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# Colours of stratified media—I. Ancient decomposed glass

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## 1. Introduction

In a memoir published many years ago, Sir David Brewster (1863) described the structures found in ancient decomposed glass as well as the optical phenomena which they exhibit and illustrated them by reproductions of coloured drawings. The effects are most beautifully shown by specimens of antique glass-ware discovered in archaeological excavations, but as these usually find their way into museums, few physicists have the opportunity of examining them and acquainting themselves with the facts by personal observation. At the Palais de la Découverte in the Paris Exposition of 1937, one of us saw in the section of Optics a very striking exhibit of an ancient vase of glass which had been excavated in Syria by the French archaeologist M Pupil. The remains of the vase together with numerous iridescent flakes resulting from its disintegration were placed in a plate-glass cabinet which was provided with viewing mirrors inclined at 45° to the vertical, and illuminated both from above and below, so that they could be seen simultaneously by transmitted and reflected light. The brilliance of the colours and the complementarity of the same as seen in transmission and by reflection were thus beautifully made evident. Through the kind offices of Prof. A Cotton, a few flakes of glass from this exhibit were presented to us by M Pupil. This gift enabled us to undertake the present investigation, which indeed we were desirous of doing, to supplement and complete our earlier work on the optical behaviour of decomposed glass of modern origin described recently in these *Proceedings* (1939). The important researches of M Marcel Guillot (1934) on the production of iridescent laminae on glass by chemical action should also be mentioned in this connection. His work indeed suggests the possibility that the processes which take place in glass very slowly under natural conditions may be reproduced in a more rapid and controllable manner by laboratory methods. Reference may also be made to the very interesting thesis by Dr Coeling (1939) in which she has studied the absorption of water-vapour by films of decomposed glass and the resulting changes of colour.

The flakes derived from the Syrian vase reflect light strongly, exhibiting an almost metallic lustre, the colour of which varies greatly. The thicker flakes amongst those given to us exhibit a bluish-white silvery lustre, while the thinner ones exhibit other tints in which greens and oranges are the most striking colours observed at normal incidence. The flakes also exhibit vivid colours by transmitted light, being in this respect much superior to the specimens of decomposed glass of modern origin considered in our recent paper. The latter show scarcely any perceptible tints when observed in transmission, while on the other hand, the flakes from the Syrian vase show colours in transmitted light which in many cases are more striking than those seen by reflected light. The transmission colours for the thicker specimens tend towards a rich red, while the thinner flakes showed colours ranging over the whole spectrum from violet to red. It is thus evident that the development of colour occurs in antique glasses to a greater depth and in a more uniform manner than in the modern specimens. This makes it all the more desirable that they should be thoroughly studied.

Figures 1 to 9 in plates I–VII accompanying this paper are photomicrographs of selected areas on the iridescent flakes of Syrian glass. Figures 1, 2, 3, 4, 5 and 9 are pictures taken by monochromatic light in transmission with a petrographic microscope, a mercury arc lamp with a green ray filter being used as the source. Amongst these figures 2 and 9 were taken with the specimens placed between crossed nicols, while in taking the other photographs the polariser and analyser were removed. The fact that these photographs have been taken with monochromatic illumination has enabled us to notice and record certain characteristic features which would not otherwise have been evident. Figures 6, 7 and 8 are pictures taken by reflection using the Leitz ultra-opak microscope and white light. We have also obtained spectrograms (with incident white light) of the transmission and reflection by several of the flakes. These are reproduced as figures 10(a) to (f) in plate VIII. The present paper deals with the question of the structure of the decomposed glass and the origin of the colours which it exhibits as indicated by the microscopic examination and by the spectroscopic studies.

## 2. The lamellar structure

That decomposed glass is composed of thin laminae adherent to each other is readily noticeable, even without the use of a microscope. The details of its structure however become more evident on microscopic examination and are quite remarkable and interesting. Figure 1 (plate I) illustrates a common but by no means universal feature, namely that the laminae, instead of being perfectly plane, consist of shallow cups shaped like watch-glasses fitting together and dividing the surface of the flake into a large number of irregular polygons bounded by straight lines. The uniformity of curvature of the surface of the cups and the perfect sharpness of the lines in which they intersect are noticeable features. The former feature is shown in figure 1 by the perfectly concentric arrangement of the interference rings between the upper surface of the lamina and a plane sheet of mica which was laid over it when the photomicrograph was taken. It will be noticed that each of the polygons shows a ring system and that all of them are very much alike, in other words, that the curvatures of the cups are nearly the same, though their areas vary greatly. It will be noticed also that the dividing lines or sides of the polygons present a very similar appearance everywhere. These dividing lines are actually seen, however, as sets of diffraction bands running parallel to the boundaries: these have a bright centre when the microscope is focussed slightly above the plane of the lamina and a dark centre when the focus is below the plane. At critical focus the diffraction bands tend to become rather inconspicuous. From these observations it may be inferred that the dividing lines between the cup-shaped areas are extremely fine, indeed of razor-edge sharpness.

The hollow cup-like forms are naturally convex on one side of the lamina and concave on the other. This is noticeable on turning the lamina over on the stage of the microscope. The curvature of the surfaces may be made conspicuously evident by illuminating the lamina rather obliquely and observing it by reflected light. Figure 6 shows this effect when the cavities are shallow and figure 8 when they are rather deep. The smoothness and uniformity of curvature of the cavities is indicated by the fact that they are capable of forming sharply defined images by reflection. This can be demonstrated with the reflecting microscope by pushing up or pushing down the objective as the case may be. Each cup-shaped area then shows an image of the illuminating annulus of the microscope, the surfaces themselves appearing dark except at the polygonal boundaries which continue to be seen faintly by reason of diffracted light (figure 7).

It is possible to find laminae which do not exhibit the cup-like cavities referred to above, but have a smooth surface either wholly or except for discrete single or multiple cavities which they may exhibit. Examination of such a lamina under the polarising microscope between crossed nicols shows the surface as quite dark when there are no cavities, but as faintly illuminated when there are shallow cavities present. The deeper cavities appear strongly luminous round their inner margins and also exhibit the phenomenon of the black cross observed by Brewster (figure 9), which is due to the rotation of the plane of polarisation of the light in its passage through the oblique surfaces of the lamina. Particularly beautiful are those cases in which there are numerous deep cavities which are discrete and do not meet to form polygonal figures. One such lamina as seen between crossed nicols is shown in figure 2 which exhibits quite a number of ellipsoidal cavities. Further, it will be seen that the luminosity is not confined to the edge of each cavity, but also extends within in the form of rings running parallel to its margin. In most cases, this appears to be a diffraction effect, but there is no doubt that occasionally also we may have multiple cavities forming concentric rings.

## 3. Colour effects

The laminar structure of the decomposed glass is shown very prettily by the colours seen in transmission through the microscope. As was remarked by Brewster, areas of different colours will be noticed around the edges where the flakes have broken off unevenly. These correspond to the different thicknesses through which the light has to pass before reaching the eye. The boundaries of these areas act as diffracting edges and appear as sharp lines in the field of the microscope. Except when the laminae have thus split off unevenly or when they have actually parted off from each other so as to admit a film of air between, the colours seen are very uniform over the surface of the flake, thereby showing that the glass, though laminar in structure, nevertheless consists of coherent layers in optical contact. Brewster's view that the colours are due to thin plates of air separating the laminae thus appears to be incorrect. Indeed, the fact to which Brewster himself drew attention, namely that the elementary films of glass adhere with such force that it is difficult to separate them, is very strong evidence that they are in optical contact and not separated by continuous films of air.

The colour effects observed with those laminae which exhibit cup-like depressions over their area are very pretty and significant. If the cups are very shallow, the colour of the laminae is nearly uniform in tint over the area of the cup, though just at and near the boundary lines the colour is slightly different. When the cups are deep, the colour near their margins differs strikingly from that near the centre. This is due to the lamina being inclined to the line of vision round the periphery of a cup, whereas at its centre it is seen normally. To show that this is the correct explanation, we have merely to tilt the lamina on the stage of the microscope. The colours then change unsymmetrically, the part of the periphery of the cup which is now seen nearly normally has the colour which was originally observed at the centre, while the part at the periphery which is now still more inclined to the line of vision changes its colour still further in the sequence of increasing obliquity. It is equally easy to follow the change of colour with increase of obliquity in these areas where there is no curvature, by merely tilting the lamina on the stage of the microscope. It is then seen that the sequence of colour is the same as over the areas which exhibit curvature. This demonstrates that the lamina is uniform in its structure and thickness in spite of the curvature and consequent apparent difference in colour over certain of its areas.

When optical contact between the layers of a lamina is actually broken, and a thin film of air enters, this makes itself evident by continuous variations of colour over the area of the flake as seen under the microscope. If the air film is sufficiently thick and, as must generally be the case, variable in thickness, regular bands of colour appear over such area. If more than one such film of air has found entry into a given flake, each film gives its own sequence of colour running along its direction of most rapid variation of thickness. We would then see two or more sequences of colour running across the film in different directions and intersecting each other. In view of the fact that the light transmitted by the flake as a whole is itself only a part of the spectrum, the colour sequences are quite different from the usual Newtonian scale. Even apart from this, they present peculiar features which become more intelligible when the incident light is monochromatic and the transmission through such film is examined under the microscope. Figures 3, 4 and 5 reproduce three photomicrographs obtained in this way which will repay careful examination. It will be seen that each of them is traversed by a series of sharply defined bright lines running more or less parallel to each other in a relatively dark field and that in some cases, (particularly figure 3) two or more such series of lines running in different directions and intersecting each other are seen. That these effects are due to air films having entered the flake and broken optical contact between the laminae is shown by gently pressing the flakes on the stage of the microscope. The bright bands are then seen to curve and move about in the field, but they recover their position when the pressure is removed. The fact that the band systems are more frequently seen near the edges of a flake where it is obviously easier for the laminae to part company and admit a continuous film of air (see for instance the left-hand side of figure 3) is another indication that they do, in fact, arise in this way.

A remarkable feature noticed in figures 3, 4 and 5 is that in the interference bands which are so prominent, the maxima of illumination are sharp lines separated by broad dark bands. This feature suggests an analogy with the sharp interferences seen in a Fabry–Perot etalon and indeed is very similar to that seen under the microscope when a wedge-shaped film of air between the silvered faces of two plates of glass is examined by transmitted monochromatic light. The two parts of the flake separated by an air-film behave therefore much as if they were heavily silvered surfaces in passing through which the light also undergoes multiple reflection, thereby sharpening the maxima. That this is the correct explanation of the sharpness of the maxima of illumination in these interference bands is indicated by the fact that the maxima are not so sharp when they are seen in air-films separated by very thin laminae (see for instance, the bands towards the thin edge of the flake in figure 3). The laminae chosen for obtaining figures 3, 4 and 5 were as nearly as possible uniform. They however exhibit a number of circular curved depressions which will be seen in the figures. When the interference bands due to the film of air are seen through such a depression, they appear wider apart or reduced in width, according as the curvature faces one way or the other. A fine example of a deep cavity is seen at the lower right-hand corner of figure 3, while several relatively shallow ones will be seen in figures 4 and 5. Three very curious examples of sharply-defined and concentric bright ring-systems will be noticed in figure 4; these however evidently arise in a different way, presumably as the result of the local separation of the laminae by a film of air over these areas. The fact that a small dark and rather elongated nucleus can be seen at the centre of each of these ring-systems is probably a connected circumstance.

## 4. Effect of immersion in liquids

It has been remarked above that the laminae in the flake are adherent to each other and form a continuous structure. The colours seen in reflection and in transmission must therefore be ascribed to this structure being stratified in such a manner that the refractive index of the medium varies quasi-periodically. An insight into the nature of the stratifications and of the resulting variations of refractive index is obtained by studying the effect of immersing the iridescent flakes in various liquids. Putting a small flake in the cavity on a microscope slide and allowing a little liquid to flow in, it will be noticed that the colours usually observed in transmission generally disappear or become inconspicuous, and that this is the case, whatever may be the refractive index of the liquid ranging from water ($\mu = 1.33$) to methylene iodide ($\mu = 1{\cdot}74$). The colours of reflection however continue to be observable, but they are altered and enfeebled to an extent depending on the refractive index of the liquid used. To enable this effect to be studied critically, it is convenient to immerse the flake diagonally inside a small glass cell of square cross-section containing the chosen liquid which is then held facing the light. The reflection by the flake at an angle of 45° deviates the light incident on it by a right angle and may be viewed through the cell against a dark background. Even a feeble reflection may then be readily recognised. Actually the iridescence is seen quite brightly if the flake is immersed in water ($\mu = 1{\cdot}33$), ether ($\mu = 1{\cdot}35$) or acetone ($\mu = 1{\cdot}36$) though a progressive diminution in intensity with increasing refractive index may be noticed. In hexane ($\mu = 1{\cdot}38$) and in paraldehyde ($\mu = 1{\cdot}40$) the iridescence is still conspicuous, though weaker. In chloroform ($\mu = 1{\cdot}44$) the coloured reflection is very weak but may still be seen. In carbon tetrachloride ($\mu = 1{\cdot}463$) it is practically unobservable. In benzene which has a higher refractive index ($\mu = 1{\cdot}50$) it is once again visible, while in chlorobenzene ($\mu = 1{\cdot}530$) and in bromobenzene ($\mu = 1{\cdot}56$) it is conspicuous. In carbon disulphide ($\mu = 1{\cdot}63$), it is quite bright, in fact, many times more intense than the reflection from the surface of a single plate of glass immersed in the liquid.

Placing the flake in a cell containing carbon tetrachloride, we may add a little chloroform and depress the index of the liquid, or add a little benzene, and raise its index until the reflection becomes observable. It is found in this way that an elevation of the refractive index to 1·470 or a depression to 1·460 is sufficient to make the reflection visible. The disappearance of the reflection may be explained by assuming that the decomposed glass has an open or "frame-work" structure into which the liquid, allowed sufficient time, can penetrate, and that when such liquid has a refractive index 1·465 the stratifications of optical density within the substance of the flake vanish and that it therefore ceases to reflect light. At the same time, according to this view, the refractive index of the saturated substance should itself be the same as that of the surrounding fluid, and its edges should therefore cease to be observable. This is actually found to be the case, and an application of the Becke immersion test under the microscope shows that the difference of refractive index between the flake and the surrounding fluid changes sign when the latter is altered from 1·460 to 1·470.

There is a definite change in the colour of the reflected light when the flake is immersed in the cell of liquid. This change is most readily seen in the colour of the normally reflected light, and is more conspicuous, the greater the refractive index of the immersion liquid. In general, it is found possible to compensate the change of colour produced by such immersion by viewing the flake obliquely. In other words, we can set off the effect of the increased index on the optical paths within the flake by the diminution in such paths due to a greater obliquity of incidence of the light on it. That an obliquity of the order of 45° or less is sufficient to achieve this indicates that the optical paths within the flake which are altered by the entry of liquid are not a very large part of the whole. It should be remarked also that though the effect of immersion is to diminish the intensity of the coloured reflections and, as stated above, also to alter them appreciably, nevertheless the liveliness of the colours is actually improved instead of being impaired by such immersion. Further, the coloured reflections by the immersed films continue to be visible at greater obliquities instead of changing rapidly to white light as is the case when the flakes are observed in air. When the refractive index of the immersion liquid approaches that of the decomposed glass, the colours may be seen by reflection even at nearly grazing incidences.

The very low index (1·465) of the decomposed glass indicated by these observations is a noteworthy feature. It must be assumed that as the result of the decomposition and of the leaching out of some of the material in the course of long years, the average index, even of the more relatively compact parts of the glass has been reduced below that of normal glass. If the openings into which liquid can penetrate are taken into account, the effective reduction of the refractive index of the substance would of course be still greater.

## 5. Spectroscopic examination

Each of the figures 10(a), (b), (c), (d), (e) and (f) in plate VIII relates to a distinct specimen of iridescent glass and contains a group of four spectra. The first and fourth of these are the spectra of the light source employed, namely, a filament lamp, and have been included for comparison with the second and third spectra in the same group which refer respectively to the light transmitted and reflected normally by the particular flake of glass.

It will be noticed that figures 10(a) and (d) present a remarkable contrast, indeed being almost exactly the opposites of each other. In the former, the entire spectrum is cut off in transmission except for a band at the red end which comes through, while in the latter, the whole spectrum is transmitted except a band at the red end which is cut off. *Per contra* again, in figure 10(a) practically the whole spectrum appears in the reflected light except the red end which is greatly weakened, while in figure 10(d), only the red end of the spectrum appears in the reflected light and the rest is cut off. Figures 10(b) and (e) also present contrasting features of the same general description, though not of so marked a character. The principal feature in figure 10(c) is the cut-off in the transmitted light of a portion of the spectrum in the yellow region and the appearance of the same region in the reflected light. Figure 10(f) shows no special feature except a large number of bands in the spectra of both the reflected and the transmitted light. The following table shows the colours of the normally transmitted and reflected light with the different specimens:

Table of colours

| Figure | Transmission colour | Reflection colour |
|---|---|---|
| 10(a) | Orange red | Blue |
| 10(b) | Brick red | Bluish white |
| 10(c) | Purple | Yellowish white |
| 10(d) | Blue-green | Red |
| 10(e) | Blue-green | Red |
| 10(f) | Neutral | Neutral |

The vividness of the colours exhibited will be readily understood from the character of the observed spectra.

It is very remarkable that, as mentioned above, specimens of iridescent glass should exhibit two entirely different types of spectra, the first in which a limited region of the spectrum is strongly reflected while the rest of the spectrum is transmitted, and the second in which a limited region of the spectrum is transmitted while the rest is reflected. The first type of spectrum is characteristic of regularly stratified media with a large number of layers when the reflecting power of an individual layer is small. In such a case, the incident light is transmitted freely except in respect of the narrow band of wavelengths for which the successive *reflections* reinforce each other by agreement in phase. This band of wavelengths is selectively reflected and disappears from the transmitted light. If, however, the number of layers is small, the reflected spectrum contains a wider band of wavelengths with subsidiary maxima on either side. There is little doubt that the transmission and reflection spectra illustrated in figures 10(d) and (e) are to be explained in this general way, though it is clear that the regularity of the actual stratifications is far from being perfect. The spectra of the second type illustrated in figures 10(a) and (b) resemble those observed when a beam of white light passes through a thin film of air between two heavily-silvered plates. In this case, as is well known, the effect of the large reflecting power of the surfaces is to reduce the transmitted light to a negligible intensity except in respect of those wavelengths for which there is an agreement in phase of the successive *transmitted* pencils. Only the narrow bands of such wavelengths appear in the spectrum of the transmitted light and the rest are cut off. The spectra reproduced in figures 10(a), (b) and (c) have evidently to be explained in this way. The difference in optical behaviour between the two classes of cases may be the result of either the spacings or their reflecting power or both being widely different. In this connection, reference may be made to a mathematical investigation by the late Lord Rayleigh (1917) in which he has shown that the passage of light through a regularly stratified medium may result in effects which are the opposites of each other, depending on the circumstances. In one set of cases, the effect of increasing the number of the stratifications is to make the reflection approach totality. In the second set of cases, the intensity of the reflected beam continues to fluctuate however large the number of stratifications might be, and might even vanish, in which case the light is completely transmitted. Rayleigh has discussed the criterion which distinguishes the two sets of cases and shown that the results are determined by the relation between the reflecting power of an individual stratification and the phase differences arising in passage through it.

The effect of tilting an iridescent lamina on the spectra of the transmitted and of the reflected light may be readily studied. In either case, the bands observed in the spectrum shift towards the violet and broaden with increasing obliquity. The actual effect of this on the observed colours is rather different in the two types of cases considered above. If, for instance, the transmission shows a dark band in the red region at normal incidence, the displacement of this towards the violet with increasing obliquity results in the colour seen changing from blue-green to blue, then to purple and yellow, while the reflected colours march from red to violet through all the rich tints of the spectrum. If, on the other hand, the transmission is initially limited to a band at the red end, the effect of increasing obliquity is to cause more and more of the spectrum from the red towards the violet to be freely transmitted. As the result of this, the transmission colour fades out rapidly, changing from red to a pale orange or yellow. The reflected colour which is initially a bluish white however becomes a richer and darker blue and finally tends towards indigo or violet.

As is to be expected, both the reflection and the transmission are strongly polarised and in opposite ways at the Brewsterian angle of incidence. Here again, there are remarkable differences between the character of the effects observed in the two sets of cases. When the transmitted light includes the whole spectrum except a limited band of wavelengths, striking changes are observed when the obliquely transmitted light is viewed through a polarising nicol. With the light vector in the plane of incidence, the film appears practically colourless, but with the light vector perpendicular to its plane, it appears richly coloured. On the other hand, the films showing only a narrow transmission band at normal incidence are practically colourless at the polarising angle, the effect of rotating the observing nicol being merely to alter the intensity of the transmitted light.

These same films however, show a remarkable increase in the richness of the colour of the *reflected* light at the polarising angle. The explanation of these facts will be clear in the light of the preceding discussion, when we remember that the reflecting power of an individual stratification is diminished for one of the components of the light vector and increased for the other by oblique incidence.

On immersing the iridescent flake in a cylindrical vessel of liquid of refractive index greater or less than that of glass, it is readily possible to observe the spectral character of the light reflected by it at various obliquities up to grazing incidence and to determine its state of polarisation. As remarked earlier in the paper, the transmission colours disappear on immersion in the liquid, but the reflected colours continue to be visible with great liveliness up to grazing incidence. They are also completely polarised at the Brewsterian angle of incidence on the liquid-glass boundary. These facts will be readily understood in the light of the preceding discussion, when it is recollected that the reflecting power of an individual stratification is greatly reduced when the lamina is immersed in a liquid. In the spectrum of the normally reflected light, narrow bands are observed which broaden out at oblique incidences, thus indicating that the iridescence is the cooperative effect of several laminations.

## 6. Effect of absorption of liquid

Very interesting phenomena are exhibited by an iridescent flake of glass when it is immersed in a liquid and then taken out and allowed to dry. As has already been mentioned, the flake appears nearly or quite colourless by transmitted light when wet, and it might be thought that the colour would reappear progressively as the drying proceeds. Actually, the colour reappears with remarkable suddenness and with extreme saturation. The phenomenon may readily be observed by holding up the film against the light from a window and watching it as it dries. Almost immediately the drying commences, the film turns black as viewed by transmitted light and silvery white as seen by reflected light. The film then gradually clears up, showing rich colours which fade away until finally the usual colours are restored. The phenomenon may be watched by transmitted light under the microscope, and this is specially advantageous when it is desired to scrutinise it in detail. A boundary of intense colour or opacity appears near the edge of the colourless film and moves inwards as the drying progresses, finally covering the whole film. It then gradually weakens and disappears.

It is not necessary for observing these effects that the refractive index of the liquid should be different from that of glass. Indeed, carbon tetrachloride which, as we have seen, causes the reflection colours to disappear, shows the effects described above just as effectively as any other liquid. If it is desired to prolong the duration of the stage of opacity, the flake may be kept under a microscope cover-slip, thus delaying the evaporation. Alternatively, a less volatile liquid may be used to wet the film. On the other hand, its drying may be speeded up by holding it in a current of hot air. This is a useful procedure when the liquid has been wetted by water or by a liquid of high boiling point.

These effects throw light on the structure of the iridescent films and show clearly that Brewster's explanation of the colours as due to thin films of air separating the layers of glass is untenable. The observations indicate that the glass has a continuous frame-work structure which is quasi-periodic or stratified. When the flake is completely wetted, the liquid penetrates into the minutest cavities and tends to render the glass optically homogeneous, though if its refractive index differs from that of glass, the stratifications persist and give observable colours by reflection. As the film dries, the liquid naturally withdraws first from the largest cavities but remains in the smaller pores and the minutest cavities. At the earliest stages of drying, therefore, the layers containing the minutest pores which continue to hold liquid have a higher refractive index and are optically more uniform than when the film is completely dry; on the other hand, the layers containing the larger cavities are empty of liquid and have the usual low refractive index. At this stage, therefore, the stratifications of refractive index are actually more pronounced than with the dry film and their reflecting power is accordingly greater. The reflection of light by the film therefore approaches totality.

The liquid held by a wetted film may be divided into at least three categories, (a) that held by the largest cavities, (b) that held by the finest pores, and (c) that molecularly adsorbed on the capillary surfaces. The rate at which the liquid evaporates and its temperature equilibrium with the vapour would naturally be different for the three categories. The colour of the film would depend on the nature and quantity of the liquid held and the manner in which it is distributed in the film. The detailed study of these effects for the iridescent films of ancient decomposed glass would obviously be of considerable interest and should enable the capillary structure of the glass to be investigated in detail. In this connection, it is important to remember that the entry or withdrawal of liquid is far from being instantaneous. The flow of liquid through very minute capillaries is a slow phenomenon especially if it be viscous, and the capillary forces which cause the flow may sometimes vanish, e.g., when the pore has a bottle-neck shape such as has been postulated to explain hysteresis phenomena in the adsorption of liquids. These considerations help us to understand the remarkable tenacity with which liquid is sometimes held by a flake of decomposed glass as is demonstrated by its influence on the observed colour.

## 7. Effect of mechanical pressure

That the decomposed glass has an open structure and that the colours arise from this structure being stratified or quasi-periodic is very clearly indicated by the observations already set out. It is a reasonable presumption that the structure is the result of the decomposition of the glass and the leaching out of the more soluble layers formed during its progress, the periodicity of the decomposition being probably analogous to the Leisegang effect as has been suggested by M Guillot. On the assumption that the colours of the ancient glass are due to a periodic distribution of cavities or pores in its structure, we must expect that mechanical pressure should destroy or reduce the colours of the glass. The equipment may readily be tried by placing a small flake covered with a piece of cellophane on the stage of the microscope, and then pressing or rolling a blunt steel point firmly on its surface. The colour is then observed practically to disappear from the area of pressure and does not recover on its removal. The success of the experiment indicates that the colours in the Syrian vase do not arise from an alternation of solid layers of different refractive index in the material of the glass. The latter supposition would also be inconsistent with the observed easy penetration of liquid into the substance of the films.

## 8. Summary

The paper describes a detailed microscopic study of the structure of the films of decomposed glass derived from an ancient Syrian vase excavated by M Pupil, and a spectroscopic examination of their iridescence. The investigation makes it clear that Brewster's explanation of the iridescence as due to films of air separating thin layers of glass is definitely erroneous. The material is optically and mechanically continuous, but has an open framework structure which is quasi-periodic or stratified. The stratifications are moderately regular with the result that in certain specimens, a limited region of the spectrum is totally reflected and the rest is freely transmitted, while in other specimens we have the opposite effect, namely that the whole of the incident light is reflected except for a limited region of the spectrum which is transmitted. At oblique incidences and especially near the polarising angle, the colours seen vary with the azimuth of the vibration and in different ways in these two cases. Mechanical pressure destroys the structure and with it also the iridescence. Liquids can penetrate into the structure of the film, but the latter becomes optically homogeneous and ceases to reflect light only when the refractive index of the liquid is equal to that of the glass (1·465). When the wetted film commences to dry, the liquid withdraws first from the larger cavities while it is retained in the smaller pores. This results in the optical stratifications being actually more pronounced than in the dry film, with the consequence that the flake appears black by transmission and silvery white by reflection.

The paper is illustrated by reproductions of nine microphotographs and twelve spectrograms.

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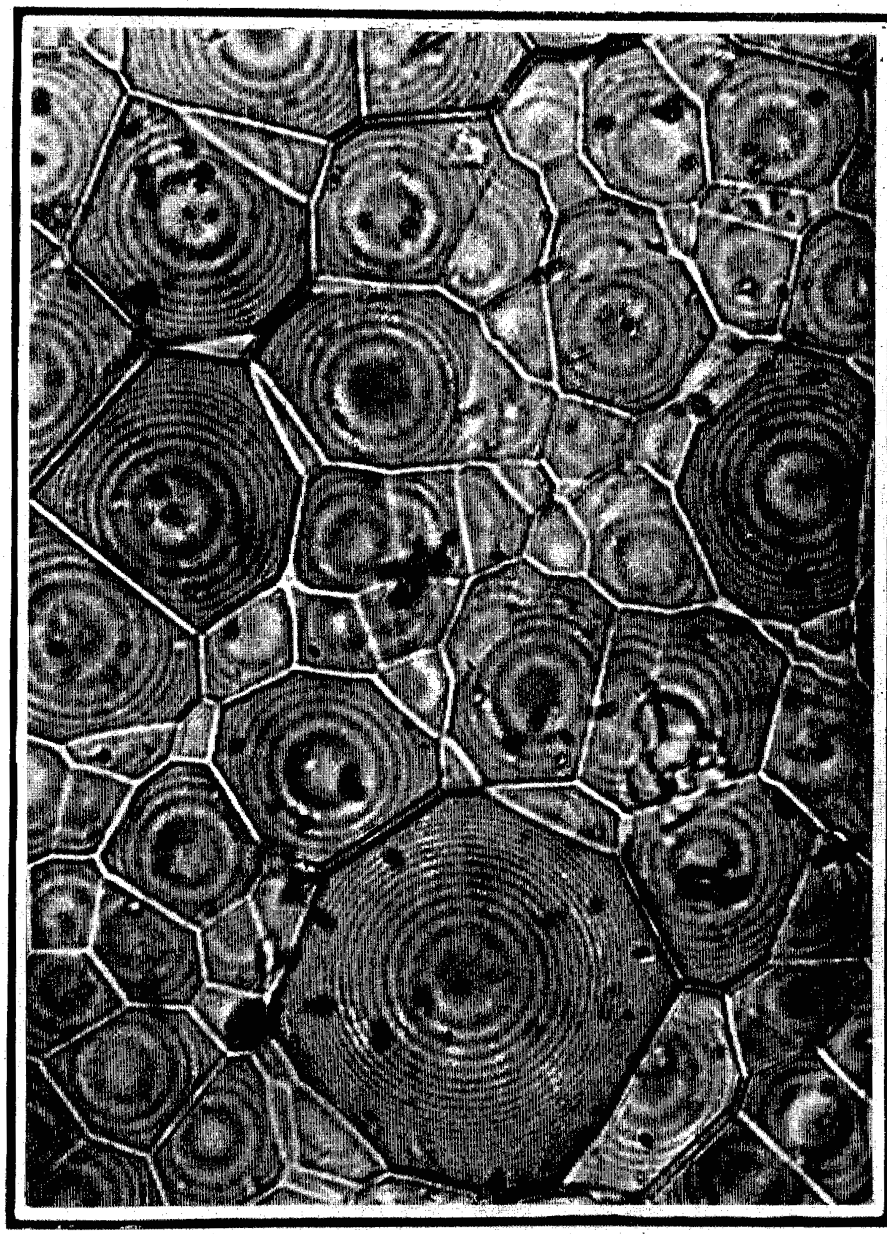

**Figure 1.** Photomicrograph of decomposed glass in monochromatic light, showing network of curved laminae.

**Plate I**

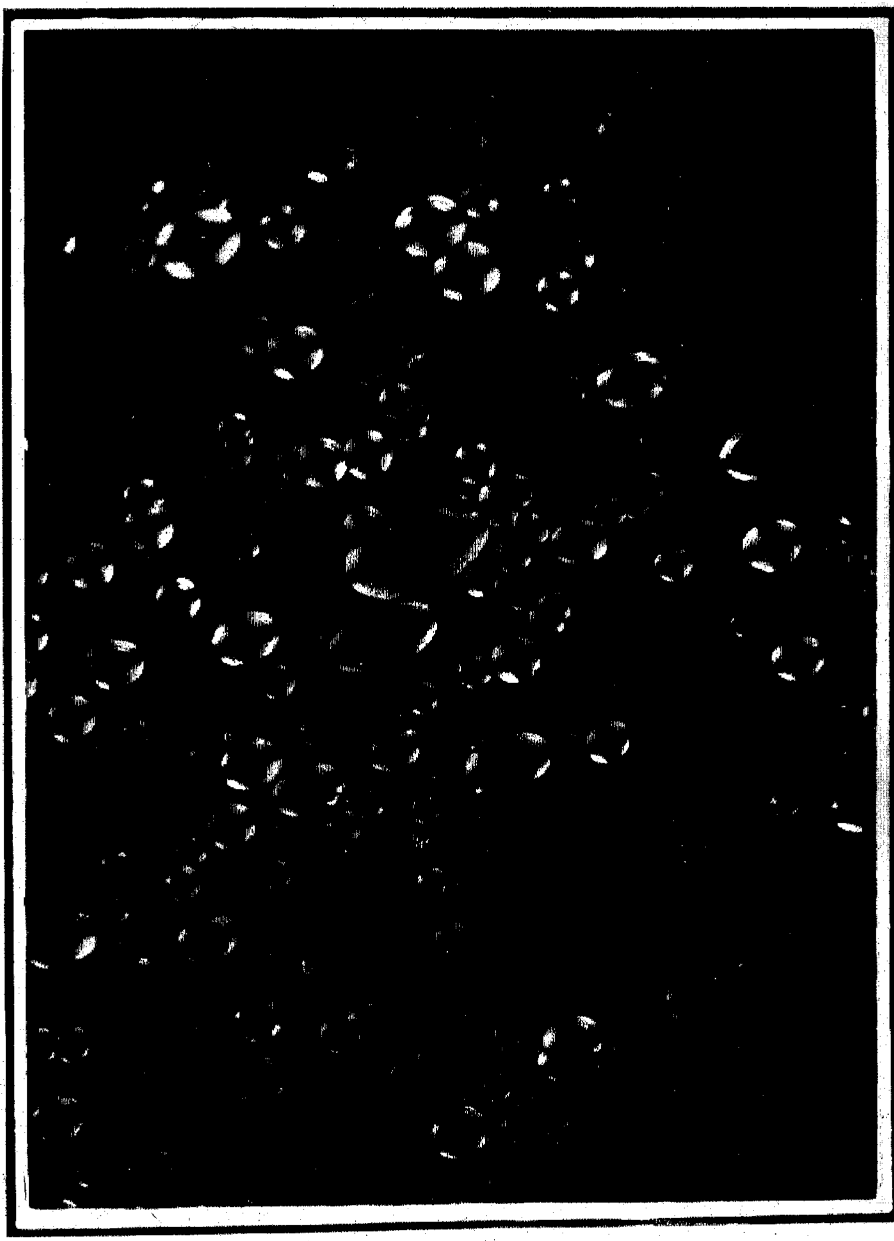

**Figure 2.** Photomicrograph of decomposed glass in monochromatic light under crossed nicols, showing spherical and ellipsoidal cavities.

**Plate II**

**Figure 3.** Photomicrograph of decomposed glass in monochromatic light, showing laminar edges, and multiple films of air.

**Plate III**

**Figure 4.** Photomicrograph of decomposed glass in transmitted monochromatic light, showing sharply defined fringes due to intruding air films.

**Plate IV**

**Figure 5.** Photomicrograph of decomposed glass in transmitted light, showing cavities and fringes due to intruding air films.

**Plate V**

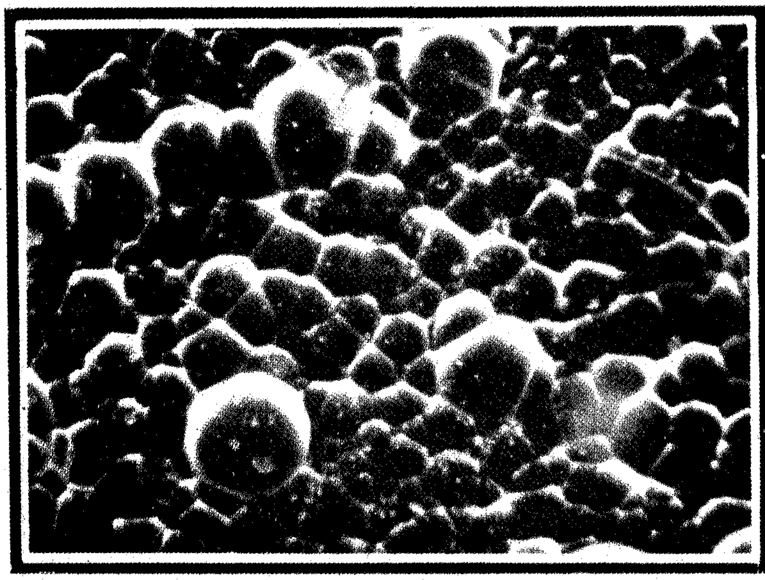

**Figure 6.** Lamina with hollows by reflected light.

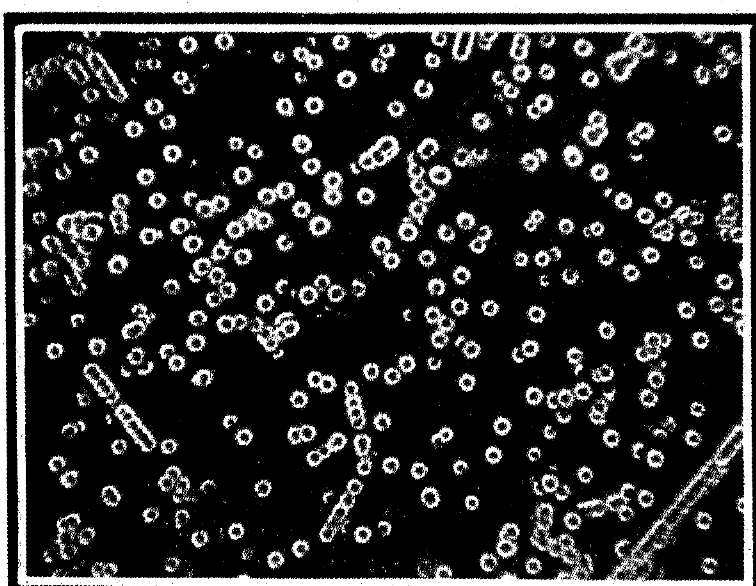

**Figure 7.** Lamina with hollows forming optical images of light source by reflected light.

**Plate VI**

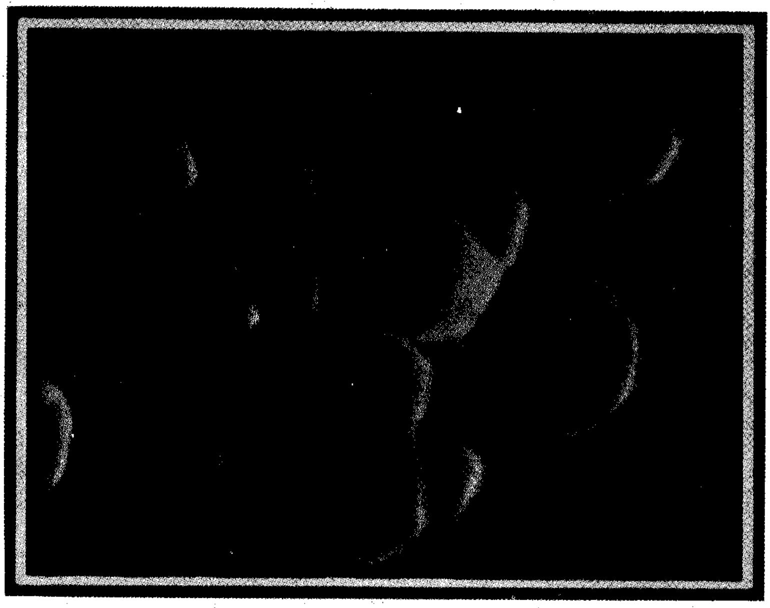

**Figure 8.** Lamina with hollows by reflected light.

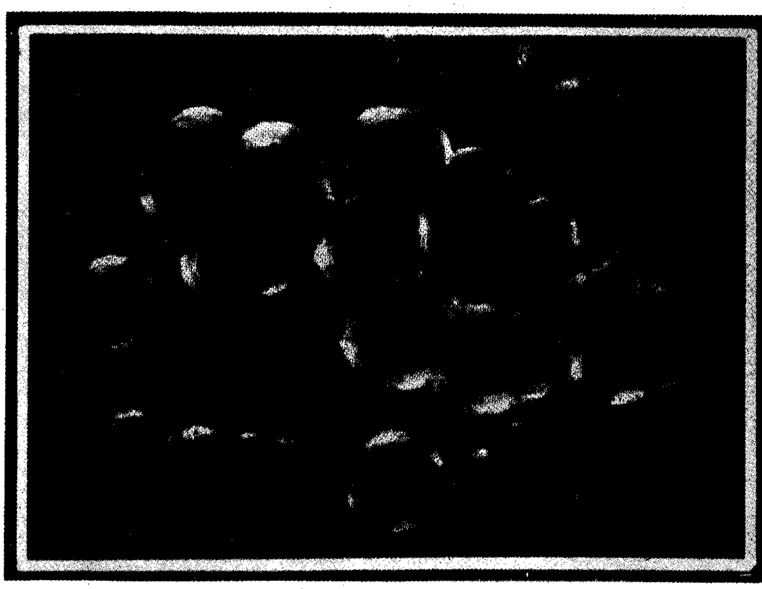

**Figure 9.** Lamina with hollows by transmitted light between crossed nicols.

**Plate VII**

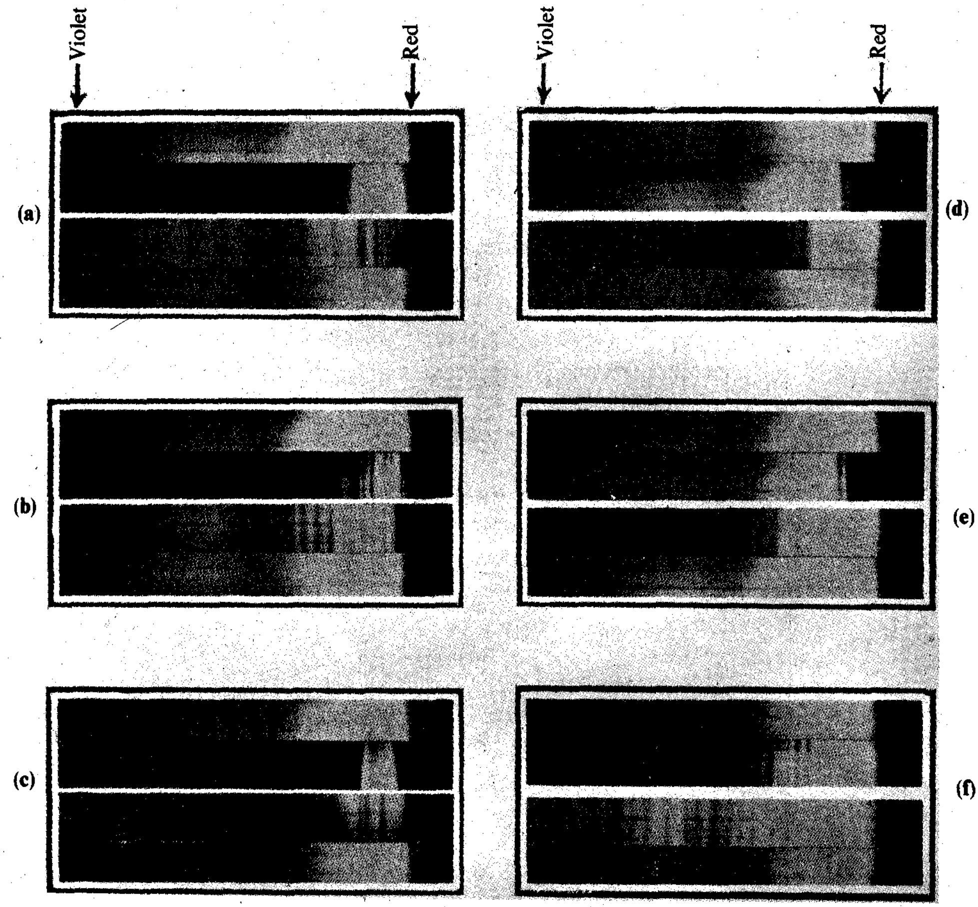


**Figure 10. (a), (b), (c), (d), (e) and (f).** Spectrograms of reflected and transmitted light of decomposed glass films.

**Plate VIII**