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# Spectroscopic research on ultrahigh pressure (UHP) macrodiamond at Copeton and Bingara NSW, Eastern Australia

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#### ABSTRACT

Millions of macrodiamonds were mined from Cenozoic placers across Eastern Australia, 98% from within the Copeton and Bingara area (85 km across) in the Phanerozoic New England region of New South Wales (NSW). Raman spectroscopy of inclusions in uncut diamond, from the Copeton and Bingara parcels, identifies them as ultrahigh pressure (UHP) macrodiamond formed during termination of subduction by continental collision. Infrared spectral properties of the two parcels are critically similar in terms of nitrogen abundance (low in zoned diamond, high in unzoned diamond), requiring a pair of different growth mechanisms/protoliths. Within each parcel, the degrees of nitrogen aggregation are relatively strong and coherent, but they are so different from each other (moderate aggregation for Bingara, strong for Copeton) that the two parcels require separate primary and local sources. The local sources are post-tectonic alkali basaltic intrusions which captured UHP minerals (garnet, pyroxene, diamond) from eclogite-dominated UHP terranes (density stranded at depth-mantle, lower crust). X-ray diffraction studies on Copeton diamond indicate a normal density, despite previous reports of anomalously high density. For nonfluorescent diamond, a 2nd order Raman peak, which is prominent in theoretical perfect diamond and in African cratonic diamond, is suppressed in Copeton and Bingara UHP macrodiamond. Pervasive deformation during macrodiamond growth probably causes this suppression, the strong nitrogen aggregation, and the exceptional durability documented through industrial use.

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#### 1. Introduction

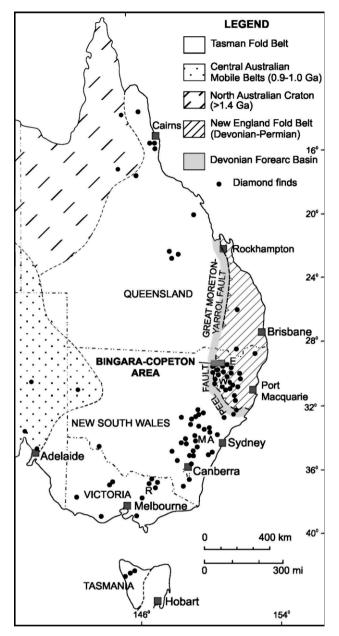
About a million carats of macrodiamonds (average 4 stones per carat) were mined from placers and paleoplacers within a  $400 \times 3000 \, \mathrm{km}$  belt in Eastern Australia [1], see Fig. 1. Despite this broad distribution, over 98% of production was confined to just 0.6% of this area: within the adjacent districts of Copeton and Bingara in the New England region of New South Wales (NSW). In the mined paleoplacers, typical diamond-related minerals are virtually absent, hampering efforts to trace the diamond back to hardrock sources. The eastern margin of Australia experienced recurrent episodes of prolonged westerly subduction, and termination of subduction due to collision with microcontinents, causing accretion and growth of Eastern Australia throughout the period late Neoproterozoic to mid Triassic [2], with most prominent episodes being of Phanerozoic age.

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Previous research on these macrodiamonds has demonstrated its unique subduction-related non-cratonic features [3–16] and two source models have been proposed: (A) diamonds were pre-existing in volumes of metastably cool lithosphere (ancient or Phanerozoic) emplaced under Australia and (B) diamonds formed during subduction, either ancient or Phanerozoic. Raman spectral determinations, of inclusion internal remnant pressures ( $P_r$ ) in uncut Copeton diamond, set up an PT array of inclusion-entrapment loci. The intersection of these restricts the diamond formation conditions to  $47 \pm 4$  kbar and 250-840 °C, identifying them as ultrahigh pressure (UHP) macrodiamond [9]. These results are consistent with an approximate thermodynamic PT locus derived from compositions of garnet and clinopyroxene inclusions [12].

Our preferred tectonic model for this UHP macrodiamond has it formed during various continental collision events which terminated repeated episodes of Phanerozoic "young" subduction. The diamond is characterized by strong pervasive deformation during growth, which has strongly accelerated nitrogen aggregation [12,14], thereby mimicking prolonged storage. Compared with cratonic diamond, anomalously high  $P_{\rm r}$  values on olivine/garnet/pyroxene inclusions are characteristic of Copeton

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**Fig. 1.** Palaeoalluvial and alluvial diamond occurrences, and relevant geological boundaries within Eastern Australia. Copeton is east of Bingara, whereas Walcha (W) is about 100 km south of Copeton.

macrodiamond [9], and inherently constrain diamond formation to low temperature UHP conditions [9]. These higher  $P_{\rm r}$  values exclude the possibility of plastic release of internal pressures on inclusions in diamond during delivery to the earth's surface, unlike the continuation of plastic behaviour in host garnet during exhumation of the exposed UHP terrane at Kokchetav [17]. Our diamond surface features indicate magmatic delivery to the earth's surface [9,12–14], most likely by post-tectonic alkali basaltic intrusions which carry relatively abundant UHP minerals (garnet, pyroxene, for analyses see [18]), captured from high density UHP terranes stranded at depth (upper mantle, lower crust) [18]. Despite this UHP link, there are no exposed UHP terranes in NSW and microdiamonds are absent.

Copeton/Bingara macrodiamonds are characterized by rounded surfaces (see Fig. 2) with more than 90% of shiny stones having abundant to overlapping microdiscs. The most common inclusion is coesite (Fig. 2d), having an equant to tapering prismatic to platy

(Fig. 2d) shape, the latter two with imposed six-sided symmetry. Important physical structures include euhedral indentations (roughly 5% of stones), pitted depressions (15%, see Fig. 2d) and naats (30%, a narrow belt with multiple twins).

Our overall research on more than 250 uncut Eastern Australian macrodiamonds showed that 97% of inclusions tested failed to give a Raman response, so the Raman inclusion technique is not an efficient way for testing random uncut macrodiamond as noncratonic in origin. Menneken [19] compiled published details about Raman spectra of microdiamond from well-defined non-cratonic sources (exhumed UHP terrane; Hawaiian mantle-circulation; back arc basin in Japan), and proposed a test for identifying non-cratonic diamond: a 2nd order Raman peak prominent in cratonic diamond is absent from non-cratonic diamond. First, the Menneken test is trialed and calibrated on African macrodiamond, then it is applied to alluvial macrodiamond from Copeton, Bingara and Walcha (all in New England NSW), Argyle (West Australia) and Cempaka (Kalimantan, Indonesia).

We make the first detailed comparison of nitrogen abundances and aggregation values in diamond from Copeton [10] and Bingara [12,14,16], and use this to demonstrate that there must be multiple local intrusions capturing diamonds from different formation rocks (at depth). Some Copeton diamond was X-rayed to obtain the theoretical density, to compare with previous reports of NSW diamond having anomalously high specific gravity [1].

## 2. Experimental

Published nitrogen abundance and aggregation results for diamond from Copeton and Bingara were derived from infrared spectroscopy using established procedures [10,12,14,16].

The Raman equipment comprises a Dilor SuperLabram spectrometer, with a holographic notch filter (600 and  $1800\,\mathrm{g/mm}$  gratings), liquid nitrogen-cooled 2000 pixel CCD detector, and a 514.5 nm Melles Griot 543 argon ion laser (5 mW at the sample). The spectral resolution was set at  $2\,\mathrm{cm^{-1}}$  (slit width of  $100\,\mu\mathrm{m}$ ). The microscope uses a  $50\times$  ULWD Olympus microscope objective, focusing the laser spot to  $2\,\mu\mathrm{m}$  in diameter and  $5\,\mu\mathrm{m}$  deep.

A PanAlytical X'Pert Pro X-ray Diffractometer with a  $\theta$ - $\theta$  geoniometer was used to X-ray powdered diamond, operating at 40 mA, 45 kV of Cu K $\alpha$  radiation, with gas-filled proportional counter, graphite monochromator, 1° divergence slit, 2° antiscatter slit and 0.1 mm receiver slit.

### 3. Theory

### 3.1. Nitrogen abundances and aggregation states

Nitrogen is the most abundant element that substitutes for carbon atoms in the diamond structure, with values up to 3300 ppm reported for macrodiamond [20,21]. Reviews [6,22] of nitrogen aggregation in diamond show N starts off as singly substituted for carbon on random lattice sites, then these rapidly aggregate to place N atoms on two adjacent sites (called IaA). By a slow process, IaA aggregates to place N atoms on four adjacent sites (called IaB). Higher N abundances, longer storage times, higher storage temperature, and strong plastic deformation during/after growth all increase the aggregation state, see Fig. 3. The dashed lines (Fig. 3) are isomaturity trends for nitrogen aggregation, calculated as a combination of storage time and temperature (see [23] for equations and coefficients, noting that the results are in "time equivalents" etc. since deformation is ignored in the calculation). A significant to major fraction of singly substituted N has been reported in microdiamond from exhumed UHP terranes [24], but also in some kimberlitic diamond from cratonic terranes [6], indicating a short period of confinement (1-20 MY) between for-

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