

## Possible origins of $\alpha$ -damage in diamonds from kimberlite and alluvial sources

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**SUMMARY.** Heating experiments provide strong evidence that the transparent green coats on some diamonds from each of many localities are caused by  $\alpha$ -particle irradiation after kimberlite injection and subsequent cooling. The natural diamonds with more or less homogeneous transparent green coats, studied in this work, appear to have received doses of at least  $5 \times 10^{13}$ – $1 \times 10^{14}$   $\alpha$ .cm<sup>-2</sup>. For Pre-Cambrian kimberlites, such doses could occur if certain regions of the diatremes contained  $\geq 20$  ppm by weight of equivalent uranium, after kimberlite injection and solidification.

Such considerations lead to the prediction that the radioelement concentrations in the Finsch kimberlite diatreme and the Bellsbank fissure kimberlite are considerably greater than those in the Premier mine, though radioelement segregation could produce the required local concentrations. Some exploratory autoradiographic measurements made on two kimberlite rock samples from Premier and De Beers Mines indicated that the radioelements were apparently distributed on a sub-micron scale, which would be a necessary, but not sufficient, condition for uniform  $\alpha$ -irradiation of diamonds.

Diamonds from various alluvial sources showing green and brown spots arising from much heavier and more localized radiation damage are also discussed.

CROOKES (1904) showed that  $\alpha$ -particles emanating from radium salts induced a green coloration in diamonds and similar results were reported by Lind and Bardwell (1923). Green diamonds occur in many kimberlite and alluvial diamond productions, and the occurrences may be divided into: true (body) colours, opaque coats of variable thickness, transparent coats, about 20  $\mu$  thick, the colour ranging from pale green to green, and transparent spots of variable size and shape, also about 20  $\mu$  thick, the colour ranging from pale green to very dark green.

Diamonds with a green body colour are rare (Wagner, 1914) although we have observed a number of olive-green stones from the Finsch Mine, South Africa. Diamonds with opaque-green coats are common in the alluvial production from the Congo (Kinshasa) (Polinard, 1929) and are also found in West Africa (Grantham and Allen, 1960).

Diamonds with transparent green spots or coats are found in many kimberlite and alluvial productions, and in the present work, they have been observed in the productions from Angola (an assorted parcel), the Congo, Kinshasa (from the Massif's 2, 3, and 5 diatremes and associated alluvials along the Bushimaie river), Ghana (an alluvial source), Ivory Coast (an assorted parcel), Sierra Leone (from Koidu Nos. 1 and 2

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kimberlite diatremes and associated alluvials), South West Africa at Consolidated Diamond Mines, Oranjemund (an alluvial source), and in South Africa from the Namaqualand coastline, and the Bobbejaan (Bellsbank), Bultfontein, Finsch, Jagersfontein, and Premier Mines. Other authors have noted similar diamonds from the Banket Conglomerate, South Africa (Denny, 1897; Raal, 1969), from diamond provinces in Siberia (Bobrievich *et al.*, 1959, p. 475), from Brazil (Leite, 1969), and from Australia, India (Majhgawan kimberlite diatreme, Panna), Liberia, Mali, Uganda, and Thailand (Grantham, unpublished work). Isolated specimens from the Banket, Brazil, and Venezuela have also been examined in the present study.

It is found in practice that a transparent green-coated diamond can be non-destructively recognized by viewing the specimen through an approximately flat face, there being a distinct concentration of colour around the edges of the stone. For diamonds with transparent green spots, it is easy to observe optically that the green coloration is confined to the surfaces (see Schiffmann, 1969).

Since the thickness of the transparent green coats or spots is about the penetration distance in diamond of  $\alpha$ -particles from U, Th, and their daughter elements, it has been proposed that such coats and spots arise from  $\alpha$ -particle irradiation (Wagner, 1914, p. 147). However, there appears to be a lack of even semi-quantitative confirmation of this proposition, although Vance and Milledge (1972) have shown that very dark green *spots* on diamond could arise from direct contact with U-rich or Th-rich minerals over a period of  $\approx 10$  Myr.

The principal aim of the present work was, firstly, to show that diamonds with transparent green coats owe their coloration to  $\alpha$ -particle irradiation and, secondly, to investigate whether the doses could arise from the radioelements now present in kimberlite.

A further object was to discuss green- and brown-spotted diamonds from various sources.

*Experimental.* The transparent green-coated diamonds used in the heating experiments to be described were from Ghana, Sierra Leone, and from the Finsch and Premier Mines, South Africa.

For the  $\alpha$ -irradiations, the source was a  $0.55 \text{ Ci. cm}^{-2} \text{ }^{242}\text{Cm}$  disc,  $1 \text{ cm}^2$  in area and a fraction of a micron in thickness. A thin Al window was placed over the source and the emergent  $\alpha$ -particles had an energy of *c.* 5.5 MeV after perpendicular traversal of the window. The irradiations, carried out at A.E.R.E., Harwell, provided doses of  $1 \times 10^{12}$ ,  $3 \times 10^{12}$ ,  $1 \times 10^{13}$ ,  $3 \times 10^{13}$ , and  $1 \times 10^{14} \alpha. \text{ cm}^{-2}$  as Vance and Milledge (1972) showed that a dose of order  $10^{14} \alpha. \text{ cm}^{-2}$  was sufficient to impart a green coloration to an initially colourless diamond. The experimental geometry was such that about eight samples were irradiated at once, the samples being  $< 2 \text{ mm}$  from the source and being close to its centre. Under these conditions, all surfaces adjacent to and parallel to the source would receive the same dose (per unit area) in a particular irradiation. The diamonds used were from the Premier Mine and the specimens consisted of colourless diamonds with cleavage surfaces (the cleavage surfaces being irradiated) as well as unbroken yellow and brown diamonds.

For autoradiography,  $\alpha$ -sensitive KO emulsion plates (Ilford) were used, the exposures being 37 days. The emulsions were placed in direct contact with flat kimberlite surfaces, produced by diamond-sawing of fresh kimberlite rocks. One kimberlite was from the 1640 ft level of the De Beers Mine and was basaltic in texture; the other was a kimberlite, containing a fairly high proportion of xenolithic material, from the 1060 ft level of the Premier mine.

The heating experiments were carried out in air, using an electric oven fitted with an automatic temperature controller, and also in an induction furnace with a graphite crucible, under a dynamic vacuum of better than  $10^{-4}$  mm Hg at temperature.

### *Results and discussion*

*Laboratory  $\alpha$ -irradiation.* The minimum dose required for an initially colourless cleavage surface of diamond to show a green coloration, observable under a low-power microscope with white light illumination, was found to be between  $3 \times 10^{13}$  and  $1 \times 10^{14}$   $\alpha$ .cm $^{-2}$ .

Unfortunately, no coloured diamonds were included in the group of specimens irradiated to  $1 \times 10^{14}$   $\alpha$ .cm $^{-2}$ , although it is very likely that the body colours of such stones would preclude visual observation of induced surface coloration at that dose level.

*Heating experiments.* Though various workers have shown that both diamonds rendered green by  $\alpha$ -irradiation (Anderson, 1963; Vance and Milledge, 1972) and green-spotted diamonds (Meyer *et al.*, 1965; Raal, 1969; Vance and Milledge, 1972) turn brown on heating to 500 to 600 °C, no directly comparative experiments appear to have been carried out on both laboratory-irradiated (green) diamonds and natural specimens with transparent green coats. For heat treatments of 1 hr in air, no colour change was found in either kind of diamond at 500 °C, both kinds went an olive colour at 550 °C, and were rendered brown at 600 °C. Both kinds of specimen turned brown on heating for 30 min at 750 °C under dynamic vacuum conditions. Subsequent etching in molten KNO $_3$  at 750 °C for 30 min confirmed the absence of a body colour in the specimens that were not irradiated in the laboratory.

Hence it was concluded that the transparent green-coated diamonds studied in the present work owed their coloration to  $\alpha$ -particle irradiation over their entire surfaces. However, the artificially irradiated specimens were irradiated on one surface only, so there is some uncertainty in direct comparison of the doses received by natural and laboratory-irradiated specimens. Nevertheless, it is estimated that the natural diamonds with transparent green coats that were examined in this work had received  $\alpha$ -doses of at least  $5 \times 10^{13}$ – $1 \times 10^{14}$  cm $^{-2}$ .

Since the intensity of colour in the natural specimens was generally weak, it is not surprising that we have found no natural diamonds with a body colour to exhibit a green transparent coat, though some coloured diamonds were found that exhibited fairly intense green spots.

The possibility that opaque green coats and brown and green body colours in diamond are due to  $\alpha$ -particle damage from finely dispersed radioactive inclusions was

considered. It is assumed that kimberlite breakthrough took place at temperatures such that the kimberlite was molten (see Kennedy and Nordlie, 1968; Dawson, 1971; Harris and Vance, 1972; but see also Harris and Middlemost, 1969). If internal  $\alpha$ -irradiation of a diamond continued for an adequate period after cooling to below about 500 °C, it would be expected that internally  $\alpha$ -irradiated diamonds would be at least olive-green, if not green. For the Premier Mine, which has a breakthrough age of *c.* 1750 Myr (Allsopp *et al.*, 1967), this argument should especially apply. But in the Premier Mine a large proportion of diamonds are brown, and olive-green and green diamonds are rare. Hence it seems that not all brown diamonds owe their colour to internal  $\alpha$ -irradiation.

Three olive-green Finsch diamonds (true body colours) and two diamonds with opaque-green coats, from Sierra Leone, were heated to 750 °C for 30 min under dynamic vacuum conditions. No colour changes were observed, which indicates that not all green diamonds owe their colour to  $\alpha$ -irradiation.

*$\alpha$ -Particle irradiation in kimberlite.* If the radioelements in kimberlite were uniformly distributed and the temperature was  $\leq 500$  °C, an even green coloration would be produced on the surface of an initially colourless diamond, assuming enough time and a sufficient concentration of radioelements to be available. However, if the radioelements in kimberlite were in the form of isolated particles rich in radioelements, such as oxides, a coat consisting of isolated green spots would be expected. As the range of  $\alpha$ -particles from U, Th, and their daughter elements is about 20  $\mu$  in such materials as diamond (Crookes, 1904; Vance and Milledge, 1972) and Si (Colella and Burgeat, 1969), it is assumed that the range of similar  $\alpha$ -particles in kimberlite is also about 20  $\mu$ . An  $\alpha$ -particle will produce appreciable radiation damage on a diamond surrounded by kimberlite only if it has traversed less than a critical distance,  $\lambda_c$ , in the kimberlite before striking the diamond. Assuming  $\lambda_c \approx 15 \mu$ , the radioelement-rich regions would have to be  $\leq 30 \mu$  apart, and *approximately equally spaced*, to produce an approximately uniform coloration.

The  $\alpha$ -dose received by a diamond in kimberlite depends on the product of the radioelement concentration of kimberlite and the time of irradiation, and the green coloration in diamonds derived from kimberlite must represent the dosage received after the temperature was  $\leq 500$  °C, i.e. sometime after breakthrough (see above). Assuming a uniform distribution of the radioelements in kimberlite, over regions greater than the size of a diamond, the  $\alpha$ -dose,  $D \cdot \text{cm}^{-2}$  received by a diamond can be calculated by first principles as:

$$D \cdot \text{cm}^{-2} = 8\rho[\text{EU}](\lambda_c/4) \times 10^{-10} (\lambda T)N_0/\bar{A}_{\text{EU}} \quad (1)$$

where  $\rho$  = density of kimberlite ( $\text{g cm}^{-3}$ ),  $[\text{EU}]$  = EU concentration<sup>1</sup> in ppm by weight,  $(\lambda T)$  = product of the decay constant of EU and the irradiation time ( $\lambda T$  assumed to be  $\ll 1$ ),  $N_0$  = Avogadro's Number,  $\bar{A}_{\text{EU}}$  = mean atomic weight of EU, and  $\lambda_c$  = critical penetration distance in  $\mu$ . For  $\rho \approx 2.7 \text{ gm.cm}^{-3}$  (Dawson, 1960),

<sup>1</sup> The equivalent uranium (EU) concentration of a material is that uranium concentration which has the same  $\alpha$ -activity as the actual radioelement mixture in the material.

$T = 100$  Myr and  $\lambda_c \approx 15\mu$ , 15 to 30 ppm of EU by weight would be required to produce a dose of  $5 \times 10^{13}$  to  $1 \times 10^{14}$   $\alpha \cdot \text{cm}^{-2}$ , and thus a transparent green coat on an otherwise colourless diamond.

To relate these figures to kimberlite sources, it must be emphasized that only a fraction of colourless diamonds in kimberlite show green coats, but that on green-coated diamonds the coats appear to be more or less uniform. To explain this observation, the EU must be distributed uniformly over volumes comparable in size to that of the diamond, but must be inhomogeneously distributed over distance  $\gg$  diamond dimensions. Since it is likely that U and Th ions will not readily enter into solid solution with the principal minerals that may crystallize from the original kimberlite magma (see, for example, Larsen and Phair, 1957, and Kleeman *et al.*, 1969), and again assuming kimberlite to be molten before injection, it is likely that U and Th will be concentrated into the last regions of the kimberlite diatreme to solidify after injection. Hence, in kimberlite, a radioelement distribution of the type mentioned above seems not unreasonable.

Dawson and Milledge (to be published) have found that the EU content of various kimberlites is of order 10 ppm by weight, so that for a Pre-Cambrian kimberlite,  $\alpha$ -doses sufficient to induce green coloration in diamond could well occur in certain diatremes, after injection.

Though the published information on the injection ages of kimberlite diatremes is scanty, it is still possible to draw some tentative conclusions about the radioelement contents of various kimberlites.

The age of the injection of the Premier Mine kimberlite has been given as 1750 Myr (Allsopp *et al.*, 1967), so from equation (1), *c.* 10 to 20 ppm EU by weight would produce transparent green coats on otherwise colourless diamonds. However, over the last few years, a study of some  $2 \times 10^5$  diamonds from the Premier Mine showed that transparent green-coated diamonds are rare in this mine, and it is suggested that, in the presently-mined part of the Premier Mine, there is *c.* 10 ppm by weight of EU (assuming an age of 1750 Myr). An EU value of 9.4 ppm by weight has been found for a 100 mg sample of Premier Mine kimberlite (Dawson and Milledge, to be published).

A parallel study of approximately  $1 \times 10^5$  Finsch diamonds shows the abundance of diamonds with transparent green coats to be at least 10 times that for the Premier Mine. Though the age of the Finsch diatreme is unknown, it may be of Post-Jurassic age (Dawson, 1970). Thus assuming the Finsch kimberlite to be approximately 10 times younger than the Premier Mine, with a higher abundance of apparently irradiated diamonds, the EU content of Finsch could well be  $\geq$  10 times that of Premier, although radioelement segregation effects may be more pronounced in Finsch than in Premier.

The Bellsbank kimberlite also contains a fairly high proportion of transparent green-coated diamonds, and although its age is unknown, it too may be Post-Jurassic (Dawson, 1970). Hence the EU content of Bellsbank should be approximately the same as that of the Finsch Mine and much greater than that of the Premier Mine. This conclusion is partially confirmed by the work of Gurney and Hobbs (to be

published), who have found an especially high EU value of 25.5 ppm by weight for Bellsbank kimberlite, but as yet EU values have not been determined from the kimberlite at the Finsch Mine.

An insufficient number of diamonds have been examined to draw any conclusions on the abundance of transparent green-coated stones from the other kimberlite sources.

During the examination of the  $2 \times 10^5$  diamonds from the Premier Mine (see above), one diamond was found that had a small transparent green spot in juxtaposition with a transparent brown spot of similar shape. This observation may possibly be explained in the following way: After injection and solidification of the kimberlite, this diamond was in contact with a small grain of material, the EU content of which was greater than that of the surrounding environment (the EU content of the general environment being insufficient to render the diamond green all over). After cooling below *c.* 500 °C, the diamond received an  $\alpha$ -particle dose sufficient to generate a green spot, and this diamond was then subsequently heated to above about 600 °C, changing the original green spot to a brown colour. After cooling once more to temperatures below *c.* 500 °C, a new green spot was formed, but a slight movement of the original EU-containing grain had occurred.

Assuming the green and the brown spot to have been caused by the same EU-rich grain, then since the intensity of both spots was approximately the same, this suggests that the reheating event occurred roughly half-way between the injection age of the kimberlite and the present time. The age of the Premier Mine has been determined as 1750 Myr (Allsopp *et al.* 1967), and since it is unlikely that the Premier diatreme was an active volcanic system for *c.*  $\frac{1}{2}$  (1750) Myr, it is improbable that the reheating event was a further kimberlite injection. However, *c.* 1115 Myr ago, a gabbroic sill cut part of the Premier diatreme (Allsopp *et al.* 1967) causing a thermal aureole in the kimberlite. Hence, if the diamond under discussion was within the zone of thermal alteration in the kimberlite diatreme, the original green spot could have turned brown, and after the sill cooled a second green spot could have formed, having about the same intensity of coloration as the brown spot.

*Autoradiography.* As stated above, to produce an even transparent green coat on a colourless diamond by  $\alpha$ -irradiation, the radioelement concentration must be reasonably uniform throughout the kimberlite surrounding the diamond. In an attempt to investigate the EU distribution in kimberlites, two kimberlites were examined using  $\alpha$ -sensitive emulsions.

On one of the emulsions was placed a flat piece of gem-quality zircon, of density 4.0 g.cm<sup>-3</sup>, which contained about 5700 ppm by weight of EU (Vance and Anderson, 1972). A study of the  $\alpha$ -tracks derived from the zircon enabled subsequent  $\alpha$ -track recognition to be quite confident in emulsions exposed to kimberlite. The measured track density from the zircon (the observed tracks were nearly continuous and 5 to 20  $\mu$  in length), was  $(1.6 \pm 0.1) \times 10^4$  mm<sup>-2</sup>. As with  $\alpha$ -particle irradiation of diamonds in kimberlite,  $\alpha$ -particles from zircon will have insufficient energy to produce a recognizable track in the emulsion if they traverse more than a certain distance in the zircon

itself. The calculated value for the critical distance,  $\lambda_c$ , for the zircon was  $8\mu$ , knowing the track density and the EU content (see equation 1). This value was also used, as an approximation, for the kimberlite samples.

Areas on the emulsions corresponding to slightly more than  $1\text{ cm}^2$  of each of two surfaces cut from the Premier Mine kimberlite were used for the track counting, as well as slightly more than  $1\text{ cm}^2$  of each of three surfaces of the basaltic kimberlite from the De Beers Mine. Over the entire areas surveyed, only three four-pronged 'stars' were observed, and assuming such stars were caused by local radioelement-rich regions, (uraninite grains for example), it is readily calculated that such regions could not have been larger than about  $1\mu$ .

However, the overwhelming majority of the  $\alpha$ -tracks caused by the kimberlite on the emulsion were in the form of single tracks, so that even if the radioelements in kimberlite were in the form of radioelement-rich regions, the size of such regions must have been  $< 1\mu$ . The largest concentration of single tracks observed was 32 in a region of  $0.01\text{ mm}^2$  from the De Beers kimberlite, corresponding to a local EU concentration of about 1500 ppm by weight (see equation 1).

The 'background'  $\alpha$ -track density, caused by scratches, etc., resembling  $\alpha$ -tracks, taken over  $0.3\text{ cm}^2$  of emulsion not in contact with kimberlite (and shielded from the zircon standard), was measured as  $0.6 \pm 0.2\text{ mm}^{-2}$ . The track densities for the three regions of the De Beers basaltic kimberlite were found, after background subtraction, to be  $6.0 \pm 0.3$ ,  $5.8 \pm 0.4$ , and  $4.6 \pm 0.4\text{ mm}^{-2}$ , and in the two regions of the brecciated kimberlite, the values were  $3.1 \pm 0.4$  and  $2.8 \pm 0.4\text{ mm}^{-2}$ . Assuming the average density of kimberlites to be about  $2.7\text{ gm cm}^{-3}$  (Dawson, 1960), the averaged EU concentrations for the basaltic and brecciated kimberlites were calculated as  $2.8 \pm 0.5$  and  $1.5 \pm 0.5$  ppm by weight respectively. These values are lower by a factor of approximately 5 than those found by Dawson and Milledge (to be published), but in view of the dilution of the brecciated kimberlite by xenoliths, and the likelihood of radioelement segregation in a diatreme after injection, the discrepancy is not serious.

Because of the low  $\alpha$ -track densities, it was not possible to reach any conclusions about the uniformity of the radioelement distribution in the kimberlite samples. But it could be concluded that the sizes of possible radioelement-rich regions were less than about  $1\mu$ , a necessary, but not sufficient condition for transparent even green coats on diamonds in kimberlite (see above).

#### *Diamonds from alluvial sources*

For diamonds showing green surface coloration, there is perhaps only one alluvial source in which radioelements are definitely associated with the diamonds (see table I and below). These diamonds tend to exhibit either multiple dark green or dark brown spots, although diamonds with similar characteristics have been found in alluvial sources whose radio element content is not proven (see Table I). In the other alluvial deposits, however, the diamonds tend to exhibit either an even transparent coat, varying in colour from green to very pale green, or pale green spots. Where the source of the alluvial diamonds is known (see below), the variation in green surface colour corresponds closely to that found on the diamonds from the associated kimberlite

source. Hence with the majority of alluvial diamond deposits, the transparent green coats or spots found on these diamonds are probably derived from radioelements in the original kimberlite, and not from detrital grains of radioelement-rich materials, accumulated with the diamonds in the alluvial deposit. If intense green-spotted diamonds are found in these alluvials, however, their presence probably indicates that these diamonds were derived from an earlier alluvial source associated with

TABLE I.  $\alpha$ -irradiated diamonds from alluvial sources

Type of coloration	Source
Intense green and/or intense brown spots on the same diamond	Rand Banket, South Africa (associated with radioelement-rich minerals); Angola, Brazil, and Venezuela
Intense brown spots	Peninsular Thailand, Western Mali
Intense green spots	Ivory Coast
Pale green spots or coats	Congo (Kin.), Sierra Leone, Ghana, South West Africa, Namaqualand (South Africa)

radioelement-rich minerals, and were not directly weathered from a kimberlite source into the alluvial deposit. The intensity of the dark green colour corresponds to localized doses of  $c. 10^{17} \alpha \cdot \text{cm}^{-2}$  (Vance and Milledge, 1972), whereas diamonds whose green surface colour was derived from the original kimberlite appear to have received fairly uniform doses of  $\approx 10^{14} \alpha \cdot \text{cm}^{-2}$  (see above).

On some diamonds from alluvial deposits, but not from kimberlite sources, *intense* green spots and *intense* brown spots may occur on the same diamond (see Table I). This could arise if a diamond received a localized  $\alpha$ -dose of  $c. 10^{17} \alpha \cdot \text{cm}^{-2}$ , was heated above 600 °C, and then received a further  $\alpha$ -dose of  $c. 10^{17} \alpha \cdot \text{cm}^{-2}$  after cooling below 500 °C. Assuming uraninite to be the radioactive source in both cases, the separate  $\alpha$ -particle doses could each have occurred in about 10 Myr (see Vance and Milledge, 1972). These times would, of course, be longer if the radioactive source was less rich in EU than uraninite.

A brief description is now given of the diamonds showing green or brown surface coloration (or both) from various alluvial sources. However, as the localities of the diamonds within both Angola and Ivory Coast are unknown, no conclusions are drawn about the possible origin of the intense green spots noted on some of these diamonds.

The earliest description of diamonds with intense green and brown surface spots was of those found during the recovery of gold from the Rand Banket conglomerate in South Africa (Denny, 1897; Young, 1917). These diamonds have recently been further described by Raal (1969).

The Banket conglomerate contains between 0.025–0.1 wt % of uraninite (Gittus 1963), and the conglomerate itself is considered to be of late Pre-Cambrian Age (Sharpe 1949). There is some doubt, however, as to the age of the uraninite in the conglomerate. Liebenberg (1957) considers it to be detrital and gives 2000 Myr for the age of the *source* of detrital uraninite, but Davidson (1957) suggests the uraninite represents an epigenetic mineralization of the conglomerate with age values ranging between 2000 to 620 Myr. Both Davidson (*loc. cit.*) and Bowie (1970*b*) mention, however,



some reworking of the uraninite in the conglomerate and give an age for this event as 110 Myr.

From field evidence (Bell, personal communication), various metamorphic events since the Banket's formation have not raised the temperature of this geosynclinal deposit to 600 °C. However, in the early days of gold production, many of the diamonds found in the Banket were recovered from the Klerksdorp area (Denny, 1897), and also from the Modderfontein 'B' Gold Mine (Johannesburg) (Young, 1917). In the Klerksdorp area (Denny, 1896), and also in many other areas of the conglomerate, the Banket is intruded by diabase dykes and sills of Karroo (Cretaceous or older age, Young, 1917). Hence, such intrusions could well have caused localized heating of the Banket sufficient to cause green spots on a diamond to turn brown. After the diabases cooled there would have been adequate time for the brown spotted diamonds to acquire further green surface coloration, assuming uraninite still to be associated with the diamond.

Much less information is available about similar diamonds from Brazil and Venezuela. The diamonds from both countries are principally mined from recent river alluvials (Leite, 1969; Fairbairn, 1971). If, however, some of these diamonds were at one stage associated with *ancient* alluvial deposits (for example, uraninite-bearing conglomerates like those of the Serra de Jacobina in Bahia State, Brazil (Bowie, 1970a)), then the green and brown spots on these diamonds may have originated in a similar way to that proposed for diamonds from the Rand Banket (see above).

One diamond from a parcel of twenty from the Phuket region of peninsular Thailand, and three diamonds from the Kenieba region of Western Mali were considered by Grantham (unpublished work, 1965, 1968) to exhibit annealed radiation damage (see table I). The Thailand diamonds were recovered from recent alluvial deposits during tin mining, and Stephens *et al.* (1966) concluded that the diamonds were derived from a local Cambrian conglomerate, but the mineral composition and thermal history of this deposit are unknown. Grantham (*op. cit.*) suggested that the Mali diamonds were Pre-Cambrian in age and may have been redistributed from the original kimberlite into a local sandstone, but again the detailed composition and thermal history of this rock are unknown.

Green to pale green spots or coats have been observed in alluvial diamonds from several sources (see table I).

The diamonds with green surface coloration from the alluvial deposits of the Bushimaie river in the Congo (Kinshasa), and from the Koidu district of Sierra Leone, are similar to the diamonds with green surface coloration found in the kimberlites of these two countries, indicating that the green coloration was derived from radioactive minerals in the kimberlite, and not from detrital radioactive grains in the alluvial deposits.

Ghanaian diamonds are mined from the alluvial deposits of the Birrim river, and are weathered from a series of metamorphic schists and grey-wackes of Pre-Cambrian age (Applin, 1972). The primary diamond source is unknown. The even green surface colour of some of these diamonds, however, suggests that it was derived from radioactive minerals in the original kimberlite, rather than from the secondary or tertiary source.

In the South West African and Namaqualand alluvial diamond sources transparent green-coated diamonds, rather than intense green-spotted diamonds, are found in Tertiary to Recent river terraces and raised beach deposits. It would appear, therefore, that these transparent coats were derived from the original kimberlite, subsequent weathering and transporting agents bringing these diamonds to the western seaboard of southern Africa, with little, if any,  $\alpha$ -irradiation taking place in these alluvial sources.

*Conclusions.* Diamonds with either pale transparent green coats or both pale and intense green surface spots were found in all the diamond productions examined in this study, and have been reported by other authors to exist in other diamond productions. The presence of green surface coatings on diamonds of various shapes and sizes, and on fractured and irregular-shaped diamonds, demonstrates that the green transparent coat or spot is a very late stage phenomenon, occurring after kimberlite breakthrough and solidification. This, of course, would be expected, since green-coated diamonds turn brown (on their surfaces) on heating above 600 °C. From the experiments described, it seems clear that any green surface feature on diamond that can be rendered brown on heating above about 600 °C (preferably in vacuum) is due to  $\alpha$ -particle damage. Assuming  $\alpha$ -particle damage to be responsible for the transparent and even green coats on diamonds found in kimberlite, then the doses seem fairly consistent with those expected from the radioelement content and ages of the kimberlite pipes.

It also seems very likely that dark green spots on diamonds from the Bantek conglomerate are due to the diamonds being in direct contact with uraninite (see also Vance and Milledge, 1972), but that for diamonds in several other alluvial sources, the green coloration was derived from the kimberlite and not from radioactive minerals in the alluvial deposit. The coexistence of intense green and intense brown spots on the same diamond has also been discussed in terms of thermal events occurring after codeposition of diamond and radioelement-rich minerals in an alluvial source.

Since the radioelements appear to be generally distributed throughout a kimberlite source,  $\alpha$ -particle damage should be present in the other minerals associated with diamonds (e.g. olivines, garnets, diopsides, ilmenites, and zircons), although the body colour of these minerals is likely to obscure  $\alpha$ -damage colour effects unless they are severe. Experiments in this work at doses of  $1 \times 10^{14} \alpha \cdot \text{cm}^{-2}$  showed no detectable colour changes in diopside, pale-green olivine, and a near colourless zircon, though experiments at higher doses ( $2 \times 10^{17} \alpha \cdot \text{cm}^{-2}$ ) showed some surface darkening in a pale-green olivine and a red-purple garnet (Vance and Milledge, 1972).

However, *fission fragments* from the spontaneous fission of  $^{238}\text{U}$  also have ranges of approximately 20  $\mu$  in most materials (Chadderton and Torrens, 1968). Though doses of fission fragments might not cause colour changes in a material, the individual fission tracks can be detected in many materials by etching (see, for example, Fleischer and Hart, 1970).

It has been shown that the minimum  $\alpha$ -dose observable by colour changes in diamond is *c.*  $10^{14} \alpha \cdot \text{cm}^{-2}$ . Assuming uranium to be the principal radioelement in the immediate environment of a diamond, then an  $\alpha$ -dose of  $10^n \text{ cm}^{-2}$  corresponds to a fission fragment dose of *c.*  $10^{n-7} \text{ cm}^{-2}$ . Since (etched) fission track densities can be

readily measured down to 10 to 100 cm<sup>-2</sup> (corresponding to an  $\alpha$ -dose of c. 10<sup>8</sup> to 10<sup>9</sup> cm<sup>-2</sup>), fission track studies on diamond surfaces offer a very sensitive means of measuring small-scale uranium distributions in kimberlite. Experiments are in progress to try to 'develop' fission tracks by etching cleavage surfaces of diamonds which have been irradiated with fission fragments from a <sup>252</sup>Cf source.

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