Thoughts on Cause of Color in Precious Opal

by

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It is commonly said that opal is amorphous, but this general statement is, perhaps, not strictly correct. There are so many varieties that they cannot reasonably be put all under one heading. It is true that common opal may be amorphous, but X-ray investigation shows that this mineral has normally some regular internal structure. (References 6 et sequi).

Precious opal must not be confused with common opal, particularly as it obviously contains areas with definite formation. Casual examination will show, in any piece of precious opal, the presence of bright colored patches. With a small loupe these are seen to be composed of numbers of approximately parallel threads lying side by side in each area. Under the microscope (using oblique lighting, not substage illumination), the structure is quite obvious and bears a distinct resemblance to asbestos, crocidolite and chatoyant or cat's eye quartz. The fibres form small sheets, not necessarily flat, and the edges of the sheets are often rectilinear, which implies crystallization, in contrast to the curves seen on the edges of fractures.

It is frequently said that the color is produced by interference caused by cracks, but the patches which show the colors are not similar to the typical fracture of opal, and in nearly all cases the colors are not interference colors (which are necessarily polychromatic), but are pure prismatic monochromatic hues. (See note A). Further, if these are cracks which have been invaded by a secondary incursion, why, it may reasonably be asked, do we never see any larger cracks? Then again no air is found trapped, liquids are enclosed, sometimes with large air bubbles, but in opal no patches of

color have air adjacent to them, as are to be seen, for instance, in iris quartz. It should not be difficult to obtain a piece of opal (either precious or common) which is obviously cracked, so that a comparison may be made between the actual crack and the hypothetical. It will be seen that there is great dissimilarity not only in (a) the outlines, and in (b) the contours of the areas, the one being almost flat, the other nearly conchoidal, but also (c) the internal surfaces of true cracks are not fibrous or striated and (d) the quality of the color generated is quite different, as different as oil on water is from a rainbow.

Some of the earlier writers (Hauy, Behrens, Brewster, Butschli, Reference 1 - 4) suggest that the cracks are regularly striated, if so, would that not prove that the opal is not completely amorphous? The areas are certainly striated, being formed of parallel fibers—layers of silica threads—but they do not appear to be fracture surfaces.

Sosman (Reference 5) has described the precristobalite state of quartz as consisting of silica threads which he says may commence to form at high temperatures while the silica is still gaseous. In the liquid state the simple tetrahedra will have linked up into fragile chains of colloidal dimensions, free to move and orient themselves as the temperature slowly decreases until cristobalite in the solid state is formed. When SiO_4 tetrahedra are generated in water, similar chains will be built up by simple polymerization, forming a colloidal hydrosol. If this gels rapidly, truly amorphous common opal will be produced, but, as stated previously some structure (believed to be cristobalite) is usually present, and when a considerable degree of orientation is possible, due to very slow gelation, precious opal will normally result.

In order that brilliant colors may be emitted it is necessary that, apart from the development of the chains and sheet structure, the inter-ionic spacing must be such that the space lattice constant in any given direction will be a near harmonic of the wave-length of the color of the beam seen from that direction, as is the case in X-ray diffraction work.

Specimens have been obtained showing definite formation on fractured surfaces of precious opal, in the form of striated blocs.

Many pieces of opal (probably over 2000) have been critically examined and indications of a "film-pack" structure are clear. It appears probable that the silica threads in the sol link up into sheet structure (of cristobalite rather than of quartz) and that in good specimens several sheets develop one behind the other like cards in a deck. Where the optical effects are most strong it is suggested that the spacing between the sheets or films is accurately related to the wave-length of a specific color, so that rays entering laterally between the films will be reflected and transmitted frontally in monitored rhythm, each film receiving colored reflections from the one behind and passing these forward in beat with the others. Thus the unusual brilliancy of precious opal—a brilliancy only equalled by that of total internal reflection — is accounted for, the changes of color are explained and also the purity of hue.

Examination of a specimen of precious opal will show that it consists of colored patches with clear opaline material between them. This is clearly visible in water opal and in some Mexican opal, where careful inspection will show that the areas emitting color are actually individual crystalline developments. In high quality gem material the large number of film-packs is confusing, but in less colorful specimens the shape and formation of individual discrete lattices may be studied, and the strength of the evidence appreciated.

Details of the geological conditions necessary for the production of opal need not be given here, but the results of detailed work indicate two major points. First, that the impervious layer of sandstone over the beds — the duri-crust — is essential to the formation of precious opal as it permits very little evaporation below it, so that the sol

takes many years to gel. Secondly, many conditions have to be fulfilled before precious opal, natural silica gel, can be deposed. (See note B). Of these the amount of silica is important, as an excess of this results in the opaque common opal known as potch. However, if the excess is only slight the silica may show as small discrete particles suspended in the gel which give the appearance of an indefinite blue mist, while a little more silica will appear as stronger blue clouds having definite outlines. Further increases cause more cloudiness and pale blue, grey or milky potch forms, according to the size and quantity of the silica particles to (Reference 17).

The production of color may be illustrated by hand specimens or micro-slides showing five nominal stages. First, the indefinite blue misty patches and, second, the definite blue clouds already mentioned. The third stage is best seen in clear water-opal or fire-opal—individual sheets or films, varicolored, tenuous and evanescent, approximately planar, with rectilinear edges.

With further crystal development we get obvious fibers — the main body of the stone is partially amorphous, but in it are areas where the silica threads have had time to orient themselves into bands, regular and fairly bright, each built up of many molecular layers, suitably spaced in the lattice and so reflecting definite wave-lengths. The fifth stage shows the final, yet still incipient, crystallization in the gel. The fibers have linked up laterally to form sheets and these are found to be in blocs or film-packs which control the light so that in any specific direction a monochromatic beam is transmitted, other wave-lengths being damped out.

Successive films act in support of each other, after the manner of resonators, and thus a much greater proportion of the light is emitted than would be reflected from a single unsupported film. (See note C).

In conclusion we may summarize by saying that the colors are not caused by refilled cracks (which would prove a cause of weakness. See reference 15), but apparently by incipient structural development of a crystalline nature whose lattice constants are in mathematical harmony with the wavelengths of the rays emitted.

$$n\lambda = 2c \sin\theta$$
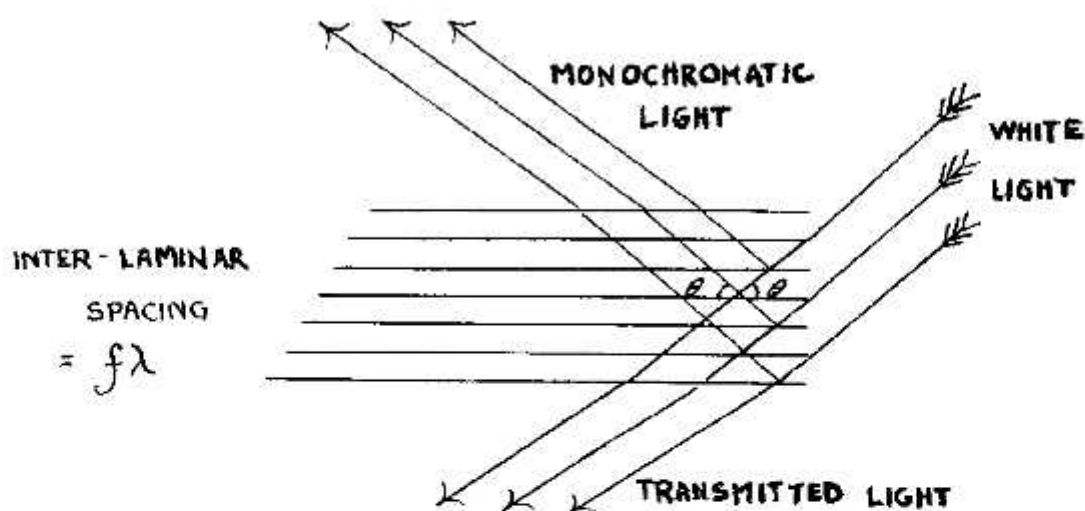
$$\lambda = \text{WAVE-LENGTH}$$

$$c = \text{INTER-LAMINAR SPACING}$$

$$\theta = \text{ANGLE OF REFLECTION}$$

Note A: The colors of thin plates are produced by the cancellation of certain wavelengths, leaving all the others operative — thus if yellow were obliterated we should see a mixture of those remaining — red, orange, green, blue and violet — a rather unpleasant tone, far from monochromatic and deficient in brightness, since all the yellow is missing; these peculiar shades are typical of interference colors, but are never seen in precious opal. Here the white light is analysed by refraction and becomes prismatic. Its monochromatic nature can be demonstrated experimentally, with the sodium, strontium or other flames.

Note B: In order for precious opal to form, the following conditions appear necessary: —

1. The ground waters must have lost all their soluble salts (NcCl, CaSO$_4$ etc.).

2. The ground waters must contain soluble silicates, Na$_2$SiO$_3$ (colloidal).

3. The surface waters must contribute an acid radicle, precipitating silica.

4. Concentration of the solutions must be sufficient to develop a light mobile gel yet not so high as to produce excess silica (potch).

5. The pH value of the initial sol must be suitable (pH5 — pH7).

6. Temperature must be satisfactory. (40° — 60°F).

7. Gelation must be extremely slow, implying impervious strata above.

8. The development of the structure must accord with a suitable wave-length.

Note C: Assuming that each film normally reflects to the front only 20% of the total light falling on it laterally, then we should get a nominal brilliance of 20%. But one film in support will increase this by a further 16% (i.e. 20% of the 80% transmitted by the first); another film as an resonator will add a further 13% and a third a further 10%, making a total of 59% reflected — three times as bright as the original 20%.

REFERENCES

1. L'Abbe Hauy, Traite de Mineralogie, Vol. II, p.456, 1801.

2. Brewster, Reports of the Meetings of the British Association for the Advancement of Science, Appendix, p.9, 1844.

3. Behrens, Structure of Opal, Ber. Akad. Wien., 64:510, 1871.

4. Butschli, Cellular Structure of Amorphous Silica., Verh. Nat. Med. Ver. Heidelberg, 6:237, 1901.

5. Sosman, The Properties of Silica, Ch. Catal. Co., Inc., New York, 1927.

6. Randall, Crystallites in Opal, Zeit. Kristallographie, 75:201, 1930.

7. Krejie & Ott, Formation of Cristobalite, Jour. Phys. Ch. Soc., 35:2061, 1931.

8. Levin & Ott, Crystallinity in Opals, Jour. Am. Ch. Soc., 54:829, 1932.

9. Greig, Crystallites in Opals, Jour. Am. Ch. Soc., 54:2846, 1932.

10. Sosman, Christobalite in Opal, Jour. Am. Ch. Soc., 59:3015, 1932.

11. Dwyer & Mellor, X-Ray Study of Opal, Jour. Roy. Soc., New South Wales, 66:378, 1932.

12. Dwyer & Mellor, Christallization of Silica, Jour. Roy. Soc., New South Wales, 67:420, 1932.

13. Dwyer & Mellor, X-Ray Study of Opal, Jour. Roy. Soc., New South Wales, 68:47, 1933.

14. Talliaferro, Some Properties of Opal, Amer. Jour. Sci., 30:450-474, 1935.

15. Bragg, Atomic Structure of Minerals, Cornell Univ. Press, p.141, 1937.

16. Hurd, Studies on Silica Acid Gels, Jour. Phys. Chem. Soc., 45:1263, 1941.

17. Copisarow, Structure of Hyalite & Opal, Nature, 157:768, 1946.

18. Johnson, Particle Size and the Scattering of Light, Jour. Am. Ch. Soc. 69:1184, 1947.

19. Shull, Micro-Structure of Gels, Jour. Am. Ch. Soc., 70:1410, 1948.

20. Raman, Labradorite, Prod. Ind. Acad. Sci. Sect. A, 31:279, 1950.