fairly well with Ab₈₅An₁₅, obtained from a measurement on the wider lamellae in a basal section.

OPTICAL CHARACTER.—The optical character of the oligoclase has been determined as negative.

INDICES OF REFRACTION.—Indices of refraction were measured by immersing cleavage flakes in liquids whose indices were known, and comparing the refraction of the flakes with that of the liquid. In this way, n_{γ} was found to be somewhat less than 1.554, and n_{α} about equal to 1.542. These values correspond to an oligoclase slightly more basic than the extinction angles indicate.

SPECIFIC GRAVITY.—The specific gravity of the oligoclase, determined by weighing in distilled water at 20°C, is 2.631. This agrees with the optical determinations of the high albite content of the mineral.

REMARKS. — While the various results of determining the composition of the feldspar as detailed above agree fairly well among themselves, they are not as strictly accordant as one would ordinarily expect when it is considered that the measurements were made on orientated sections of very fresh material. When under these favorable conditions such discrepancies arise, the need of caution in stating the composition of feldspar as determined by optical tests on unorientated crystals in a thin section is apparent.

THE COLORING OF THE DIAMOND BY RADIUM RADIATION*

S. C. LIND and D. C. BARDWELL

Rare and Precious Metals Experiment Station, U. S. Bureau of Mines, in Cooperation with the University of Nevada

The results of various investigators attempting to color the diamond by radium radiation have not been entirely uniform and in the main were unsuccessful. Doelter¹ classes diamond as the most difficult of all minerals to color, and reports for the most part failures or very slight coloring. On the other hand,

* This article is a reprint of a paper by the authors which appeared in the October number of the Journal of the Franklin Institute, (Vol. 196, pp. 521-28). Published by permission of the Editor of the Journal and the Director of the U. S. Bureau of Mines.

¹ C. Doelter, "Das Radium und die Farben," Steinkopff, Dresden, 1910, p. 123.

Crookes² obtained in a few cases bluish-green and sage-green colors and A. Miethe³ produced a yellow color from a colorless diamond in one instance.

As reported in the preceding paper,⁴ all minerals except diamond, that can be colored by radium radiation, are colored by the penetrating (beta and gamma) rays transmitted through the glass tube containing the radium salt. Experiments of this character, using 250 mgs. Ra for thirty days on five Cape diamonds of four to ten carats of bright straw-yellow color, failed in all cases to produce any change in color. Two other cut diamonds of the same color were sealed inside a tube with 10 per cent. RaCl₂ for the same length of time. The one which was entirely covered by the 10 per cent. salt became fairly green (about a half grass-green) in thirty days, while the other, lying on the surface of the salt, took on a distinct but fainter green.

It was therefore quite evident that direct (probably alpha) radiation is necessary to color the diamond. This experiment explains why most investigators, working with penetrating radiation only, have failed to produce color changes.⁵ To confirm this, four straw-yellow Cape cut diamonds of two to six carats were completely covered with 10 per cent. Ra Cl₂, sealed in a glass tube, and left for seventy-five days. A deep grass or sage-green was produced which was of quite uniform shade in all, and as far as could be detected, seemed to penetrate through the crystal. No trace of the yellow color appeared to remain.

The apparently deep penetration of color is very surprising, since alpha rays can penetrate diamond to a depth of the order of 0.001 inch only. This will be more fully discussed later. The non-penetrating character of alpha radiation also led to a second consideration, namely, that alpha rays will be able to reach the surface of the diamond only from those layers of salt immediately in contact with it (from a layer less than 0.001 inch). The rest of the salt absorbs its own alpha radiation and is wholly ineffective

² Sir Wm. Crookes, Proc. Roy. Soc., 74, 47 (1904); Nature, 94, 300 (1914); Pluil Tr Roy. Soc., 214A, 433-45 (1914); Sci. Amer. Supplement, No. 2270 (July 5, 1919).

³ A. Miethe, Ann. d. Phys., 19, 633 (1906).

• Jour. Frank. Inst., September, 1923, p. 375. Also Am. Min., 8, p. 171.

⁵ Crookes (*loc. cit.*), who obtained positive results, worked with the direct radiation of radium salts.

JOURNAL MINERALOGICAL SOCIETY OF AMERICA 203

in producing any color changes. This suggested that radium emanation could be much more effectively employed, since at low gas pressure, the absorption in a container not too large will be practically negligible, and the proportion of effective radiation area of diamonds exposed

will depend principally upon the ratio: $\frac{\text{area of diamonds exposed}}{\text{total area exposed}}$

Accordingly seven cut diamonds from 1.5 to 9.5 carats, of original color white to yellow, were sealed in a glass tube (about 10 cms. long and 1.5 cms. in diameter) with 114 millicuries (m.c.) initial radium emanation. The fluorescence was marked in all cases but not uniform, varying through greenish-blue to bluish-green shades without reference to the original color of the stones, which were supposed to represent one Brazilian and all the principal South African fields.⁶ That the intensity of radiation was greater than in 10 per cent. salt was evident from the increased rate of coloring, though this cannot be accurately judged owing to the rapid coloring of the glass tube container. After an exposure of nine days, during which time 91.4 m.c. of emanation disintegrated, the tube was opened and all seven were found to be colored a moderate shade of green. They were put into another tube of the same dimensions and exposed for nine days to an initial quantity of 161 m.c. of emanation. At the end of this time all had deepened in green shade and were nearly as deep as the ones exposed for seventy-five days to 10 per cent RaCl₂. A third exposure was begun, but was continued for only a few hours, owing to the cracking of the glass tube. Upon examining some of the diamonds visually it was observed that they had apparently developed "carbon spots."7 This was very puzzling, as "carbon spots" have never been reported as being produced artificially. The original crystals had been perfect, nor had any "carbon spots" been produced in the four diamonds colored to a deeper shade of green by 10 per cent. RaCl₂. Attempts to remove the spots chemically by washing in ammonia, hot HNO3, hot chromic mixture, organic solvents, etc., failed. Examination under the high-power microscope revealed round black spots like fly specks at a considerable depth below the surface (as much as 1 to 2 mm.), a few of which

⁶ Since all the diamonds examined had passed through commercial channels their origin cannot be given with certainty.

⁷ Small specks of carbon, possibly graphite, which frequently are found in natural diamonds.

were surrounded by lighter brown halos as if the "carbonization" process were spreading from a centre.

The rest of the investigation was devoted to an attempt to solve the puzzling problems presented by the coloring and the "carbon spots," in connection with the nature of the latter, and the production of both at depths beyond the range of alpha particles in the diamond.

The coloring itself can be described in a few words. In five specimens of light-yellow cut diamonds no color by penetrating rays (through glass $\frac{1}{2}$ to 1 mm. thick) could be produced in thirty days, as already stated. In more than thirty specimens of cut and one uncut diamond varying in color from colorless to yellow to brown, no failure to produce color by direct contact with radium salt or radium emanation was encountered. Green and only green color was produced; even the shade was the same for the same radiation intensity and the same original color. Prolonged heating to 500° C. restores the *original* color, leaving one to suppose that the green color is superimposed and merely masks the original color.

An estimation of the initial alpha radiation from emanation received per unit surface of diamond, in the experiment where "carbon spots" were first obtained, indicated that the alpha ray intensity was about five-fold that in the case of diamonds buried in 10 per cent. RaCl₂, when no spots were developed. Assuming that intensity is the controlling factor, exposure in 50 per cent. RaCl₂ ought to produce "carbon spots" also. A test was made with four Cape cut diamonds of about two carats each (three white and one slightly yellow) buried in 50 per cent. RaCl₂ for seventy-four days. A very deep grass or emerald green color was produced, practically identical in shade in all cases, but no "carbon spots" were produced. In all later experiments it was found that "carbon spots" are produced only in contact with emanation, never in salt, but not universally even in emanation, nor could we determine the factor which controls the carbonization. It was apparently not a characteristic inherent in the individual diamonds, because there were no variations among different specimens under the same conditions; either all or none developed "carbon spots" in a given test. One light vellow cut diamond of eight carats, which developed "carbon spots" in emanation and from which they had been removed (see later), failed to develop any on being radiated with 50 per cent. RaCl₂ till it again colored green.

The nature of the spots also remained unsolved. It appeared plausible to suppose that under radiation, diamond, which is known to be an unstable form of carbon at ordinary temperature and pressure, should revert to the stable form graphite, just as white phosphorus has been shown to be converted to red by radium radiation.⁸ That such a process might be taking place appeared probable from the brown halos surrounding a few of the spots. To explain the localization of discrete spots, rather than a general darkening similar to that observed by Crookes⁹ under cathode-ray bombardment, it appeared plausible to suppose that there exist initial "seeding centres" where the change originates and from which it spreads. This theory was greatly shaken if not entirely upset by the observation that prolonged heating in air at a red glow caused the "carbon spots" to disappear permanently. We can hardly assume a reversion to diamond under these experimental conditions. We cannot attribute it to oxidation unless we assume the diffusion of oxygen into and possibly the diffusion of CO2 or CO out of the diamond. A very ingenious hypothesis was proposed by Dr. R. B. Moore, namely, that there may be in the diamond minute pockets of CO₂ or CO under high pressure, not detectable by the microscope, which may be decomposed by the radiation into C and O2 which recombine in loco upon heating. CO is the more probable, since CO₂ is very slightly decomposed, even by intense alpha radiation.¹⁰ The depth of the pockets from the surface also imposes a difficulty since they cannot be reached by alpha rays, and beta and gamma rays are much less effective in producing chemical action. This theory also does not conform with the idea of gradual growth (brown halos); some of the spots become large enough to be visible to the naked eye, and all of them are easily visible with a small hand lens. That the spots are in reality some form of carbon is supported by the fact that the simultaneous exposure in emanation of a large number of different transparent minerals, together with the diamond, gave these spots only in the latter.

A most puzzling question is the explanation of any effect,

⁸ H. Becquerel, Comp. rend., 133, 709-12 (1901).

⁹ Sir Wm. Crookes, Sci. Amer. Supplement, No. 2270 (1919).

¹⁰ E. E. Wourtzel, Le Radium, 11, 346 (1919).

THE AMERICAN MINERALOGIST

either production of color or of "carbon spots," at a depth in the crystal exceeding the range of alpha rays—if one is to suppose the effect due to alpha rays. The question may be divided into two: (1) Are we sure that the effects are deeper than the alpha ray range? (2) Are they due to alpha rays? (1) There can be no doubt about the depth of the carbon spots below the surface, which were observed by the microscope (change of focus) and confirmed by the diamond-cutter who stated it would be necessary to cut away one-half to two-thirds of the crystals (of several carats) to remove the spots. In regard to the depth of penetration of *color* there is much doubt, and that point will therefore be referred to again. Question (2) can best be considered generally for either effect as follows:

The possibilities of the production of effects beyond the range of alpha rays appear to be the following: (a) By penetrating (beta or gamma) rays; (b) by soft beta rays which could not penetrate the wall of the glass container, but which do penetrate diamond sufficiently to produce the effects observed; (c) by a secondary penetrating radiation set up at the diamond surface by some form of non-penetrating radiation (including alpha rays); (d) by transmission by an electronic or atomic "chain effect" of a disturbance set up at the surface, possibly equivalent to (c); (e) (applying to color only) by the diffusion through the crystal of color produced at the surface. Hypothesis (a) can apparently be dismissed by comparing the negative results of exposure outside a tube with the positive ones inside. The possibility of (b)is not excluded, but it should be remembered that such soft radiation could not extend the depth beyond a layer equivalent to $\frac{1}{2}$ to 1 mm. of glass, which would be insufficient to penetrate the entire crystal in the case of large diamonds. Regarding (c), no such form of radiation in diamond is known, nor would the low atomic weight of carbon indicate that it would have much penetration; on the other hand, not much would be required, for the same reason. Both (d) and (e) appear intangible at present. None of these hypotheses receives any support from the observations of Joly¹¹ and of Rutherford¹² on pleochroic halos in mica,

¹¹ J. Joly, "Radioactivity and Geology" (1909), pp. 64-9; *Phil. Mag.* (6), 19, 327 (1910).

 $^{12}\,\mathrm{E.}$ E. Rutherford, "Radioactive Substances and Their Radiation" (1913), p. 310.

206

which have been shown to have exactly the diameter predicted by the range of alpha rays emitted from a point source, and which have persisted through geological periods of time.

We are therefore forced to leave the question of the penetration of the effects unsettled in regard to "carbon spots" and will conclude the discussion with the observations contained in the following paragraphs on the penetration of color.

As far as can be determined optically (even by gem experts), either with the unaided eye or with the microscope, the color produced in diamonds persists throughout the crystal. On the other hand, this is not consistent with a direct alpha-ray effect, though rigidly it does not need to be, since "carbon spots" are produced by some influence beyond the alpha-ray range and possibly color also. But color production is a much more general phenomenon than that of carbonization and need not be produced by the same radiation. Crookes (*loc. cit.*) found that the removal of the surface from a cut diamond destroys the color. This would appear to show that it is a surface coloring only, but it must be remembered that cutting or grinding a substance of such hardness is a prolonged process involving the generation of much heat, so that the destruction of color may be due to a heat effect.¹³

Two attempts were made to solve the question without cutting or grinding. A small brown cut diamond was colored green by radiation and then broken into two or three fragments. Examination under a high-power microscope failed to reveal any surface zone of color; each fragment with a colored (polished) surface appeared to be colored throughout, but the reflection and diffraction of green light from the polished (green colored) surfaces was so great that one could not be sure that the appearance of internal color was not merely due to an optical effect. The masking of an internal yellow or brown by green seems to demand more than a thin surface layer of green, but again one must recall that this thin green layer is produced at polished surfaces from which light is reflected in all directions, and also that the masking is not complete in *thick brown* crystals. An uncut chip of colorless

¹³ It is interesting in this connection that in 1915, F. P. Mennell (*Mineral*ogical Magazine, 17, 202) described the occurrence of natural green diamonds in an alluvial field situated on the borders of the Somabula Forest in Southern Rhodesia. He states that the color is entirely lost in the usual course of cutting and polishing for use in jewelry.

THE AMERICAN MINERALOGIST

diamond was colored green by radiation and then cleaved into two pieces. Viewed through the cleavage plane, the fragment still looked green, but again one could not be sure it was not due to reflection. The question might be settled by covering all the facets but one with foil or paint during radiation, and later covering the colored surface with an opaque covering and looking through the others. This experiment was not attempted.

The question of the color penetration remains unsettled with some evidence in favor of non-penetration, which, however, is not conclusive.

The question of the permanency of the green color to light is a much simpler one. Experiments, both in diffused and direct sunlight, showed that the color is quite permanent, perhaps in the same class with glass colored by radiation, though the observations have not yet extended over so long a period as with glass. This confirms the general rule already stated, that the more difficult a mineral is to color the more permanent the color. As stated, the color can be removed by heating to 500° or higher for half an hour or less (depending on the temperature). In this way green color can be toned down in shade by interrupting the heating at the desired point.

The question of imparting permanent radioactivity to the diamond by radiation by radium, which was the subject of experiments by Sir Wm. Crookes (*loc. cit.*), has not been investigated in the present work, but there seems to be no reason to believe this cannot be explained by "active deposit" (or radium itself in case of contact with radium salt), in the light of adsorption and surface phenomena as now explained by Langmuir and others.

SUMMARY

1. Diamond is not colored by the penetrating (beta and gamma) radiation from radium salt in glass tubes 0.5 to 1 mm. thick.

2. Diamond is always colored green by direct radiation, either from high-grade radium salts or from the gas, radium emanation. No exceptions were met in more than thirty specimens.

3. The green color is apparently light-permanent, but can be removed by heating to a dull red.

4. Yellow and brown diamonds are also colored green, but are restored by heating to their original colors. In the case of large deep brown stones the color masking by green was not complete.

5. Recutting the crystal after coloring removes the green color. This is possibly a heat effect during cutting, and does not prove that the coloring is merely a surface effect.

6. The depth of penetration of the color zone could not be determined. If it is an alpha-ray effect, it would be limited by the range of alpha rays in diamond, about 0.001 inch. But reflection of green light from the highly polished surface interferes with optical observations.

7. Radiation by emanation is more effective than by salts, owing to the greater absorption of alpha rays by the salt itself.

8. In some cases in emanation, but never in salt, the radiation resulted in the production of "carbon spots" in the interior of the crystal. The black spots are probably a form of carbon, the nature of which was not definitely determined. They can be removed by prolonged heating in a blast flame.

9. The "carbon spots" are undoubtedly beyond the range of alpha rays in the crystals, which suggests that the color may be penetrating also and that both are produced by some form of secondary radiation more penetrating than alpha rays.

NOTES AND NEWS

MINERAGRAPHIC NOTES ON MANGANESE MINERALS. ERNEST E. FAIRBANKS. Somerville, Mass.

Coronadite from the Coronado vein, Clifton, Graham County, Arizona, was examined in polished section and found to consist of a mixture of hollandite (psilomelane) and an unidentified lead mineral. Lindgren,¹ who first described coronadite, shows that it consists chiefly of lead and manganese oxides.

A polished section gave the following results: galena white, very brittle mineral with a hardness of about 5.5 and with a brown streak. Throughout this mineral there is a finely disseminated substance which shows polarization with crossed nicols.

Following are the microchemical tests made on the surface containing the two minerals:

HNO₃-Fumes tarnish slightly, otherwise negative.

HCl-Fumes tarnish slightly, also faint tarnish produced which is not persistent. KCN-Negative.

FeCl₃-Tarnishes brown, rubs lighter, and is fairly persistent.

¹ Lindgren, Waldemar; Copper deposits of the Clifton-Morenci district, Arizona, U. S. G. S. Prof. Paper 43, (1904) pp. 103-6.

209