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The structure and optical behaviour of pearls

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1. Introduction

The pearl holds a unique position amongst gems by reason of its natural beauty which needs no enhancement at the hands of the lapidary. It is also distinguished from the rest of them by the fact of its origin within the living body of a mollusc. For these reasons, it is a product of great and general interest. The aim of the present paper is to lay bare and express in the language of science the effects which pearls display and for which they are so highly admired.

The investigations here reported may be regarded as a sequel to a recent paper[1] by us in these *Proceedings* which concerned itself with iridescent shells. It was shown in that paper that the birefringence of nacre notably influences the spectral character of its iridescence. Further, it was shown that the discrete structure of the material results in characteristic diffusion phenomena which, no less than the iridescent reflections, make themselves evident in the observed optical behaviour of the shells. In view of the general similarity in nature of the material composing pearls and of the nacre in iridescent shells, it is to be expected that analogous effects would be observed in both cases. The spherical or nearly spherical shape of a pearl has however to be considered in this connection. Further, neither the structure of the material nor the nature of the stratifications is necessarily identical in the two cases. In the present paper, observations are presented bearing on these questions. The most noteworthy outcome of the investigation is to show that the diffusive properties of the material composing pearls play a major role, which is indeed no less important than the reflection of light by the stratifications, in making pearls the beautiful objects that they are.

2. The birefringence of pearls

It is well known that in the production of cultured pearls, a sphere of extraneous material (usually mother-of-pearl) is inserted within the mollusc, as a result of which, it deposits a whole series of layers in the course of years around the nucleus thus provided. By cutting a cultured pearl in two halves, it is easy to remove the nucleus and obtain two hemispherical shells of the deposited material. These shells are in a very useful form for a study of the optical characters of the substance composing pearls.

Placing such a hemispherical shell between crossed polaroids and viewing it normally along its axis through a magnifying lens against a source of light, one observes an interference figure (see figure 15 in plate IV) which is very similar to the conoscopic figure of a plate of a biaxial crystal seen under the polarising microscope. It is remarkable that such a figure is observed in *parallel light* in the present case. The explanation of it is the hemispherical shape of the shell coupled with the fact that its optic orientation everywhere follows the curvature of the surface. Hence, when traversed by a parallel beam of light, the interference figure is localised at or near the curved surface of the shell. If the hemisphere is rotated round its axis, the isochromatic lines in the figure are observed to rotate with it, while the isogyres pass through a cycle of changes. If, on the other hand, the hemispherical shell is rotated around a diameter, the entire pattern drifts along the surface of the shell but remains substantially unaltered.

Another simple and effective way of exhibiting the birefringence is to grind one of the hemispherical shells cut from a cultural pearl to a flat surface on both sides and to mount the plate thus obtained in canada balsam between glass coverslips. Since the stratifications in the material are curved, a specimen thus prepared and mounted reflects light like a spherical shell. However, when held up against a lighted window, it becomes evident that the plate has a circular bevelled edge where the original spherical surface meets the parallel plane faces. This bevelled edge acts like a circular prism, and since the material is doubly refracting, two concentric rings of light are seen around a source of light when the latter is viewed through the plate (see figure 1, plate I). On tilting the specimen, the two circles become eccentric with respect to each other (see figure 3, in plate I). On further tilt, both the rings of light become elliptic, the outer one much more so; the two ellipses touch each other at one end of the major axis and are widely separated at the other end (see figure 5, plate I). At such oblique settings the rings also exhibit distinct colours due to dispersion. That these effects arise from the birefringence of the material is confirmed by viewing the rings of light through a polaroid. It is then noticed that the rings are extinguished in certain regions which shift around as the polaroid is rotated (see figures 2, 4 and 6 in plate I). In the case in which the source of light is viewed normally through the plate, the rings are extinguished at the ends of two diameters which are mutually perpendicular to each other.

3. The structure of the pearly layers

The shells of the bivalves[2] exhibit conoscopic figures closely resembling that reproduced as figure 15 in plate IV. It follows that the orientation of the aragonite crystallites is very similar in the two cases. In other words, the crystallites in pearls have their *c*-axes more or less exactly normal to the laminations, while their *a* and *b* axes also have fairly defined orientations in the plane of the laminations. This is confirmed by X-ray diffraction studies, all that is necessary being to send a fine pencil of X-rays along the axis of the hemispherical shell and to record the results on a flat film. Two such photographs recorded at this Institute by Mr A Jayaraman are reproduced as figures 7 and 8 in plate II. The former was obtained with the filtered X-radiation from a copper target and the latter with unfiltered molybdenum radiation. In both records, we notice that the bright arcs have a finite spread, thereby indicating that the orientations of the *a* and *b* axes in the plane of the stratifications have a tolerably large range of variation. It may likewise be assumed that the direction of the *c*-axis is also sensibly variable, as has been established by X-ray methods[3] in the case of the bivalve shells. Comparison of figure 8 with the figures reproduced in the papers of Rama Swamy quoted show that the fluctuations in the orientation of the *a* and *b* axes are of the same order of magnitude in the two cases.

When a bright source of light is viewed normally through a thin plate of an iridescent shell having its faces parallel to the stratifications, a characteristic diffusion halo is observed; for the bivalves, this consists of a diffuse central spot overlying the source and two other diffuse spots lying one on either side of it[1]. The observation indicates that the aragonite crystallites are arranged in parallel rows along the lines of growth of the shell; the spacings of the rows calculated from the separation of the diffuse spots agrees with that directly observed with thin sections examined under the microscope[2]. If the material in cultured pearls had a similar structure, we should expect to be able to observe it when thin layers are examined under the microscope and also to see similar diffusion haloes. These anticipations are to some extent supported by the results of observation. We do indeed find that thin plates of the material under a microscope exhibit a fibrous structure in the plane of the laminations. We are also able to observe in some cases, diffusion haloes similar to those of the bivalve shells (see figure 13 in plate IV). On the other hand, in other cases, a different form of diffusion halo is observed, consisting of a pattern of six diffuse spots arranged in the form of a hexagon and surrounding the central diffuse spot; two of these spots at the end of a diameter are brighter than the rest (see figure 14 in plate IV). In the deposition of pearly material round a spherical nucleus, it is difficult to see why any particular direction on the surface should be favoured. It is therefore a little surprising that parallel rows of crystals, as in bivalve shells, are found in many cases. *Per contra*, it is not surprising that in other cases a halo indicative of a grouping of the crystallites in hexagonal order in the plane of the laminations is observed.

4. The optical behaviour of pearls

Some general remarks—The appearance which any object presents to an observer is determined by the manner in which it is illuminated and by the particular way in which the light falling on the object is returned to the eye. Consider, for example, a case in which the object under examination is a polished metallic sphere held at some distance from an open window and viewed by the observer against a black background. The more perfectly polished the sphere is, the less would be the intensity of the light scattered by its surface. All that the observer would see in these circumstances is a well-defined image of the lighted window formed by reflection at the convex surface, while the sphere itself would hardly be visible. It is clear from this example that the possession of a high reflecting power by a spherical body would not by itself make it appear an attractive object.

The reflection of light by a pearl, however, occurs not solely or even principally at its surface. A whole series of spherical laminations participate, and since the material is optically heterogeneous, the reflections at the successive layers would be accompanied by a strong diffusion which would appear spread out over a range of solid angles, instead of forming a well-defined image of the luminous object. Hence, unless the source of light be of very small angular dimensions, its reflected image would be overlaid by diffuse light, and hence prevented from its being readily observed; *per contra*, the illusion would be created that the pearl is itself a lustrous and brilliant object. It is thus evident that the diffusion of light in directions approximating to that of regular reflection contributes notably towards making the pearl an attractive gem. The diffusion in other directions also plays an important role, as will appear later.

The reflection-diffraction spectra—With our earlier paper, we reproduced several photographs of the reflection-diffraction spectra of iridescent shells. These were recorded by allowing a narrow pencil of light to fall on the surface of a shell and receiving the reflected and diffused light on a photographic film held a few centimetres away. The photographs show very clearly that the diffraction of light by the corrugations at the external boundary and the reflection of light by the internal stratifications form a single coherent series of spectra, the iridescence usually appearing as the last of the series on one side. A similar phenomenon can also be observed with pearls when a magnifying lens is focussed on the image of a distant source of light formed by reflection at the surface of the pearl. A series of spectra are then seen in focus whose separation depends on the angle at which the stratifications meet the surface; if they were strictly parallel to the surface, the spectra would all overlap and we would observe only a single coloured image of the source. More generally, however, the spectra are seen clearly separated.

The diffusion-halo of reflection—Surrounding the reflected image of the source (or the whole group of reflection-diffraction spectra) is observed a phenomenon which may be designated as the diffusion-halo of reflection. The structure of this halo varies to some extent from specimen to specimen. It is most clearly seen when the external surface is truly spherical and the stratifications are parallel to the surface, so that we observe only a single image of the source formed by reflection and exhibiting the characteristic iridescence. Through this image runs a straight band of light terminated by two diffuse spots and exhibiting over its length approximately the same colour as that of the iridescent reflection. This band is encircled by two coloured arcs lying on either side of it and exhibiting a tint complementary to that of the iridescence. The obvious analogy between this and the diffusion haloes by transmitted light reproduced as figures 13 and 14 in plate IV makes it clear that its origin is to be sought for in the discrete structure of the stratifications reflecting the light. The reason why the reflection halo exhibits complementary colours in the different parts of it is not difficult to fathom. The most prominent features of the halo are those due to light selectively reflected and diffracted by the upper layers of the stratifications and hence exhibiting the characteristic colour of the iridescence. The rest of the light thus filtered out would penetrate deeper into the stratifications and would be returned by diffusion, thereby appearing spread out over a wider range of angles, and this would exhibit the complementary colour.

Some illustrative examples—Figures 9 and 10 in plate III illustrate some of the observations set out in this and the preceding section. They are photographs respectively of a natural and of a cultured pearl illuminated in each case by two filament lamps set one on either side of the pearl. The cultured pearl appearing in figure 10 was a rather perfect specimen of spherical shape. On the upper right of that picture can be seen a small bright spot being the coloured image of the source formed by reflection and this appears surrounded by an extended area of diffuse radiation. Lower down in the picture can be seen the image of the second lamp surrounded by a rather complicated pattern of diffused light.

The natural pearl which appears in figure 9, though larger than the cultured pearls in our possession and far more expensive, had a rather irregular shape, and the optical effects exhibited by it were rather irregular. In the patch of light seen in the upper right hand side of the picture, the long streak and the diffuse spot separated from it record the reflection-diffraction spectrum exhibited by that part of the surface. These features appear surrounded by a strong patch of diffuse light. In the lower part of the same picture, effects of the same general nature due to the second source of light appear.

The whispering gallery effect—The photographs reproduced as figures 9 and 10 in plate III were recorded with the pearls placed on a sheet of black glass and in a dark room with blackened walls. Nevertheless the whole of the pearl shows up quite clearly. Not only are the spherical margins clearly seen, but they are actually somewhat brighter than the central regions of the pearl. Closely connected with the explanation of these facts is the very remarkable phenomenon illustrated in figures 11 and 12 of plate III, recorded respectively with a cultured and a natural pearl. In figure 11 the pearl was illuminated tangentially on the side remote from that in which it was observed. Nevertheless it will be seen that the rear face is seen brilliantly illumined, the edges in particular being very bright. In figure 12, the pearl was illuminated at a part of the surface a little to the rear of that visible in the photograph. Nevertheless it can be seen that the entire periphery of the sphere appears lit up, the part on the right much more so.

The explanation of these effects is evidently related to the phenomenon of the lateral or transverse diffusion of light in nacre described and illustrated in a paper[4] published in these *Proceedings* twenty years ago. It was there shown that light penetrates nacre by diffusion through far greater distances parallel to the laminations than perpendicular to them. It is clear that a similar effect is operative also in the case of pearls. Light diffuses in directions parallel to the spherical laminations through considerable distances with relatively little loss, thereby enabling the pearl to be lit up and become visible in areas where no light falls directly.

The spectral character of the iridescence—Owing to the spherical shape of the pearls and the relatively small sizes available, the recording of the spectrum of iridescence presents some difficulties. The angle of incidence of the light would vary rapidly from point to point on the surface of the pearl and so also the spectral character of the iridescence. Hence it would be necessary to severely restrict the aperture of the incident beam and of the reflected beam as well. The iridescence exhibited by pearls varies considerably in its hue and saturation from specimen to specimen. The pearls taken up for examination were naturally those that exhibited the most vivid colours. Even in their cases, however, the results were rather disappointing. Complex spectra exhibiting a great many maxima of illumination, some stronger than the rest, were recorded. Further, the spectrum also showed large variations in different areas of the same specimen. See figures 16(*a*), (*b*), (*c*) and (*d*) in plate V recorded with a cultured pearl and figures 17(*a*), (*b*) and (*c*) in plate VI recorded with a natural pearl. These were obtained with the light beam incident normally on the specimen.

It would seem from these facts that the stratifications in pearl are somewhat different in nature from those in nacre. It appears not improbable that they consist of distinct groups of layers, each having a separate spacing and not a single continuous sequence. However, this is only a surmise which has to be tested out by further study of different specimens and a careful comparison with the nacre from the same species of mollusc.

Effect of birefringence on the iridescence—In spite of the complicated nature of the spectra, it has been found possible to establish their bifurcation at oblique incidences due to birefringence, as has already been done by us with nacre.

Figures 16(*e*), (*f*) and (*g*) are the spectra of a cultured pearl recorded respectively with vibrations perpendicular to the plane of incidence, with vibrations in the plane of incidence and with the vibrations unpolarised. The shift of the bands as between figures 16(*e*) and 16(*f*) is readily seen, while figure 16(*g*) is clearly a superposition of both. Figures 17(*d*), (*e*) and (*f*) are similar records obtained with a natural pearl and analogous effects are there noticed.

Figures 16(*h*), (*k*) and (*l*) in plate V obtained with a cultured pearl exhibit an interesting effect. They were recorded in circumstances similar to those of figures 16(*e*), (*f*) and (*g*) but with the beam incident on the surface of the pearl at an angle of about 60°. As in all other cases, the sphere was kept immersed in liquid paraffin to minimise disturbing effects at the external surface. It will be seen that the iridescence is almost completely polarised with its vibration direction perpendicular to the plane of incidence. Surprisingly enough, this effect was observed to persist over a wide range of angles of incidence.

Transmission spectra—Figure 18 in plate VI shows the spectra of the light transmitted by the hemispherical shell obtained from a cultured pearl, the incidence being normal in (*a*), (*b*) and (*c*) and a few degrees away from normal in (*d*), (*e*) and (*f*). The three spectra in each group were recorded respectively with light having its vibration direction perpendicular to the plane of incidence, the same lying in the plane of incidence and with the vibrations unpolarised. It will be noticed from the pictures that there is a noteworthy extinction of the component in the plane of incidence at oblique incidences, a phenomenon clearly analogous to that already noticed by us in the case of nacre.

5. Summary

The paper embodies a study of the structure of the material composing pearls and of the optical effects which they display. The following topics are dealt with: (1) birefringence, (2) X-ray-diffraction patterns, (3) the reflection-diffraction spectra, (4) the diffusion haloes of reflection and transmission, (5) the whispering-gallery effect, (6) the spectral character of iridescence and the influence of birefringence thereon, (7) the transmission spectra. The most noteworthy result of the investigation is to show that the diffusive properties of nacre play a major role no less important than that of the reflection by its stratifications in the optics of pearls.

Bibliography

A detailed account of the origin and structure of pearls will be found in F Haas' book *Bau Und Bildung Der Perlen*, (Leipzig, 1931). See also W J Schmidt's *Die Bausteine Des Tierkorpers* (Bonn 1924), pages 166–169.

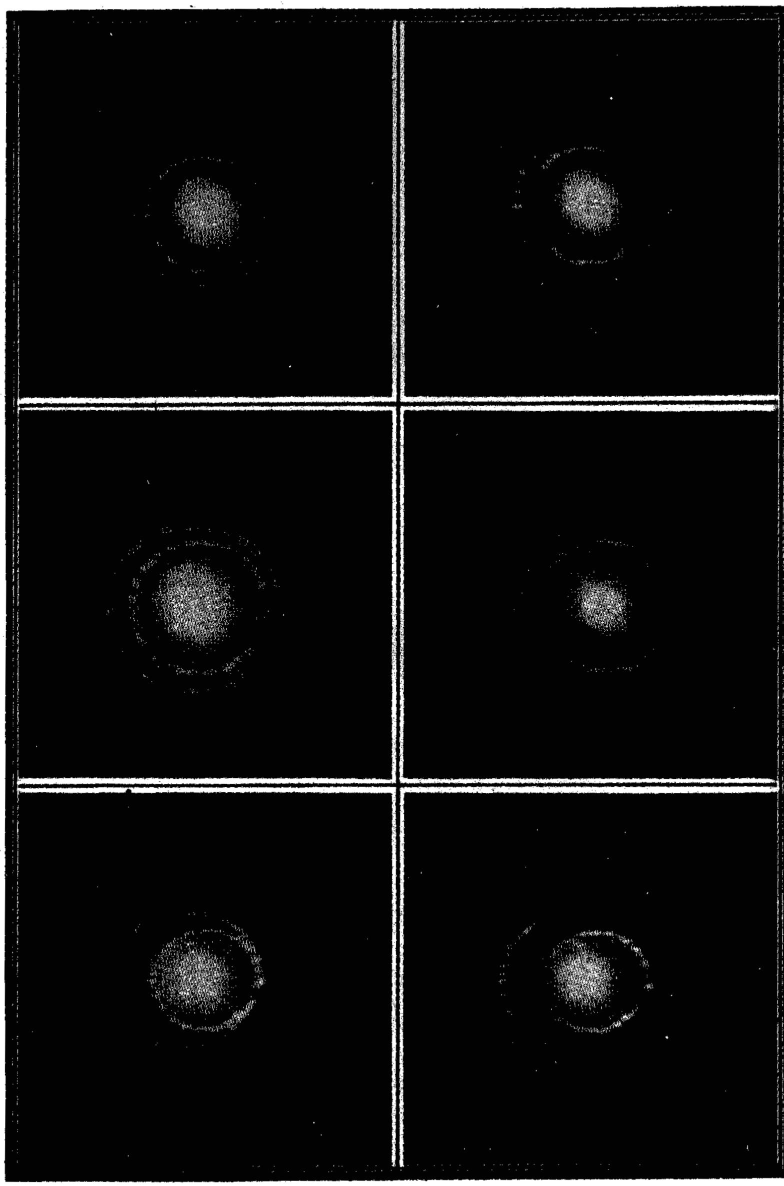
The culturing of pearls is an important Japanese industry and several laboratories in that country are at present investigating its problems and have published papers dealing with various aspects of the subject which are collected together in Bulletin No. 1 of the Nippon Institute for Scientific Research on Pearls (1953). The following papers are those referred to in the text.

1. Raman, C V and Krishnamurti, D, *Proc. Indian Acad. Sci.*, **A39** 1 (1954).
2. Rajagopalan, V S, *ibid.*, **A3** 572 (1936).
3. Rama Swamy, S, *ibid.*, **A1** 871 (1935); **2A** 345 (1935).
4. Raman, C V, *ibid.*, **A1** 859 (1935).

(1) (2)

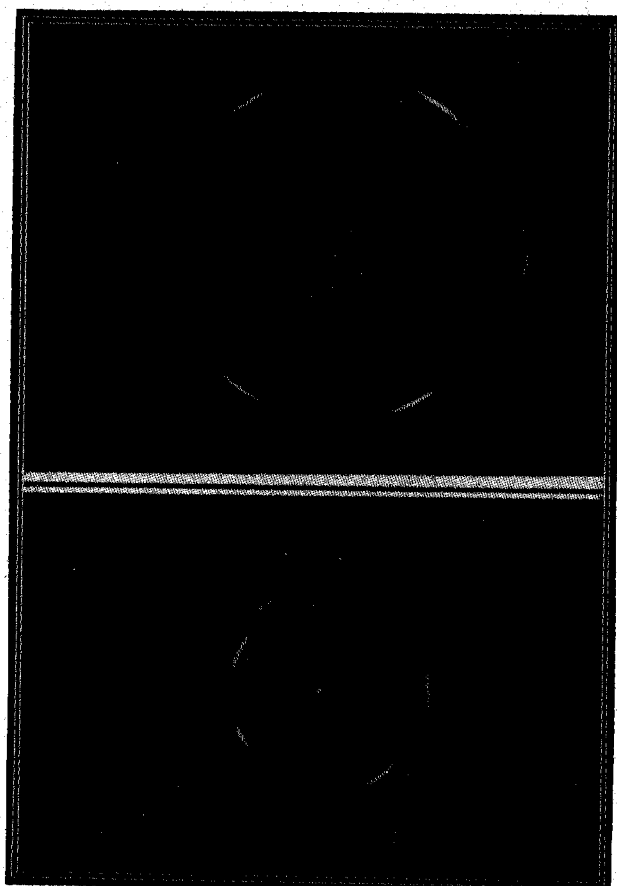
(3) (4)

(5) (6)



Figures 1–6

Plate I



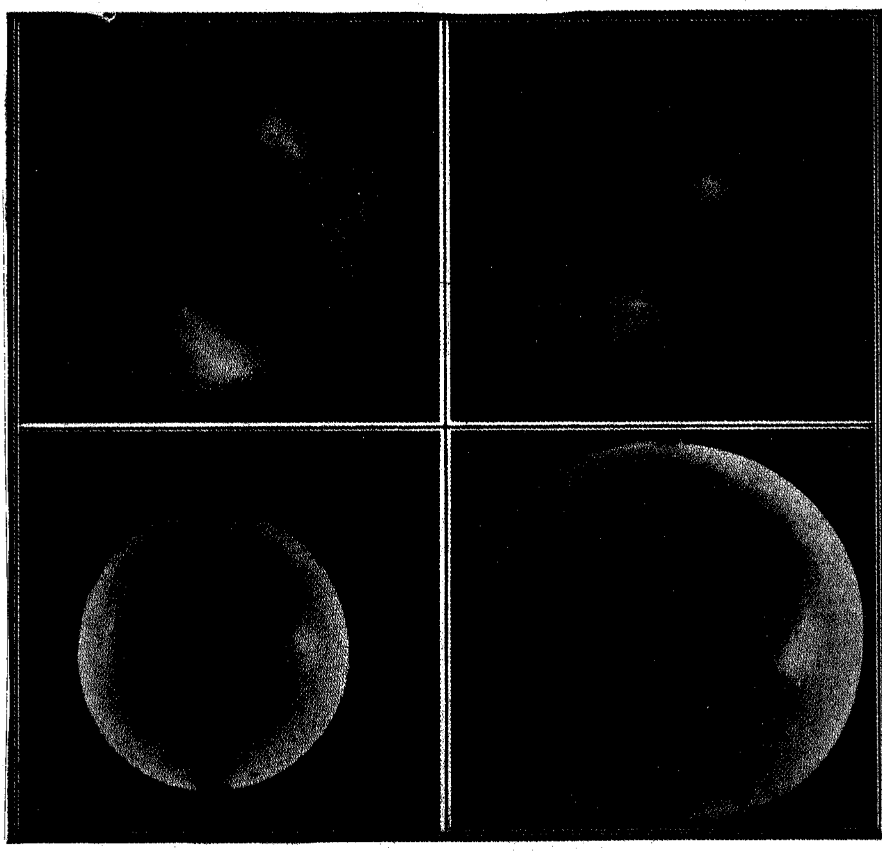
(7)

(8)

Figures 7 and 8

Plate II

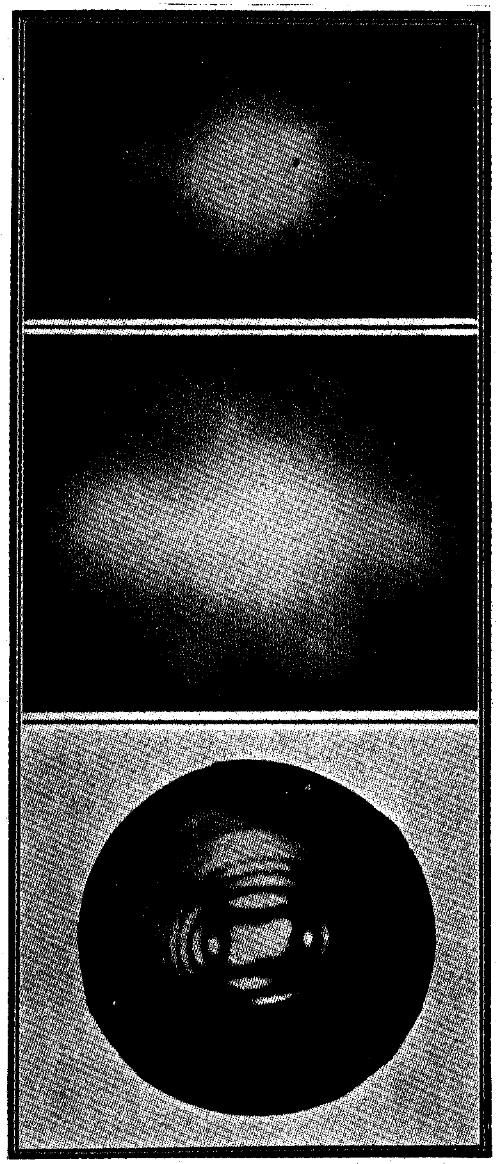
(9) (10)



(11) (12)

Figures 9–12

Plate III



(13)

(14)

(15)

Figures 13–15

Plate IV

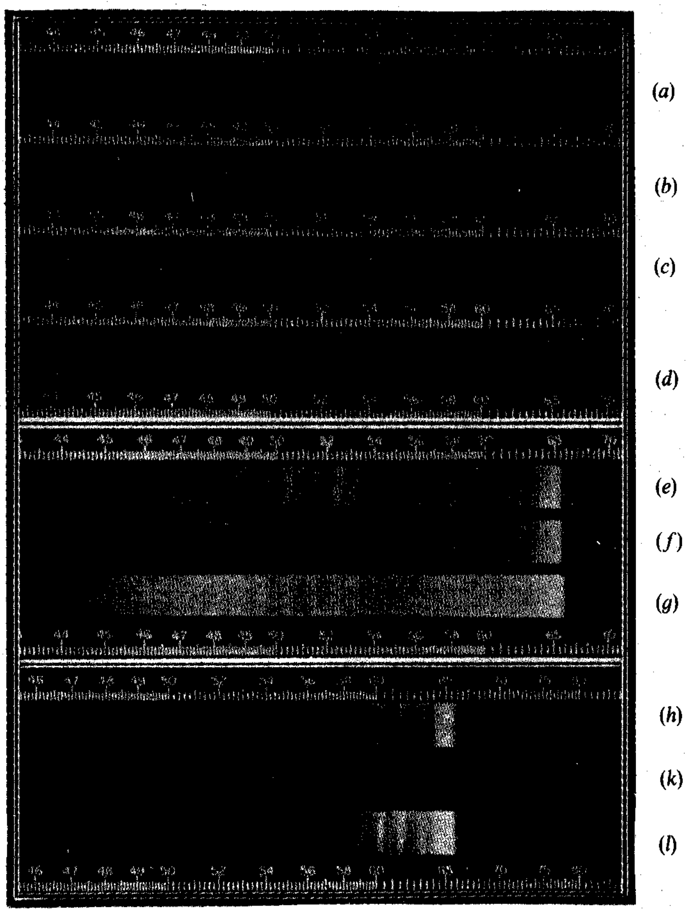


Figure 16

Plate V

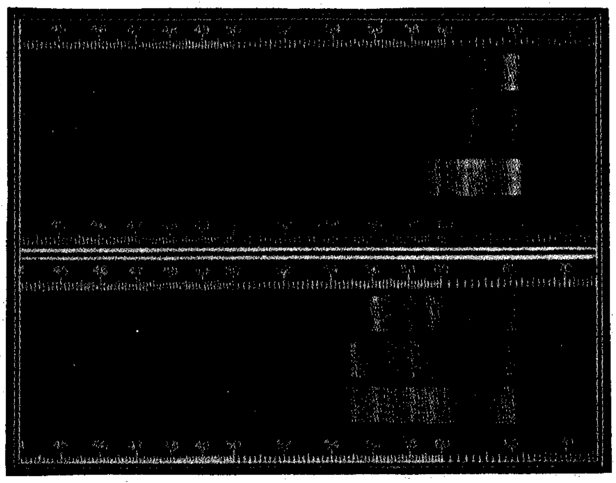


Figure 17

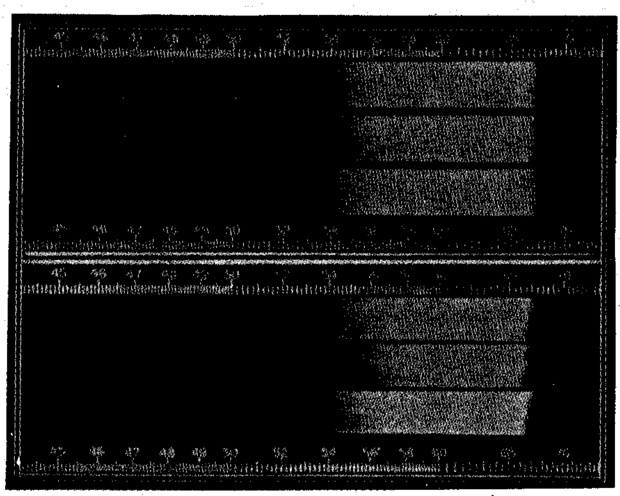


Figure 18

Plate VI