	0	—	+0.43
EtOH*	0	—	+0.40
<i>n</i> POH*	0	—	+0.49
MeF	0	+0.41	+0.86
EtF	0	+0.21	+0.69
<i>n</i> PF	0	+0.11	+0.48
MeAc	0	+0.15	+0.50
Bzn	0	-0.03	+0.32
Tol	0	-0.08	+0.13
<i>p</i> Xyl	0	-0.30	-0.07
<i>n</i> Oct	0	0	0
<i>n</i> Dec	0	-0.27	-0.52

* From measurements on a second column

$R_{oct\tau} = \log r_{oct\tau}(\text{polymer } i) - \log r_{oct\tau}(\text{copolymer } 2)$

Thermodynamic data of the interaction between binder and solvent

As is also shown by Guillet et al.⁹, absolute thermodynamic data of the interaction forces of the solvents with a binder can be obtained from gas chromatographic measurement, provided that some physical properties of the binder are known. Using equations (8) and (9), activity coefficients γ^∞ and free energies of mixing ΔG_o^{mix} for three solvents on polyvinyl acetate as the stationary

phase, the pva having a molecular weight, M_L , of about 40,000, were evaluated from gas chromatographic experiments at various temperatures. In Table 6 the corresponding free energies of demixing at 100°C, $\Delta G_o^{demix} = -\Delta G_o^{mix}$, see equation (7), are shown. The increase in positive values of ΔG_o^{demix} indicates increased interaction forces of solvents with polyvinyl acetate from *n*-octane through benzene to methyl formate. Under the same experimental conditions, the enthalpies of demixing, ΔH_o^{demix} and the entropies of demixing, ΔS_o^{demix} , were determined from the temperature dependence of $\ln \gamma^\infty$, and from an equation similar to equation (2), respectively.

Moreover, the corresponding thermodynamic data for the process of evaporation of the solvents from polyvinyl acetate, which has a density at 100°C of 1.15g ml⁻¹, were also evaluated from gas chromatographic measurements, using equations (2) to (4). Data regarding the process of vaporisation of the pure solvents have then been obtained from equation (7).

As shown in Table 6, gas chromatographic values of the enthalpy and the entropy of vaporisation of the pure solvents, ΔH_o^v and ΔS_o^v respectively, are in good agreement with values reported in the literature.

Table 6

Thermodynamic data for polyvinyl acetate(1)/solvent systems at 100°C from gas chromatographic measurements

	Methyl formate	Benzene	<i>n</i> -Octane
Boiling temperature τ (°C) at mean column pressure	37.0	88.0	133.0
Evaporation of solvent from binder:			
ΔG_o^{ev} (kcal mol ⁻¹)	+ 2.18	+ 2.99	+ 2.20
ΔH_o^{ev} (kcal mol ⁻¹)	+ 6.60	+ 8.75	+ 6.60
ΔS_o^{ev} (cal deg ⁻¹ mol ⁻¹)	+ 11.8	+ 15.4	+ 11.8
Demixing of solvent and binder:			
$-\Delta G_o^{mix}$ (kcal mol ⁻¹)	+ 3.76	+ 3.46	+ 1.67
$-\Delta H_o^{mix}$ (kcal mol ⁻¹)	+ 0.30	+ 1.37	+ 1.44
$-\Delta S_o^{mix}$ (cal deg ⁻¹ mol ⁻¹)	- 9.30	- 5.60	- 8.30
Vaporisation of solvent:			
ΔG_o^v (kcal mol ⁻¹)	- 1.58	- 0.47	+ 0.53
ΔH_o^v (kcal mol ⁻¹)	+ 6.30	+ 7.38	+ 8.04
ΔS_o^v (cal deg ⁻¹ mol ⁻¹) ^(a)	+ 21.1	+ 21.0	+ 20.1
Reported value of ΔH_o^v (kcal mol ⁻¹) ^(b) ..	+ 6.96	+ 7.18	+ 8.65

(a) Trouton constant: 23.3 (cal deg⁻¹ mol⁻¹)

(b) calculated from Watson equation¹⁵ and reference¹⁶

Effect of the individual interaction forces

From the values of the gas chromatographic solvent retention on different binders given in Table 5, it may be seen that the solvent scale with respect to these values differs from binder to binder. This result is to be seen more clearly from Fig. 4, where ΔI_{100}^{sq} values of solvents on polyvinyl acetate and on cyclised rubber, respectively, are plotted against the polar solvent solubility parameter, δ_a , of Hansen. Whereas δ_a has a constant value for each substance, ΔI_{100}^{sq} values, which should be representative of the total polar interaction between the solvents and binders, are actually influenced by the binder. In addition to this, the shape of the graph of ΔI_{100}^{sq} versus δ_a varies from binder to binder.

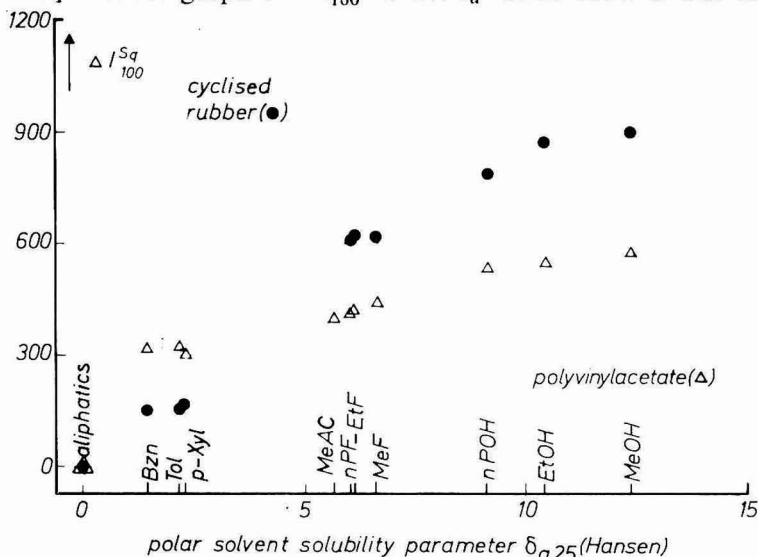


Fig. 4. Correlation of gas chromatographic data for solvent retention on different polymer binders with polar solvent solubility parameter increment, δ_a

As Rohrschneider^{12, 13} has deduced from gas chromatographic experiments on a number of common stationary phases, the total interaction of solvents with the stationary phase comprises at least five different kinds of polar interaction, which are represented by the retention behaviour of benzene as a π — electron donor, of methyl ethyl ketone (MEK) as a dipolar substance, of nitromethane as an electron acceptor, of pyridine as a n -electron donor and a proton acceptor, and of ethanol as a proton donor, respectively. The results shown in Fig. 4, are probably due to the different proportions of the individual polar interaction components with respect to the different binders. In order to confirm this assumption, retention index increments, ΔI_{100}^{sq} , were calculated from retention indices I_{sq}^{100} of some typical solvents (e.g. benzene, methyl ethyl ketone, acetone, ethanol and methanol) on typical stationary phases, which were reported by Kaiser¹¹. For the calculations, the following stationary phases were chosen: polyethylene glycol (Carbowax 4,000) as a n -electron donor (11)*, polyethylene glycol adipate as a dipolar substance (30), dimethyl

* The numbers in brackets refer to the curves in Fig. 5.

stearamide (Hallcomid M 18) as a proton acceptor (33), sorbitol as a proton donor (64) and a nitrile silicone (XF 1150) as a strongly dipolar substance (76). In Fig. 5, ΔI_{100}^{sq} values were plotted against Hansen's polar solvent solubility parameter δ_a . Again, changes in the shape of the curves, representing the different proportions of the distinct interaction components, have been obtained on the individual stationary phases.

From recent gas chromatographic experiments (Table 7), using typical solvents on various binder materials, similar results were found, indicating the effect of different proportions of the individual polar interaction components that yield the total polar interaction of the solvents with binders.

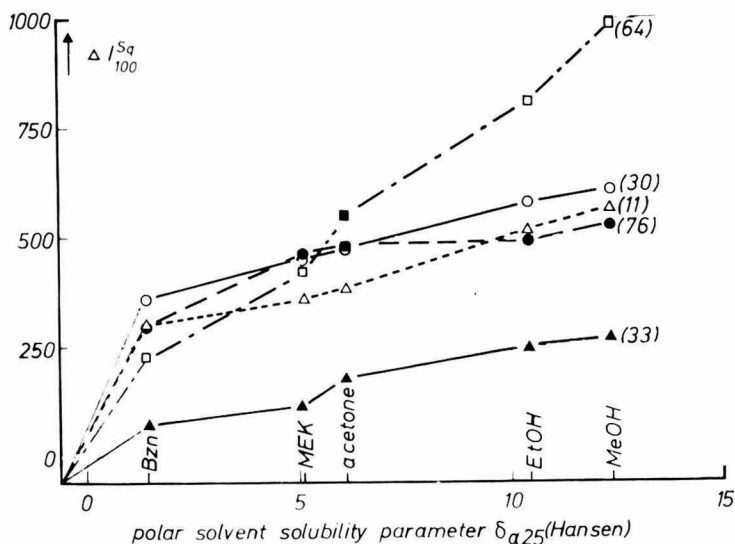


Fig. 5. Correlation of calculated retention index increments, ΔI_{100}^{sq} , for typical solvents on typical gas chromatographic stationary phases, with polar solvent solubility parameter increment, δ_a (ΔI_{100}^{sq} values calculated from reported I_{100}^{sq} , Kaiser¹¹)

Table 7

Correlation of experimentally obtained retention index increments ΔI_{100}^{sq} for typical solvents on different polymer binders

Binder	ΔI_{100}^{sq} (a)				
	<i>n</i> -Decane	Benzene	MEK	Nitro methane	EtOH
Alkyd melamine resin	0	191	253	381 (b)	341
Polyvinyl acetate(1)	0	315	—	—	580
Urea resin ..	0	362	460	637 (b)	605
Cyclised rubber ..	0	148	—	—	878

(a) I_{100}^{sq} values from R. Kaiser¹¹

(b) $I_{100}^{sq} = 501$ ¹⁴

Conclusion

From the above experiments it has been shown that characteristic empirical indices, and even absolute thermodynamic data, indicating the total interaction of solvents with binders can be obtained by means of gas chromatographic measurements of the solvent retention volumes on binders as a stationary phase in the column. As has been further shown, these quantities, obtained from direct measurements on concentrated binder/solvent systems, also take into consideration the different proportions of the individual polar interaction of solvents with each binder, whereas the solvent solubility parameter is a constant for each solvent regardless of the binder used. When dealing with a multi-component parameter system, reported by Rohrschneider to exist for the interaction of solvents with the stationary phases commonly used in gas chromatography, it should be possible to predict the unknown properties of binder/solvent systems by performing only a few additional experiments.

Acknowledgment

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Discussion at the Torquay Conference

MR J. BOROKY remarked that Dr Reichert had said that gas chromatographic investigation might be of assistance in determining the compatibility of binders. Had he any experience of this type of investigation?

DR K. H. REICHERT said that he was not sure if the compatibility of polymers could be determined, but it was possible to measure the change in polymer/solvent interaction when changing, for example, from one homogeneous copolymer to another containing the same monomer constituents but having a different monomer ratio.

MR BOROKY said that the method of investigation quoted was valid only at infinite dilution when the binder was at infinite dilution in the solvent, so that the physical laws were followed. Under normal conditions, mixtures or dispersions were being dealt with, where the concentrations were much higher. To what extent could correction factors be used?

DR REICHERT replied that the values obtained directly from gas chromatographic measurements were generally dependent on the concentration of the solvent in the inert gas phase. As the measurements were carried out at low solvent concentrations and as the experimentally obtained values were extrapolated to zero solvent concentration, the method was valid for the practical case of a dried paint film, where the solvent concentration in the binder was also extremely low. In the case of a wet paint film, solvent concentrations in the binder were somewhat higher. By gas chromatographic measurements at higher solvent concentrations, at which the distribution equilibrium was changed, this fact could be taken into consideration.

DR D. H. SCHÄRER asked what was the lowest temperature at which the polymer/solvent interaction could be measured. Was it possible to measure it at room temperature by the gas chromatographic method?

DR REICHERT said that gas chromatographic measurements could, in principle, be carried out at low temperatures when the gas chromatographic equipment was fitted for low temperature control. From theoretical considerations it followed, however, that the basic distribution equilibrium existed only above the glass transition temperature, but similar experiments below the glass transition temperature had been reported by Canadian authors, who studied, in this region, the equilibrium of adsorption of solvents on solid polymer surfaces.

DR W. FUNKE (Chairman) said that this work had been carried out by Professor Guillet, who had shown that, with the help of gas chromatography, it was possible to determine crystallinity and glass transition temperatures.

It was, of course possible to measure the influence of temperatures above the glass transition range by changing the temperature of the gas chromatographic column.

DR REICHERT added that the left-hand section of the reciprocal temperature dependence diagram (Fig. 2) represented the behaviour of solvents at higher temperatures. In this region a true equilibrium, the absorption equilibrium, existed. On the right-hand side, at low temperatures, there existed an adsorption equilibrium. In the medium field of glass transition it was possible to obtain information about the diffusion of solvents in polymers.

MR A. R. H. TAWN remarked that a previous speaker had asked whether it was possible to use this technique to investigate the compatibility of polymer pairs. He and co-workers had carried out research in this field, and the only meaningful results obtained with polymer pairs had been when the polymer pairs were compatible. When they were incompatible the results had been completely unreproducible. The technique used had been developed some years ago, and differed from Dr Reichert's method because the Kovats Retention Index was not then known. It had therefore been necessary to have a highly reproducible technique for packing the column in

order to get comparable results between runs. The technique used consisted of using mixtures of polymers to coat ballotini in a uniform manner. Using this technique no meaningful results had been obtained for incompatible pairs.

He thought it was possible to infer a good deal about the potential compatibility of a pair of polymers if thermodynamic parameters of each one were measured independently; if they matched reasonably closely it could be assumed that, at all but the very highest molecular weights, they were likely to be compatible.

DR FUNKE stated that an important point when considering compatibility of polymers was that there were both low molecular weight and high molecular weight types. The determining factor in the high molecular weight types was the enthalpy, while the determining factor in the lower molecular weight types was mainly the entropy. This was, perhaps, an explanation of why only the compatibility of lower molecular weight resins could be determined by this method.

MR TAWN added that a most important factor in this technique, as Dr Reichert had brought out, was that it was a very simple method of separating the terms of the free energy equation. Hitherto entropy had been ignored, but individual terms for enthalpy and entropy could now be obtained by this technique, which was very easy to use.

MR A. KJELLIN said that a lot of work on solubility parameters had been reported in the last decade, and in his experience the paint industry had been very slow in adopting the results. Possibly this was a communication problem, which meant that the scientists should explain their findings in more clear terms. Dr Reichert had said that it would be possible to foresee some important properties of a paint film by using this technique. Did this mean that it could be of value in determining formulations and film properties which were of immediate interest?

DR REICHERT replied that the number of experiments performed at the present time was not sufficient to give a full scheme of values for many types of solvents and binder materials, so no predictions could be made at present. When information from more experiments was available, it should be possible to predict the behaviour of a new binder in binder/solvent systems from gas chromatographic retention experiments with the five typical solvents representing the different individual interaction forces, for instance with regard to solvent evaporation and solubility properties. At present, such work was being carried out.

DR R. BULT asked if it would also be possible to take this work further so that it would give a measure of solvent retention in paint films. The solvent retention of paint films had, of course, been determined by other gas chromatographic techniques, and he was interested to know if Dr Reichert had any ideas in this direction.

DR REICHERT agreed that this was possible, but only above the glass transition point. In the region below this point, solvent retention would be dependent upon other factors, especially upon effects which cause hindrance of solvent diffusion in paint films, such as the shape of solvent molecules and the distance between chain segments of polymer molecules in the film.

DR BULT agreed and asked if there was any information on the glass transition temperatures of normally-used paint layers in the solid state, for example, air drying alkyd resins.

DR REICHERT replied that he had performed gas chromatographic experiments using copolymers with different amounts of vinyl acetate. The glass transition temperatures of polyvinyl acetate (25 C), of a copolymer with 40 per cent of vinyl acetate (35 C) and of a copolymer with 15 per cent of vinyl acetate (ca 45 C) were expressed in the reciprocal temperature dependencies of the gas chromatographic values for solvents: the points of deviation from the linear relationship increased with the increased glass transition temperatures of the polymers.

MR T. R. BULLETT inquired if the application of these methods to the adhesion of solution-type paints had been considered. He suggested that, in general, increased interaction between solvent and polymer molecules was likely to give polymer configurations favourable to improved contact between the polymer and substrate and therefore better adhesion. On the other hand, this same increased interaction would mean that, for the same molecular weight, there would be a lower concentration of polymer at a given viscosity and thus a lower film build, a result less attractive to paint makers. The adhesion of the polymer to the substrate was also dependent upon the residual amounts of solvents in the film, since some attractive groups on the polymer would be saturated by the solvent. There was experimental evidence that adhesion of solution polymers changed over a long period after application. Films of high molecular weight materials could initially be stretched and peeled off very easily, but as time passed there was a continuous improvement in adhesion, presumably because solvent molecules were leaving the interface, allowing more polymer/substrate bonds to develop.

DR REICHERT agreed that this was a very interesting point of view. No such investigations had yet been carried out.

DR FUNKE said that in principle it must be possible to obtain some indications for film adhesion. Pigment materials could be used to fill the gas chromatographic columns, and the retention of solvents on the pigments determined. Powdery material with a similar composition to the surfaces of metals could also be used, for example, iron oxide. From comparison of the retention values of binders in the same solvent on these two systems, it should be possible to obtain information on adhesion, at least in principle.

DR REICHERT referred to some experiments using pigment-filled columns. In logarithmic plots of the retention volumes of solvents, related to the specific surface area of the pigment, linear relations upon the energies of wetting were obtained.

DR FUNKE said this work was part of an extensive study going on in Stuttgart. For example, the surface energy of various different materials, particularly pigments, was being measured. The aim was to obtain some indication of the dispersibility of pigments in certain vehicles from these energy values. The conditions in these experiments were, of course, not as complicated as they were in practice. Very clean materials were used, whereas in paint films many additives might be present which could interact at the interface. However, the study of the surface activity of pigments would produce values which could be related to the interaction parameters obtained by gas chromatography. By comparing the surface energy values and the interaction values some information on the criteria affecting the dispersion of pigments in vehicles might be obtained.

Next month's issue

The Honorary Editor has accepted the following papers from the Torquay Conference for publication, and these are expected to appear in the October issue:

"Film appearance and its dependence on solvent formulation; a more systematic approach," by *D. H. Schärer and L. A. Tysall*

"Basic lead silicochromate," by *R. P. Bates*

"Opacity of roller coatings: Part I—Practical aspects," by *D. G. Dowling and D. F. Tunstall*.

Reviews

REVIEWS IN MACROMOLECULAR CHEMISTRY, VOL. 4

EDITED BY G. B. BUTLER AND K. F. O'DRISCOLL. New York: Marcel Dekker Inc. 1970, pp. viii + 419. Price £8.35

This volume, which follows its predecessors in general standard and presentation, covers seven topics. These are: Polymer enzymes and enzyme analogues (Lindsey), Stability of polycarbonate (Davis and Golden), Cross-linking—effect on physical properties of polymers (Nielsen), Synthesis of thermally stable polymeric azomethines by polycondensation reactions (D'Alelio and Schoenig), Dehydrochlorination and stabilisation of polyvinyl chloride (Mitsuo Onozuka and Mitsuo Asahina), Recent advances in the development of flame-retardant polymers (Delman), and Thermodynamics of polymerisation (Hideo Sawada).

We have here a nice blend of the special with the general. The general must be absorbed by all with pretensions to current awareness in any area of polymer science. The special will naturally be of more limited interest unless one is seeking to extrapolate far into the future. Thus, the coatings scientist will naturally turn to the sections on cross-linking, pvc, flame retardant polymers, and polymerisation thermodynamics.

Nielsen is as clear and concise as ever. His contribution is rather weighted towards the behaviour of rubbers, but his discussion of the characteristics of cross-linked materials is little short of impeccable. He provides a most useful review of the effects of cross-link density on chemical behaviour, swelling, elastic modulus, creep, glass transition and stress-strain behaviour. He is at pains to emphasise the oft-forgotten observation that, below T_g , most properties are insensitive to the nature of the cross-linked structure: we have much to learn of the factors affecting behaviour in the glassy state.

Careful reading of the Japanese contribution on pvc shows that, notwithstanding the widespread investigations which have been undertaken on pvc stability, our understanding of this much-used and long-standing polymer is far from complete.

Delman is not alone as a recent writer on the topic of flame retardancy in polymers, but his review is salutary in pointing out the still dominant role of additives, which often create as many problems as they solve. There are several unresolved and competing theories of the mechanism of flame retardancy yet to be argued.

This reviewer found the chapter on thermodynamics of polymerisation a mixed bag. He felt that the discussion of ceiling temperature gave insufficient credit to the classical work of Dainton and Ivin (though it was referenced), but at the same time was gratified to find such a useful presentation of experimental methods and tables of data for this and heats of polymerisation. The final part of this chapter, on polycondensation, appeared disappointing. Granted, the thermodynamics of polycondensation are little different from those of simple condensation, but the complications of ring formation are dismissed in 10 lines by reference only to Stockmayer and Jacobson's work

of 1950; Harris and Kilb apparently do not exist. Moreover, the effect of unequal reactivity, discussed at length by Case, is not mentioned, any more than the important recent contributions of Gordon. One concludes, unavoidably, that the author is less at home when discussing polycondensation than when dealing with addition polymerisation.

This is a most useful series which is obviously maintaining the high standard originally set. Those who have the earlier issues will certainly want to acquire subsequent volumes; those who acquire Volume 4 will be greatly inspired to order back—as well as future—issues.

A. R. H. TAWN

MICROBIAL ASPECTS OF METALLURGY

EDITED BY J. D. A. MILLER. Aylesbury: Medical & Technical Publishing Co. Ltd. 1971, pp. 202. Price £3.40

This book is based on a course given by its eight authors at UMIST on the role of micro-organisms in metallurgical processes. It forms a fascinating introduction to an area on the boundaries of biology, chemistry and corrosion science that involves expensive destructive processes but also has potentially favourable economic interest. All biological growth processes involve organisation of matter, a loss of entropy which must be balanced by degradation of the environment to a higher entropy state—metallic corrosion is one such degradative process and not surprisingly is often associated with microbial activity, even when the primary source of energy is some other chemical reaction.

The two most important biochemical effects discussed in this book are the reduction of sulphate by, for example, *Desulfovibrio Spp.* and the oxidation of sulphur and, by some species only, ferrous iron by *Thiobacillus Spp.* The sulphate reducers thrive in anaerobic conditions, for example in heavy clay or submarine mud, and accelerate corrosion mainly, it is now considered, because hydrogen is consumed in the reduction reaction, thus depolarising the cathodes; in the worst conditions—soils of low resistivity and redox potential more negative than +400mV with respect to the normal hydrogen electrode—heavy cast iron pipes can be perforated in one year. The *Thiobacilli* are almost all aerobic but can be equally destructive through developing high concentrations of sulphuric acid. Bacteria are not the only source of trouble. Fungi and algae can promote corrosion by establishing regions of differential aeration, purely by physical blockage, and acidic by-products of micro-biological growth can also attack many substrates. *Cladosporium resinae*, a mould with a strange appetite for paraffins, which has given rise to corrosion in aircraft fuel tanks, is particularly troublesome in that it appears to eat through many organic coatings as well. But not all life is hostile—the last two chapters of the book tell how *Thiobacillus ferro-oxidans* is already helping to reclaim hundreds of thousands of tons of copper from low grade ores by oxidising sulphides to water-leachable sulphates, and point the way to many other reclamation processes that may become worthwhile as primary high grade metal ores become exhausted.

Summing up, this little book is interesting general reading for any industrial scientist. For the corrosion technologist or the engineer concerned with protection of materials in any natural environment, there is much to be noted. Not least important is the emphasis on adaptability of micro-organisms. Some *Thiobacilli* can thrive on pH 0.5, whilst others can adapt to tolerate copper in concentrations up to 30,000ppm. Too generous use of biocides can result in development of resistant species.

Chapters by different authors vary in depth of treatment—the best, for example microbial corrosion of buried and immersed metals, are fully documented with an extensive bibliography, but for most serious readers this should be only an introductory text.

T. R. BULLETT

Courses available 1971-72

Details of courses dealing with surface coatings technology have been received from a number of colleges, and these are shown below. It is emphasised that this list is not fully comprehensive, as it has not been possible to obtain information from all the relevant colleges.

Manchester Polytechnic—John Dalton Faculty of Technology

Department of Polymer Technology, Chester Street, Manchester M1 5GD.

The City and Guilds Paint Technicians Certificate Parts I and II. The Higher National Certificate Endorsement Subject, (for LRIC purposes) "Chemical technology with special reference to polymers and surface coatings."

The Associateship of the Society of Dyers and Colourists, (including Branch 5 relevant to paints, lacquers and printing inks).

Enrolment for the above courses will take place at the college on Monday 13, Tuesday 14 and Wednesday 15 September, during the periods 10.00 a.m.—12.00 p.m., 2.00 p.m.—4.00 p.m., and 6.00 p.m.—8.00 p.m.

In addition to the above part-time courses, a short course of a general nature will be held during Easter 1972.

Those requiring further information on any of the above courses should contact the Head of Department, Dr V. G. Bashford, at the above address, or discuss the matter with members of staff during enrolment week.

Polytechnic of the South Bank

Department of Chemistry and Polymer Technology, Borough Road, London SE1.

BSc Honours Degree in Chemical Technology. A four-year sandwich course with specialisation in polymers, and plastics or surface coatings. Higher National Diploma in Applied Chemistry, a two year full-time course with alternative specialisations including one in polymer science and technology.

Licentiatehip of the Royal Institute of Chemistry, one-year part-time courses leading to Certificates of Supplementary Studies in surface coatings, polymer chemistry or polymer science and technology. (For LRIC purposes.)

Certificate of the Polytechnic in Surface Coatings Technology.

Principles of Surface Coatings Technology, an intensive two-day full-time course, to be held in May 1972.

Enrolment for part-time courses will take place on Tuesday 14 September between the hours 9.00 a.m.—12.00 a.m. and 5.30 p.m.—8.00 p.m. Those interested in full-time courses, or requiring details of any course, should contact Dr B. E. Weller at the address given above.

East Ham Technical College

Department of Science, High Street South, London E6.

The City & Guilds Paint Technicians Certificate Parts I and II.

The Higher National Certificate endorsement in surface coatings leading to LRIC.

The City & Guilds Paintmakers Certificate.

“Essentials of paint technology” a 30 week part-time course intended to give a technical grounding to non-technical staff in the paint and associated industries.

Enrolment for these courses will take place on Monday 13, Tuesday 14 and Wednesday 15 September between the hours 9.00 a.m.—5.30 p.m. and 6.30 p.m.—9.30 p.m. Full details of the courses may be obtained from Mr S. W. Stuart, Head of the Department of Science, at the above address.

London College of Printing

Elephant and Castle, London SE1.

Higher National Diploma in printing, a three year sandwich course.

Printing ink technology (Advanced) leading to the City & Guilds Printing Ink Technicians Advanced Certificate.

City & Guilds Printing Ink Ordinary Certificate, City & Guilds Printing Ink Technicians Certificate.

Ordinary National Certificate in printing.

Higher National Certificate in printing.

Several other short courses are offered, mainly for scientists and technologists in the printing and printing ink industries. Full details of all courses and application forms for the full time course may be obtained from the Senior Administrative Officer at the above address; enrolment for part-time courses will take place from Monday 13 September to Friday 17 September at 6.00 p.m.

Information Received

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

The Association was pleased to be represented at the recent opening of the new polymer laboratories at the **Polytechnic of the South Bank** by the President of the Plastics Institute, Sir Harry Melville. The total floor area of the new laboratories and associated facilities is almost 13,000sq.ft, and this includes two laboratories devoted entirely to surface coatings: one to manufacturing aspects, and the other to testing. A number of courses of interest to coatings technologists are offered, and details of these appear elsewhere in this issue.

The Color Systems Division of **Kollmorgen Corporation** has introduced a new computer compatible automatic colour measuring instrument, the *KCS-18 Automatic Colour-Eye*. The new instrument is claimed to be the lowest priced instrument of its type capable of providing tristimulus and spectrophotometric data for both fluorescent and non-fluorescent materials. Four-filter tristimulus measurements are provided automatically in under 15 seconds, and 19 point spectrophotometric measurement is obtained in less than 50 seconds.

Faserit, a fibrous spray coating material for textured wall covering is now available through approved contractors of the manufacturer, **Faserit (Great Britain) Ltd.**, a member of the **Ault & Wiborg Group**. Applied in a three-part process by a special spray-gun, the coating can be applied direct to brick or block-work, thus dispensing with the need for plastering.

New editions of the "Seibt Export Directory of German Industries," in English, French and Spanish, have been published by **Seibt-Verlag**. The book gives the German suppliers for a wide variety of products, together with lists of trade names. Copies may be obtained from Seibt at a cost of 44 DM including postage.

Fabwerke Hoechst AG has recently introduced *Hansa Orange RN 01*, a new, improved quality of Permanent Red GG. Advantages claimed for the new pigment are higher tinctorial strength, yellowness, brightness, gloss and transparency than Permanent Red GG or the earlier *Hansa* pigment, *Orange RN*; it is expected to be of particular interest to the printing ink industry.

The construction of the new **Lennig Chemicals Ltd.** plant at Seal Sands, Teesside, is on schedule, it has recently been announced, and the first phase of the project will be ready for commissioning in November. Lennig expects the project to be completed by summer 1972 and to be operating at full capacity the following year.

Vinyl Products Limited has announced that the manufacture and marketing of its synthetic resin emulsions and solutions to all users in Germany, Holland, and Scandinavia, and to the paint industry in Belgium and Luxembourg, is to be undertaken by the **Scado** group of companies, all of which, like Vinyl Products, are part of **Unilever's** Polymers Division. Scado will market these products in Austria, Poland, Russia and East Germany. Vinyl Products state that significant market penetration is foreseen in the territory to be covered by Scado, owing to the commercial and technical strength resulting from the close links now established between the two companies.

The *Panel-Rak*, a stainless steel rack for drying of sample panels, is now available in the UK from **Wentworth Instruments Limited**. Manufactured by **Atlas Electric Devices Company** in the USA, the *Panel-Rak* will hold fifteen prepared panels for air-drying or baking without any sticking, scratching or marring of the panel surfaces.

Two new innovation studies are now available from **Arnold Services**. Entitled "New ideas for surface coating polymers" and "Fusion coating by electrostatic deposition," both studies were written by Dr R. L. Smith, and can be obtained at a price of £400 each.

A series of printing inks that dry on exposure to ultra-violet radiation has been introduced by **Fishburn Printing Ink Company Ltd**. Based on the company's *Ultraking* range of inks, the inks are formulated on a photosensitive machine, and dry almost instantaneously on exposure to UV radiation of the correct spectral characteristics. Of particular advantage in sheet-fed work, the ink has been demonstrated by Fishburn on a two-colour litho press operating on paper and board at speeds of 6,000 iph with drying on the press. No stacking of sheets to allow curing is required, and since no solvent evaporation is involved, the problems of pollution control encountered with heat-set inks are not encountered. This UV cure technology can be applied equally to letterpress printing, and Fishburn are carrying out work on overprint varnishes, functional coatings and flexo/gravure inks.

Over 500 enquiries were received on the **Albright & Wilson Ltd**. stand during OCCA 23 at Olympia in June. Nearly 60 per cent of the enquiries were from overseas, involving visitors from 23 countries including the USA, Australia and all of the West European countries.

This response was some 50 per cent higher than the company had at OCCA 22, and considerable penetration into overseas markets for its **Accomet C** and **Corinac** products is expected as a result.

The **Paints Division of ICI Ltd**. has recently undergone a major reorganisation. The division has now been formed into three distinct business groups, paints, wall-coverings, and the **Hyde group**, which manufactures coated fabrics and composites.

At a press conference given to introduce the board of the reorganised division, ICI was quoted as being the largest paint manufacturer in Europe, and about the third largest in the world; sales of £100 million per annum were confidently expected by the end of the decade. The increasing share of the market being captured by the company's vinyl wallcoverings was stressed; in the long term it was foreseen that usage of dispersymers would increase, particularly in automobile applications, and that combinations of dispersymers with the plastic manipulations being produced by the **Hyde Group** would lead to interesting composite materials.

An open day was held recently at the new £2 million distribution centre for **ICI Ltd. Dyestuffs Division** at Heywood, Lancs, which will centralise the storage and despatch of goods manufactured at all the Division's factories and will shortly deal with the whole of the new **Organics Division** when **Nobel Division** is merged with **Dyestuffs**.

The warehouse, which is a single storey building, covers an area of 450,000sq.ft and will accommodate approximately 25,000 tons of material, amounting to about 500,000 individual packages and covering some 3,500 products. Storage is in drums upon wooden pallets housed in long rows of tubular steel racks reaching to a height

of 32ft. Stacking to this height is achieved by the use of specially designed high reach fork lift trucks.

In addition to this main warehouse, there is a separate store covering 40,000sq.ft for flammable materials and 56,000sq.ft of open air storage. It is expected that the warehouse will have a daily throughput of about 1,000 orders amounting to around 1,200 tons.

The Distribution Administration Department is housed as a separate unit on the same site. Orders are received by teleprinter via the computer controlled switching centre at Piccadilly Plaza (Manchester) and from there via the computer installation at Hexagon House, Blackley.

Heywood was chosen as the site for the distribution centre because it is within a few miles of the most strategic position with regard to factories, customers and ports, because suitable land was available and because of its proximity to existing and projected motorways. ICI has a sufficient area to double the size of the warehouse.

The *Super-plus* range of polyurethane gloss and vinyl-gel emulsion paints has been introduced by **Carson-Paripan Ltd.** The polyurethane gloss is available in 12 shades, as well as black and brilliant white, and contains a high percentage of polyurethane to promote chip resistance and silicone to reduce dirt retention. The vinyl-gel emulsion is produced in matching shades with a satin finish.

The names of some of the water repellent wood preservatives marketed by **Preservation Developments Limited** have been revised, it was recently announced. In future the pigmented water repellent finishes for hardwoods for exterior cladding and industrialised timber buildings will be known as *Timseal Red* (formerly *Super Timproof*), and the colourless version as *Timseal Clear* (formerly *Super Timseal*).

The *Timproof* range will then consist of water repellent primers, named *Timproof Clear*, *Timproof Pink* and *Timproof White*.

Silver Paint and Lacquer Co. Ltd. has announced that it has purchased Cross Bank Mills in Batley, Yorks. This will add 47,000sq.ft to the company's warehousing and distribution depot.

Derussol 345 is a new aqueous pigment dispersion from **Degussa**. Intended for use in paper and light-weight building board, the new product contains 45 per cent furnace black, and, it is claimed, has a particularly good degree of dispersion and colouring power.

Nopco Hess Ltd. has been appointed exclusive distribution and service agents for *Dion 3-800 LC*, a catalytic resin for rapid-cure epoxy adhesives, manufactured in the USA by the **Diamond Shamrock Chemical Co.** A mercaptan terminated liquid polymer, *Dion 3-800 LC* promotes rapid curing in amide/epoxy and polyamide/epoxy systems, both in thin films and at low temperatures. Full technical data is available from Nopco Hess.

Section Proceedings

Scottish

Eastern Branch

Modern developments in titanium dioxide pigments with particular reference to printing inks

The third meeting of the session was held in the Carlton Hotel, Edinburgh, on Wednesday 16 December, with Mr P. Gower in the chair. Mr D. G. Dowling of British Titan Products Ltd. spoke on "Modern developments in titanium dioxide pigments with particular reference to printing inks."

Mr Dowling opened his talk by stating that modern pigments were chemical products produced to close specifications and citing the requirements for titanium dioxide pigments. The theoretical basis of opacity was then discussed and the optimum pigment particle size calculated.

The two processes for the manufacture of titanium dioxide were discussed; the sulphate process gave a product containing trace impurities and with poorer colour, and the chloride process gave a very pure product, but had not given all the advantages originally hoped for.

Mr Dowling went on to describe the four possible modifications of titanium dioxide: dry milling, in which the dry pigment was dry ground and the required particle size selected; coating and refining, in which dry milled pigment was coated by wet slurring and precipitating aluminium or silicon on to the surface; coating and super-refining, manufactured as the previous type, but to closer specifications and heavy coating, in which a higher percentage of inorganic oxide was precipitated on to the pigment surface.

Mr Dowling concluded this most interesting talk by describing the influence of the printing stock, emphasising that in most applications a printing ink was not printed in a sufficiently thick film to give true opacity.

J.H.S.

Money-go-round

The fourth ordinary meeting of the session was held in the Carlton Hotel, Edinburgh, on Wednesday 20 January, with Mr P. Gower in the chair. Mr M. Burt of the Scottish Stock Exchange gave a talk entitled "Money-go-round."

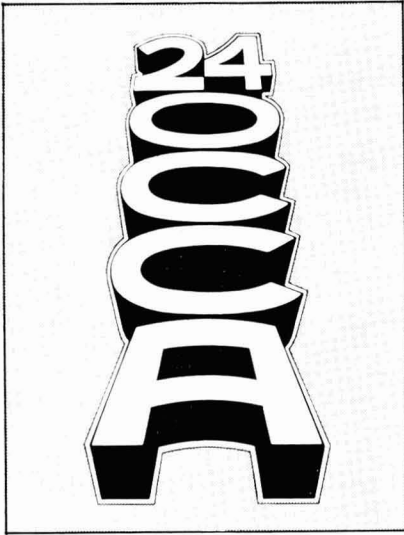
A short film was shown which outlined the procedure necessary for a private company to go public. The film particularly emphasised the thorough investigation into the company's financial soundness conducted by the Stock Exchange before that company was allowed to offer shares to the general public and obtain a Stock Exchange quotation.

Then Mr Burt briefly traced the history of the Stock Exchange. In the 11th Century the De Medici family who, up till that time, had been merchants, offered a loan to the people of Florence. Interest was paid on the loan, and the De Medici thus became the first moneylenders. To this day the three brass balls, the coat of arms of the De Medici family, were hung outside pawnbrokers' shops.

The first Stock Exchange as it was known today was opened in Amsterdam, and this was followed shortly afterwards by the London Stock Exchange which opened in 1744. Other Stock Exchanges were opened in different parts of the country soon afterwards.

The remainder of the meeting was devoted to questions and a vigorous discussion period took place.

J.H.S.



Technical Exhibition

17-21 April 1972

As previously announced, copies of the Invitation to Exhibit for the Twenty-fourth Technical Exhibition, to be held at the Empire Hall, Olympia, London, W14 from 17-21 April 1972, were despatched in July.

Resulting from the success of OCCA-23, many companies, both from the UK and overseas, have already applied for stand space. Any company, therefore, wishing to exhibit is urged to contact the Director and Secretary immediately at the address shown on the front cover as the closing date for applications is **1 October 1971**.

The times when the Exhibition will be open are given below:

Monday	15.00-18.30
Tuesday	09.30-18.30
Wednesday	09.30-18.30
Thursday	09.30-18.30
Friday	09.30-16.00

Interpreters for French, German, Italian and Spanish will be in attendance throughout the period of the Exhibition and all the overseas visitors are asked to sign the Visitors' Book at the Information Centre.

The Association is making arrangements with various hotel groups for hotel accommodation for visitors to the Exhibition and information will be published in the *Official Guide*.

Institution of a Professional Grade for Ordinary Members

Members will be aware that Council set up a Working Party on Education, Training and Qualifications in 1967 (as part of its Forward Thinking exercise).

The report of this Working Party was adopted in principle at the Council Meeting on 28 January 1971. Subsequently, Council appointed a Committee, composed of members whose interests embraced all aspects of technical education in the industries covered by the Association, to institute the scheme for an optional professional grade for Ordinary Members, as proposed by the Working Party.

The Members of this Committee—to be known as the Professional Grade Committee—are:

Mr A. W. Blenkinsop (President)
 Mr A. S. Fraser (Immediate Past President)
 Mr T. W. J. Apperley
 Dr S. H. Bell
 Mr H. J. Clarke
 Mr R. R. Coupe
 Mr S. R. Finn
 Dr H. W. Keenan
 Mr A. McLean
 Dr W. M. Morgans
 Mr A. T. S. Rudram
 Dr F. M. Smith
 Mr A. R. H. Tawn
 Mr C. J. A. Taylor

Council feels that members will appreciate having details not only of the final recommendations of the Working Party but also of those parts of its Report that clarify the Council's intentions in instituting this professional grade.

Extracts from the Report

1. *The problem*

From time to time since the foundation of the Association in 1918 there has been a feeling that some kind of professional

grade should be available to members (e.g. Chapter 5 of the *History of OCCA*) but, on the other hand, there has been the fear expressed that to introduce such a scheme would of necessity alter the whole character of the Association. The Working Party, therefore, examined ways in which an optional scheme could be introduced with the minimum alteration to the existing organisation and at the same time giving flexibility so that not only could the Section Committees overseas advise on educational facilities in their countries (in order to establish comparable standards) but also consideration could be given to exemptions by examinations taken through other institutions in the UK and overseas.

2. *The intention*

The Working Party particularly wish to emphasise that its whole thinking has been directed towards the younger member who at present might, after successfully taking certain qualifications in technology and chemistry (e.g. City & Guilds or Higher National Certificate), wish to obtain the same sort of educational identity with his industry as is available in other similar industries. In this way it was suggested that many more young men of good technical standard would either be attracted to the industry or remain therein, to the benefit of both the industry and the Association. The Manchester Section Survey, as published in *JOCCA* 1968, 51, 359, showed strikingly that membership of the Association does not seem to have the same appeal to those under 30 as to those over 30—a finding supported in other Sections' surveys. It follows, however, from the institution of a Licentiate grade for younger members, that the progression (to Associate and Fellow) would have to be made clear. Existing Ordinary Members could then apply, if they so wished, for entry into the professional grade at a higher level. For example,

Council feels that Members will like to know that it is suggested that the following shall be immediately invited to apply for admission into the Professional Grade:

All Past Presidents of the Association.

Members of the Working Party on Education, Training and Qualifications.

Members of the Professional Grade Committee.

3. *The scheme in outline*

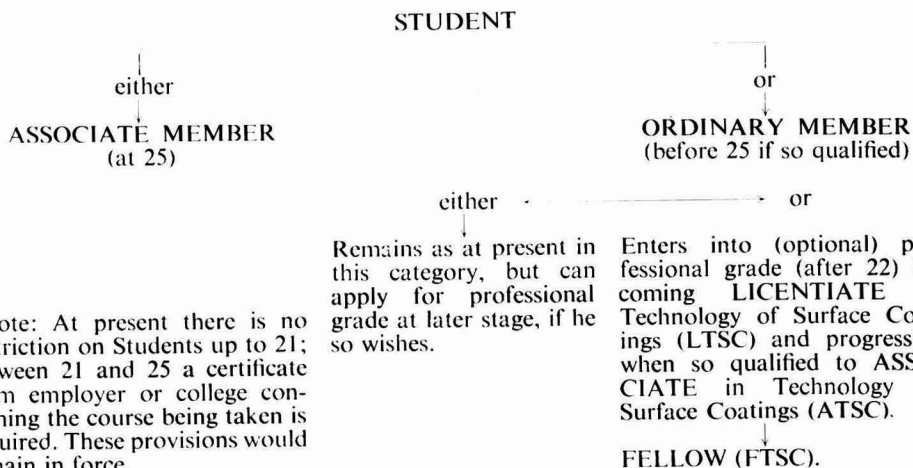
(a) Since it follows that a member wishing to apply for entry in the professional grade must already be eligible for the Ordinary Membership class, it is not necessary to alter the present membership structure of the Association, but only to offer an *optional* grade to those Ordinary Members who wish to apply. In the case of a Student wishing to take up Licentiate-ship it follows that he must have already obtained the necessary qualifications to enable him to enter the Ordinary Membership category in any case. Further it must be recalled that under Articles 6 (iv) and 8 a Student is not a member of the incorporated association and has no voting rights, so that it would be logical for a Student to transfer to the Ordinary Membership category before applying for Licentiate-ship of the Association.

(b) Except for the use of designatory letters, the Ordinary Members who are admitted to the professional grade will have no additional rights, will remain in the Ordinary Membership category but will be denoted in both the Association's and Sections' records or membership lists by designatory letters. (The present membership categories Ordinary, Associate, Honorary would remain with the (non-member) Students and it will be recalled that a Retired Member remains in the same category as he was in on retirement but without any loss of the rights attached to that category.)

(c) The name of the Association will not be changed and designations will be as follows:

- (i) Fellow (of the Oil and Colour Chemists' Association) in the Technology of Surface Coatings, designated FTSC.
- (ii) Associate (of the Oil and Colour Chemists' Association) in the Technology of Surface Coatings, designated ATSC.
- (iii) Licentiate (of the Oil and Colour Chemists' Association) in the Technology of Surface Coatings, designated LTSC.

The following diagram shows the suggested progression of a Student (up to 25 years of age)*.



*Note: At present there is no restriction on Students up to 21; between 21 and 25 a certificate from employer or college confirming the course being taken is required. These provisions would remain in force.

4. *The scheme in detail*

(a) *Procedure*

(i) *Institution of the Professional Grade:*

In order to obtain the widest possible support and interest for this scheme, copies of this statement will be distributed to appropriate organisations and colleges. Council seeks the co-operation of all senior members of the Association, particularly in encouraging younger personnel to apply for entry to this grade.

(ii) *Regulations:* Regulations for admission to the various classes of the professional grade are set out as an Appendix to this report. It is stressed that these may not be wholly appropriate to the Sections in South Africa and New Zealand, but the advice of these Section Committees will be sought for regulations which will be acceptable equivalents in those countries. Already correspondence has been received from the overseas Sections about the indigenous examinations and courses.

It will be noted that the regulations state that admissions to the Licentiate grade and in some cases to the Associate grade shall attend a viva voce examination where, *inter alia*, the breadth and depth of the candidate's technical experience will be examined. Clearly, many candidates in the United Kingdom and Ireland will be examined in London but there will be candidates who by reason of geographical situation will be examined locally under the supervision of the Committee. Similarly, a candidate submitting a dissertation or wishing to be examined on a very specialised subject might be referred by the Professional Grade Committee to assessors. Abroad, in the three Sections arrangements will be made for viva voce examinations to be held with results referred to the Professional Grade Committee in London and for the General Overseas Section; the co-operation of distinguished members of that Section in various centres will be sought.

Eventually it is intended to agree the syllabi for courses at some technical colleges with written examinations

moderated by the Professional Grade Committee, but for reasons given in the preceding paragraph it will clearly not be possible to have written examinations for every centre or for every candidate.

(iii) *Certification Fees:* Since the emphasis is on the Licentiate grade, these candidates (particularly Students) will pay only a nominal fee, since they must first transfer (from Student to Ordinary membership) at the relevant age shown in the regulations, and would thus in any case pay the Ordinary Membership subscription. It is stressed that, after paying the fee for the particular grade, the Ordinary Members would then pay only the annual membership subscription applicable to Ordinary Members. Council has decided that the Certification Fees shall, in the first instance be as follows: Licentiate £1.00; Associate £3.00; Fellow £5.00.

A remittance for the appropriate fee must accompany each application and, in cases of application for those grades requiring viva voce examinations and for assessments of theses or dissertations, the fees paid will be non-returnable, but where an application for the Fellowship grade is unsuccessful only a proportion of the fee will be retained to cover the cost of administration.

(iv) *Recognition of the Professional Grade:* The Association has taken steps to discuss the institution of a professional grade with interested organisations and feels confident that this step will be welcomed both by the industries and the educational institutions. Lists of the names of Ordinary Members in the Professional Grade will be published each year in the December issue of the *Journal*.

(v) *Organisation and sponsorship in the first year:* Each Ordinary Member entering the professional grade will receive a numbered certificate which will be returnable for cancellation on transfer from one grade to another or on resignation from the Association. On resignation from the Association, the entitlement to use designatory letters ceases immediately.

Although the scheme states that applicants have to be sponsored, this will be waived for the first year (until October 1972) for existing Members and Students.

Application forms for candidates for entry into the Professional Grade are available from the Director & Secretary at the Association's offices, and one is enclosed in each copy of this issue being sent to Members. After completion, the forms should be returned, together with the appropriate remittance, to the Director & Secretary at the Association's

offices (except in the case of those Members attached to the Auckland, South African and Wellington Sections, who should address their forms to their Section Hon. Secretaries).

Students wishing to apply for entry into the Professional Grade must first make application in writing for upgrading to Ordinary Membership, giving the reasons for their eligibility for such regrading. Applications, together with the appropriate remittance, should be addressed as for application for admission to the Professional Grade.

Appendix

Regulations for admission to the Professional Grade, September 1971

Note: For the sake of simplicity, at this stage reference is made only to UK examinations etc., but equivalent qualifications overseas will naturally be accepted.

A. Licentiate

1. Shall be an Ordinary Member of the Association and have been an Ordinary Member or Student of the Association for not less than one year.
2. Shall have attained the age of 22.
3. (a) Shall be a Licentiate of the Royal Institute of Chemistry in Coatings Technology (*viz.* Higher National Certificate + Endorsement in coatings technology + 1 year approved experience in the science or technology of coatings after passing the endorsement exam).

OR (b) Shall be a Licentiate of the Royal Institute of Chemistry in another relevant subject such as advanced analytical chemistry, colour chemistry or polymer science, and shall have two years approved experience in the science or technology of coatings since so qualifying.

OR (c) Shall hold the Full Technological Certificate of the City & Guilds of London Institute in a relevant subject as approved by the Professional Grade Committee

and shall have two years approved experience in the science or technology of coatings since gaining the FTC.

OR (d) Shall have passed Higher National Certificate or Higher National Diploma with three years approved experience in the science or technology of coatings since qualifying, but two years approved pre-qualification experience shall be deemed equivalent to the third post-qualification year.

OR (e) Shall be graduate in relevant subject with not less than 1 year approved experience.

OR (f) Shall have passed such other qualifications as approved by the Professional Grade Committee from time to time.

4. Shall be required to satisfy the Professional Grade Committee, or some other body approved by the Professional Grade Committee in a viva voce examination and submit a dissertation on a topic previously approved by the Professional Grade Committee.

5. Shall normally be sponsored by three Ordinary Members of the Association in the professional grade (either Associate or Fellow) at least one of which must be a Fellow.
6. Shall have paid the fee stipulated by the Council and have paid the current subscription payable by an Ordinary Member.

B. Associate, being already a Licentiate

1. Shall, since his election to the Licentiateship, have practised the science or technology of coatings for not less than three years.
2. Shall provide evidence acceptable to the Professional Grade Committee of his superior professional skill and maturity.
3. Shall hold the City & Guilds of London Institute Insignia Award OR shall submit a thesis or dissertation of comparable level on a topic previously approved by the Professional Grade Committee OR shall have published work which, in the opinion of the Professional Grade Committee, is of comparable merit.
4. MAY be required to satisfy the Professional Grade Committee or some other body as approved by the Professional Grade Committee in a viva voce examination.
5. Shall normally be sponsored by three Ordinary Members of the Association in the professional grade (either Associate or Fellow) at least one of which must be a Fellow.
6. Shall have paid the fee stipulated by the Council and have paid the current subscription payable by an Ordinary Member.

C. Associate, not already a Licentiate

EITHER

1. Shall be not less than 24 years of age.
2. Shall be an Ordinary Member of the Association and have been an Ordinary Member or Student of the Association for not less than two years.

3. Shall hold the Graduateship of the Royal Institute of Chemistry or Institute of Physics or a University or Council of National Academic Awards degree recognised by the Royal Institute of Chemistry or Institute of Physics as giving full exemption from the Graduateship Examination.
4. Shall have not less than two years approved post-graduate experience in the science or technology of coatings.
5. Shall normally be required to satisfy the Professional Grade Committee, or some other body approved by the Professional Grade Committee, at a viva voce examination.
6. Shall normally be sponsored by three Ordinary Members of the Association in the professional grade (either Associate or Fellow) at least one of which must be a Fellow.
7. Shall have paid the fee stipulated by the Council and have paid the current subscription payable by an Ordinary Member.

OR

1. Shall be not less than 30 years of age.
2. Shall be an Ordinary Member of the Association and have been an Ordinary Member of the Association for not less than three years.
3. Shall have been engaged in practising the science or technology of coatings for not less than seven years and shall have attained a position of considerable standing in the industry.
4. Shall normally be required to satisfy the Professional Grade Committee in viva voce examination of his professional competence.
5. Shall normally be sponsored by three Ordinary Members of the Association in the professional grade (either Associate or Fellow) at least one of which must be a Fellow.
6. Shall have paid the fee stipulated by the Council and have paid the current subscription payable by an Ordinary Member.

D. Fellow

Note: This is the senior award of the professional grade and signifies that the holder has made outstanding contributions to the science or technology of coatings or has reached a position of eminence in the industry through the practice thereof. The Professional Grade Committee will require substantial evidence of professional maturity in the science or technology of coatings although commercial experience will be taken into account in assessing the merits of candidates.

1. Shall be not less than 33 years of age.
2. Shall have been an Ordinary Member of the Association for not less than five years.
3. Shall be engaged in a position of superior responsibility in the coatings industry.

4. EITHER (a) shall have been an Associate of the professional grade for at least eight years
OR (b) shall have not less than fifteen years' experience of the science or technology of coatings in a position of superior responsibility.
5. Shall submit, with his application, an account of his experience, with due reference to scientific and technological interests, achievements and publications.
6. Shall normally be sponsored by three Ordinary Members of the Association in the professional grade, all of whom must be Fellows.
7. Shall have paid the fee stipulated by the Council and have paid the current subscription payable by an Ordinary Member.

All correspondence regarding the Professional Grade should be addressed to:

R. H. Hamblin, MA, FCIS, Director & Secretary,
Oil and Colour Chemists' Association,
Wax Chandlers' Hall, Gresham Street,
London EC2V 7AB.

OCCA Australia

The report for the year 1970-71 of the President of Oil and Colour Chemists' Association Australia, Mr D. M. D. Stewort, has recently been received, and it is felt that Members will be interested in the following extracts.

"With this annual meeting, the second year of this Executive's term of office comes to a close. Many of the predictions and comments I made in last year's report have now come to fruition but many still remain. However, progress has been made with, I think, a notable success in the launching of a new surface coatings course in Sydney. It is my hope that we shall form a Technical Education Sub-Committee at this annual meeting to

co-ordinate and advance our educational activities in other states. Other industry service activities of this type must now be found to further develop the image of OCCA Australia within the industry.

During this period and especially through our educational activities, a closer relationship has been developed between OCCAA and the Australian Paint Manufacturers Federation. Through our industry service activities I hope this "dialogue" will be further strengthened in the future.

A prospectus has been drafted and, if accepted at this meeting, should prove useful in attracting new members by giving a summary of the Association's

activities and benefits of membership.

Like most other associations and companies, we are meeting the problem of rising costs. So far the position has been held fairly well through the benefit of rising interest rates. Postage increased significantly during the year, contributing to the P & N deficit. When a new capitation fee is negotiated for *JOCCA* at the end of the next year the fee can be expected to rise sharply. At that time the incoming Committee will have to consider raising subscriptions.

P & N now needs some help to regain advertising accounts. All Sections should do what they can to assist in this regard.

All are now familiar with the third edition of the Raw Materials Index. Corrections and additions have recently been returned and the big job of collation and printing will soon be under way. The RMI has proved very popular within the industry and many copies have been sold. It is hoped that this pleasing trend will continue.

Last year's work on technical education has borne fruit with our new surface coating technology course. Reports on an "OCCA guide to coating surfaces" and a prospectus have been completed. This year it is expected that OCCA will launch their proposals for professional membership grades in the Association. The Forward Planning Sub-committee

will thus encounter a very big job in making recommendations for considering a similar system in Australia. These recommendations may be expected to involve a lot of proposals with regard to our ideas on OCCA International and how the professional grades may be dovetailed into the system. Forward Planning is also involved in making recommendations on how OCCA Australia should be involved in Standards Association Committees. Such representation is potentially another area where OCCAA may be of service to industry.

Through these two years we have endeavoured to maintain the AFC's position as an advisory body with the major activities being arranged at Section level. This system seems to be well established and operating successfully. Through the introduction of the "President's Page" in P & N, I have tried to keep members informed on those AFC affairs which would be of general interest. Possibly this type of communication could be used more beneficially in the future.

Finally, I would like to thank members and committee members, for the favour given me to serve them as President. It has been a privilege to do so and I have sincerely tried to fulfil the obligations of the office

D.M.D. STEWART."

OCCA and West Riding Golf Trophies

The competition for the OCCA and West Riding Golf Trophies was held at Ilkley Golf Club on 11 June 1971.

The trophies were again won by Mr A. Bridgewood of Laporte Industries Ltd. and the runner-up was Mr R. E. Frost of Sandoz Products Ltd.

This was a very close finish, with Mr Bridgewood and Mr Frost both having

the same number of points. Mr Bridgewood, however, gained the majority of his points during the last nine holes and the club professional decreed him the winner.

Fifteen players took part in the competition and although played in very wet conditions, it was a very enjoyable outing.

D.M.



The Chairman of the West Riding Section, Mrs K. Driver, presents Mr A. Bridgewood with the OCCA and West Riding golf trophies, watched by the runner-up, Mr R. E. Frost.

Hull Section

Dinner Dance

The annual Dinner Dance of the Hull Section will be held at the Hotel Eden, Willerby, Nr. Hull, on Friday 8 October 1971, reception 7.00 p.m.

Application for tickets should be made to:

The Hon. Soc. Secretary,
Hull Section,

The Oil and Colour Chemists' Association,
3, Briarfield Road, National Avenue,
Hull, Yorks.

Telephone: 482 41441 (Day) 0482 400525 (Evening).

Requests for accommodation should be made direct to the Hotel Eden. Telephone: 0482 652616.

News of Members

Mr D. C. Timpson, an Ordinary Member attached to the London Section, has been appointed chief chemist of Ripolin Ltd. Mr Timpson will be in charge of the company's Southall laboratories, and his responsibilities will include research and development.

Mr E. P. Gotham, an Ordinary Member attached to the Thames Valley

Section, who was previously chief chemist at Ripolin, is now based in France with Ripolin Georget Frietag.

Chemical Society Summer School in Rheology

Preliminary arrangements for a summer school in 1973 on the subject of rheology have been announced by the Chemical Society. The summer school will be held at the School of Pharmacy, London on

9-13 April 1973, and will cover both basic principles of rheology, and applications to specific systems; at present it is intended to deal with dispersed systems—such as emulsions, suspensions and pastes—plastics and rubbers, and solids, both

crystalline and amorphous.

Further details, when available, may be obtained from Dr M. D. Robinson, Education Officer, The Chemical Society, Burlington House, Piccadilly, London W1V 0BN.

Obituaries

Dr T. I. Kyle

Ian Kyle's very many friends will be shocked to hear of his death on 5 April in Christie Hospital, Manchester, after a short illness. He was 46 and leaves a widow and two children at school, to whom we offer deepest sympathy.

Mr Kyle received his academic training at Heriot Watt College and Edinburgh University, where he obtained his PhD. Apart from a short spell at ICI Billingham before going to Edinburgh University, his first post in industry was with Monsanto Chemicals Ltd. at Ruabon, from where in 1956 he went to British Titan Products Ltd. as a Senior Technical Officer. In 1964 he moved to Hardman & Holden Ltd. as Head of the Pigment and Surface Coating Departments, and in 1968 was appointed Group Director of Research and Development for SCC Colours Ltd.

Whilst in Wales he was a council

member of the RIC. In 1968 he was elected to the committee of the Manchester Section, and in 1970 was elected Hon. Treasurer of the section.

Cremation was at Stockport Crematorium on 10 April and contributions in place of flowers were to the Cancer Research Fund, Christie Hospital, Manchester.

Mr W. M. Aldous

It is with regret that we announce the death of Mr W. M. Aldous on 3 May in Scotland. Mr Aldous, who was a member of the Association for many years until his retirement from business in 1961, was a director of Burrell & Co. Ltd. for 14 years and joint managing director for five years.

He took a great interest in the work of the Paint Research Station, and was chairman of the technical panel dealing with pigments for a number of years.

Register of Members

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

Ordinary Members

STRUMHAUS, JERRY M., Bonaval GMBH, Bruhlerstr. 2-20, 53 Bonn, W. Germany.
(Overseas)

Associate Members

BEG, ARSHAD, Taba Chemie Co., 88W Takhte Jamshid Eve, Tehran, Iran.
(Overseas)

RIZZOTTO, RINO, Cabot Corporation, 1 Via Winckel Mann, Milano, Italy.
(Overseas)

VAQUERO MORILLO, JOSE, Industrias Proa. SL, Apartado 280, Vigo, Spain.
(Overseas)

Forthcoming Events

Details are given of meetings in the United Kingdom up to the end of the month following publication, and in South Africa and the Commonwealth up to the end of the second month.

Tuesday 14 September

West Riding Section: "Flexographic printing inks" speaker to be announced, to be held at the Griffin Hotel, Boar Lane, Leeds at 7.30 p.m.

Wednesday 15 September

Manchester Section—Student Group: "Formulation of two-pack epoxy paints" by Mr A. McWilliam (England, Hughes Bell & Co. Ltd.), to be held at the Manchester Literary and Philosophical Society at 4.30 p.m.

Friday 17 September

Irish Section: "The efficient use of TiO_2 pigments in organic compounds" by Mr J. G. Hoogerbeets of NV Titaandioxydefabriek Tiofine, to be held at the Clarence Hotel, Wellington Quay, Dublin, at 8.00 p.m.

Midlands Section: Ladies' Evening, to be held at the Westbourne Suite, Botanical Gardens, Birmingham, at 7.00 p.m.

Thursday 23 September

Thames Valley Section: "The Consumers' Association—the aims and achievements" by Mrs A. Williams of the Consumers' Association, to be held at the Beech Tree Hotel, Maxwell Road, Beaconsfield, Bucks, at 7.00 p.m.

Friday 24 September

Midlands Section: "Chlorinated rubber marine paints" by Mr C. G. Reid of General Chemicals Group, Imperial Chemical Industries Ltd., to be held at the Birmingham Chamber of Commerce and Industry, PO Box 30, 75 Harborne Road, Birmingham B15 3DH, at 6.30 p.m.

Friday 1 October

Scottish Section—Student Group: "The properties of anti-corrosive pigments" by Mr J. Bowden of Craig-Hubbuck Ltd., to be held at the St. Enoch Hotel, Glasgow, at 10.00 a.m.

Monday 4 October

Hull Section: "Practical pigment dispersion" by Mr J. R. Groom and Mr M. Baker (Sandoz Products), to be held at the Queen's Hotel at 7.00 p.m.

Tuesday 5 October

Thames Valley Section—Student Group: "Metal pretreatment" by Mr P. Burden of Pyrene Ltd., to be held at the Main Lecture Theatre, Slough College, at 4.00 p.m.

Thursday 7 October

Bristol Section: "The future of plastics." Joint meeting with the Birmingham PVL Club, to be held at the Imperial Hotel, Birmingham.

Newcastle Section: "The fibre optics colorimeter and its applications for industrial colour control" by Dr I. G. H. Ishak of The Paint Research Association, to be held at Royal Turks Head Hotel, Grey Street, Newcastle upon Tyne, at 6.30 p.m.

Friday 8 October

Manchester Section: "The fibre optics colorimeter and its applications for industrial colour control" by Dr I. G. H. Ishak of The Paint Research Association, to be held at the Bolton Institute of Technology, Deane Road, Bolton, at 6.30 p.m.

Hull Section: Dinner Dance, see p. 923.

Tuesday 12 October

West Riding Section: "Pigmentation of plastics" by Mr J. E. Todd of CIBA-GEIGY (UK) Ltd., to be held at the Griffin Hotel, Boar Lane, Leeds at 7.30 p.m.

Wednesday 13 October

London Section: Chairman's Forum, "Cost effectiveness and profitability in the paint industry" by Mr D. E. Eddowes, to be held at the South Bank Polytechnic from 2.15 p.m. to 5.30 p.m.

Manchester Section—Student Group: "A review of printing inks and printing processes" by Mr F. Lewis of WPM Colour & Adhesives Co. Ltd., to be held at the Manchester Literary and Philosophical Society at 4.30 p.m.

Newcastle Section—Student Group: Visit to Tioxide International Weathering Station at Carlton.

Scottish Section: "Personalised marketing" by Mr W. A. Croom of the International Life Assurance Co. (UK) Ltd., to be held at the St. Enoch Hotel, Glasgow, at 6.00 p.m.

Thursday 14 October

Midlands Section—Trent Valley Branch: "Corrosion and the automobile" by Mr H. L. Quick of Chrysler UK Ltd. Joint meeting with the East Midland Branch of the Institute of Metal Finishing, to be held at the British Rail School of Transport, London Road, Derby, at 7.00 p.m.

Friday 15 October

Irish Section: "Factors which affect the efficiency of ball milling" by Dr W. Carr of CIBA-GEIGY (UK) Ltd., to

be held at the Clarence Hotel, Wellington Quay, Dublin, at 8.00 p.m.

Midlands Section: "The importance of electrokinetics in electrodeposition" by Professor G. D. Parfitt of Tioxide International Ltd., to be held at the Birmingham Chamber of Commerce and Industry, 75 Harborne Road, Birmingham, at 6.30 p.m.

Wednesday 20 October

Scottish Section—Eastern Branch: "Developments in water-based surface coatings" by Mr A. J. Becalick of Lennig Chemicals Ltd., to be held at the Carlton Hotel, North Bridge, Edinburgh, at 7.30 p.m.

Thursday 21 October

Thames Valley Section: "Pollution" by Dr R. B. Brown, Scientific Adviser to the Greater London Council, to be held at the Beech Tree Hotel, Maxwell Road, Beaconsfield, Bucks, at 7.00 p.m.

Friday 22 October

Manchester Section: Annual Dinner Dance, to be held at the Piccadilly Hotel, Piccadilly Plaza, Manchester 1.

Friday 29 October

Bristol Section: Ladies evening, "Paint, art, colour and heraldry" by Dr S. H. Bell of The Paint Research Association, to be held at the Royal Hotel, Bristol, at 7.15 p.m.

Irish Section: Annual Dinner Dance, to be held at the Tara Towers Hotel, Dublin. Reception at 8.00 p.m.

Late News

London Section

Developments in resins for surface coatings
A special course of six lectures under the above title is being organised from 25 October to 29 November 1971 by

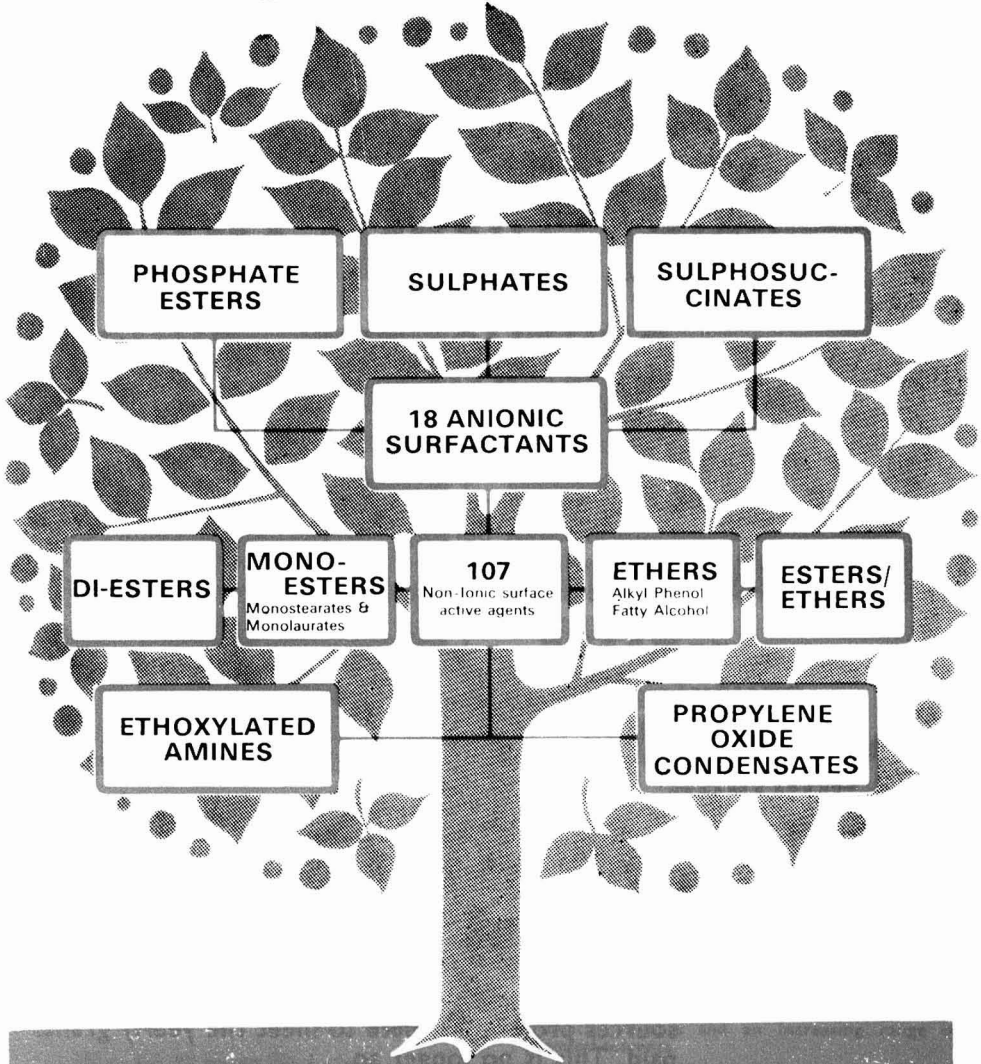
Scottish Section

Please note that the lecture by Mr W. A. Croom will now take place on Wednesday

East Ham Technical College, in conjunction with the London Section. Full details will appear in the October issue.

13 October, as shown above, and not as stated in the August issue.

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THE PAINT TRADE

BENEVOLENT FUND

The Fund was established in 1935 to provide assistance by grants or loans to necessitous cases amongst persons engaged in the colour, paint and varnish industries and to the widows and other dependants of persons lately engaged in those industries.

Pension schemes and the "Welfare State" are unfortunately not always adequate to meet difficult circumstances at a given time. It is in such cases that the Fund can be of practical assistance.

Finance is obtained by voluntary gifts from firms and individuals by way of covenanted subscriptions, annual subscriptions and donations, supplemented mainly by money raised by an annual golf competition. The Fund costs very little to administer as all services are given voluntarily.

For many years the income from these sources proved adequate to meet the yearly grants paid. This is no longer so.

Contributions are therefore greatly needed and should be sent to :

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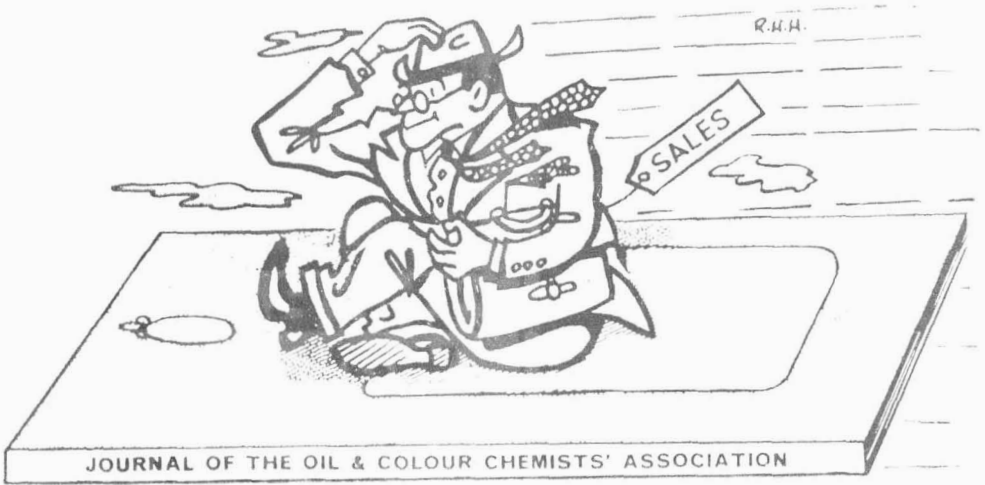
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JOURNAL OF THE OIL AND COLOUR CHEMISTS' ASSOCIATION
Wax Chandlers' Hall, Gresham Street, London, E.C.2

CLASSIFIED ADVERTISEMENTS

Classified Advertisements are charged at the rate of 50p per line. Advertisements for Situations Wanted are charged at 12½p per line. A box number is charged at 5p. They should be sent to the Director & Secretary, Oil & Colour Chemists' Association, Wax Chandlers' Hall, Gresham Street, London, E.C.2. Telephone: 01-606 1439.

SITUATION WANTED

SITUATIONS WANTED

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Development; Raw Material Testing;
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TECHNICAL EXHIBITION

OCCA 24 17-21 APRIL 1972

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paints

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The position is based in our laboratories at Darwen and is likely to be filled by a person with degree standard or maybe HNC in Chemistry and proven ability in the formulation of decorative emulsion paints. A final C & G qualification will only be considered if the applicant can show an extensive knowledge in the specialised field of emulsion paint technology.

There is a strong commercial bias to the work of this department which maintains close links with the adjacent Marketing and Production departments. Together they are helping the Company to maintain its growth record in a highly competitive industry, and applicants can be assured of some exciting and valuable experience.

ASSISTANT CHEMIST

Product Development

The Industrial Paints section of the Development Laboratories of Crown Paints, based in Darwen, have one vacancy for an Assistant Chemist. The work will involve Product Development of a non-routine nature, and demands customer contact through the Sales Department in order to provide imaginative but technically sound solutions to practical problems.

Suitable applicants are likely to be aged up to 26 years, with 4-5 years' experience in industrial paint technology, and ONC Chemistry is a minimum qualification. Ideally, candidates should be qualified to HNC or City and Guilds final level with development experience.

TECHNICAL SERVICE REPRESENTATIVE

The Industrial Paints Division of Crown Paints require a Technical Service Representative for South-East England, including Greater London. The work will involve industrial paint application trials on site and the provision of a technical advice and problem-solving service to industrial customers. Responsible to the Technical Service Manager, in Darwen, representatives work closely with sales representatives and laboratory staff to provide a customer service for their territory.

Suitable applicants, ideally in the age range of 23-35 years and qualified to ONC standard in Chemistry or Paint Technology, would have general experience in paint technology and industrial paint applications.

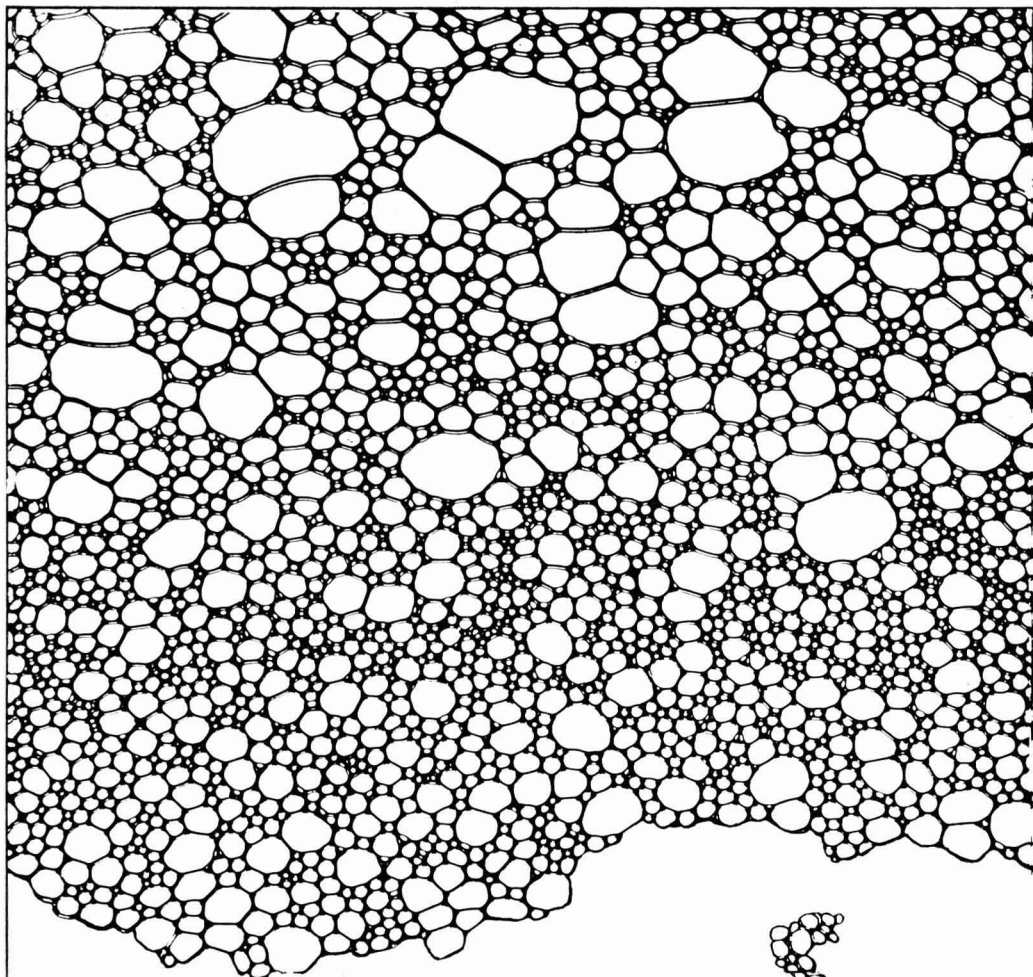
Roller coating methods are an important feature of the work and knowledge of these will be an added advantage. Self-motivation and the ability to plan and control his own activities are important.

Salary will be by negotiation. Company car and normal expenses are provided, and removal assistance for suitable candidates will be considered.

For the above vacancies, please write with full details to:

**J. V. Duckett, Personnel Officer, Crown Paints,
Crown House, Hollins Road, Darwen, Lancs.**





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