

## MINERALOGICAL NOTES

### A STUDY OF SOME GOLD MINE DIAMONDS

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#### ABSTRACT

A study of diamonds recovered from gold mines suggests that the stones were subjected to charged particle irradiation and subsequent annealing of the radiation damage at temperatures not exceeding 500°C. Other unique absorption features are thought to be the consequence of the annealing process time-scale.

#### INTRODUCTION

Recently a small diamond was recovered in cleaning out a tube mill on one of the Witwatersrand gold mines. This rekindled interest in some other diamonds, 38 in all, that have been found in gold mines from time to time and are now in museums. In some of the older gold recovery processes the crushed ore was sent over corduroy which retained the gold particles in its folds and allowed the lighter materials to be washed over. Being relatively heavy the diamonds were found with the gold. A non-destructive study of these unique diamonds, which have kindly been made available to us, has now been made.

#### APPEARANCE AND MORPHOLOGY

The 38 stones, the largest of which weighs 1.53 carats and the smallest 0.08 carats are shown in Figure 1. As can be seen they are all of reasonable crystallographic habit and a few well-shaped octahedrons and dodecahedrons are in evidence. The diamonds are all colored various shades of yellowish-green or green, ranging from a barely perceptible green tinge in one instance to a dark-green, almost black, color in another. The coloration appears to be only skin-deep as is the case, in our experience, with most green diamonds. The skin-deep coloration is consistent with the diamonds having been irradiated with high energy alpha- or beta-particles (Dugdale, 1953). These emanated, in all probability, from uranium (or one of its degradation products) since this element is abundantly present in the gold mines where the diamonds were found.

#### TYPE DESIGNATION

Robertson *et al.* (1934) were the first to draw a distinction between Type I and Type II diamonds and noticed that diamonds of the latter category have nondescript shapes as opposed to those of the Type I variety with good crystallographic features. Kaiser and Bond (1959) showed that all Type I diamonds contain varying amounts of nitrogen in substi-



FIG. 1. Diamonds recovered during gold mining operations. They are all colored consistent with having been irradiated by high energy alpha or beta particles.

tutional position in the diamond lattice and that they are characterized by a strong absorption band at  $7.8\mu$  in their infra-red spectra. Elliot (1960) proposed that this nitrogen in Type I diamonds is segregated into a layer configuration and Evans and Phaal (1962) observed platelets in (100) planes in these diamonds, which could only be explained as being due to segregated substitutional nitrogen atoms. A qualitative study of the infra-red absorption spectra of the 38 diamonds revealed the presence of the  $7.8\mu$  band in all instances. It is concluded, therefore, that as is the case with about 98 percent of all diamonds from more conventional sources, the gold mine stones are all of the Type I variety, *i.e.* they contain appreciable amounts of substitutional nitrogen in platelet configuration.

#### OPTICAL ABSORPTION FEATURES AND DISCUSSION

Three octahedrons with relatively smooth parallel-sided faces lent themselves to optical studies of a more quantitative nature. The optical spectra obtained for these three stones showed identical absorption features although the strength of the bands varied in degree. Spectra were recorded at liquid nitrogen temperature ( $80^{\circ}\text{K}$ ) and a typical spectrum

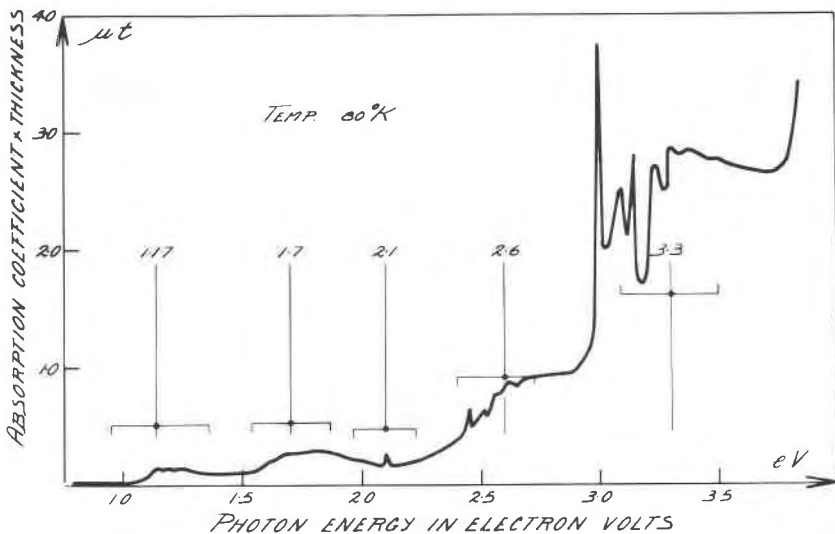


FIG. 2. A typical absorption spectrum.

is reproduced in Figure 2, where the product of absorption coefficient and thickness of the diamond is plotted as a function of the photon energy of the incident radiation.

As can be seen there are prominent absorption band systems centered at 3.3, 2.6 and 1.7 eV respectively. In addition, there are present two absorption systems with main bands at 2.1 and 1.17 eV respectively which to our knowledge have never been observed in diamond before. It is probable that these are a result of annealing of irradiation damage in the diamond over many thousands of years at a temperature which might have been well below but not exceeding 500°C.

The 1.7 eV band, designated G.R.1, is known to be associated with radiation damage in diamond (Clark *et al.*, 1956). Experiments in this laboratory have shown that on heating irradiated Type I diamonds to 500°C G.R.1 can be annealed out completely with time to produce the band at 2.6 eV. At lower temperatures G.R.1 is annealed out only partially and an equilibrium with the 2.6 eV band is reached dependent on the temperature of heating.

Since both the G.R.1 and 2.6 eV bands are present in the gold mine diamonds the inference is that these diamonds were initially irradiated by a natural radioactive source, and at some subsequent stage in their histories were heated to a temperature not in excess of 500°C, and most likely considerably less than this value, to partially anneal out the G.R.1 system.

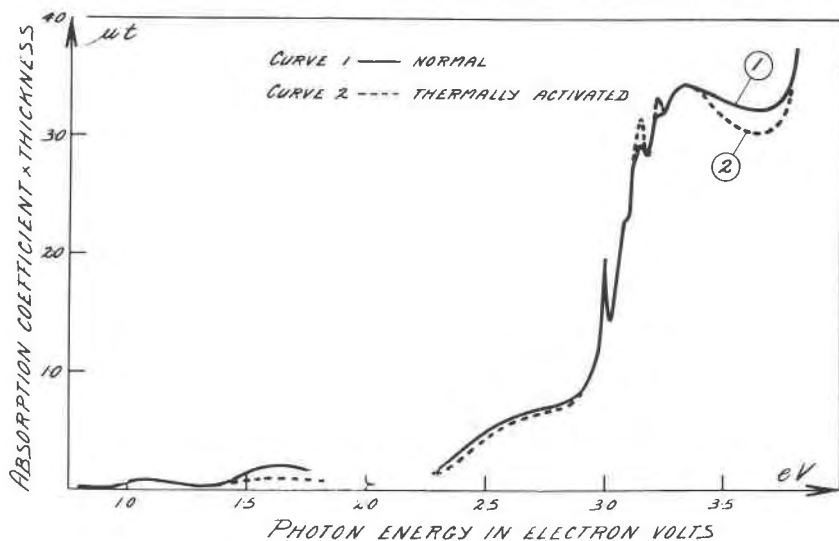


FIG. 3. The absorption spectrum in its normal and thermally activated states.

Figure 3 depicts the absorption spectra at room temperature of one of the selected octahedrons before and after the diamond had been heated in the dark at  $400^{\circ}\text{C}$  for 10 minutes. Curve 1 is that of the diamond in its normal state whereas curve 2, superimposed on curve 1, is that of the diamond in its thermally activated condition. On exposure to ultra-violet light or daylight curve 2 reverts to curve 1, *i.e.* the activated state reverts to the normal condition.

As can be seen there is a difference in light absorption between the normal and activated states. By heating the diamond in the dark the G.R.1 and 2.6 eV systems are reduced and the one at 3.3 eV enhanced. The G.R.1 and 2.6 eV bands being in the visible part of the spectrum, one would expect a visible color change to accompany the heating and light exposure cycles. This is indeed the case. After heating in the dark at  $400^{\circ}\text{C}$  for a few minutes the diamond appears yellow, which rapidly changes to yellowish-green or green on exposure to light. Similar observations have been made in this laboratory on natural Type I diamonds which have been irradiated (Dyer and du Preez, 1965). Our understanding of this chameleon behavior of diamond substantiates the claim that the gold mine diamonds were subjected to irradiation as well as annealing of the radiation damage at temperatures probably very much less than, but not exceeding,  $500^{\circ}\text{C}$  at some stage during their existence.

## ACKNOWLEDGMENTS

Thanks are due to the Anglo American Corporation of South Africa Limited for the loan of the diamonds and to Industrial Distributors (1946) Limited for permission to publish this paper.

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THE AMERICAN MINERALOGIST, VOL. 54, JANUARY-FEBRUARY, 1969

ABOUT THE STRUCTURE OF IOWAITE<sup>1</sup>

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## ABSTRACT

Iowaite, which weathers to pyroaurite and is similar to it in crystal data and powder-line intensities, should be isostructural with it and be reformulated as  $[\text{Mg}_4\text{Fe}(\text{OH})_{10}]^+ \cdot [\text{Cl}(\text{H}_2\text{O})_{x-1}]^-$ ,  $x=4$ ,  $Z=3/5$ .

Kohls and Rodda (1967, to be referred to as KR) have recently described the new mineral iowaite,  $4\text{Mg}(\text{OH})_2 \cdot \text{FeOCl} \cdot x\text{H}_2\text{O}$  ( $x \leq 4$ ). All the data reported by them indicate a strong similarity (Table 1) between iowaite and pyroaurite. The latter is one of two polymorphic modifications, described as pyroaurite I and pyroaurite II by Aminoff and Broomé (1930). Frondel (1941), who established the presence of  $\text{CO}_3$  in the chemical formula, called the hexagonal modification sjögrenite, reserving the term pyroaurite for the rhombohedral one. Allmann and Lohse (1966) determined the crystal structure of sjögrenite with structural formula  $[\text{Mg}_6\text{Fe}_2(\text{OH})_{16}]^{2+} \cdot [\text{CO}_3(\text{H}_2\text{O})_4]^{2-}$  and, for pyroaurite, proposed a layer sequence that was later confirmed by two structure determinations independent of each other (Ingram and Taylor, 1967; Allmann, 1968).

In view of the similarities in crystal data (Table 1) and in powder-line intensities, further supported by the fact that iowaite alters to pyroaurite on weathering (KR), it would seem that the structures of these two

<sup>1</sup> Work supported by NSF grant GA 1343 to The Johns Hopkins University.