

The origin of sapphires: U–Pb dating of zircon inclusions sheds new light

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Abstract

Uranium–lead isotope dating of two zircon inclusions in sapphires from the Central Province, NSW, gives ages of 35.9 ± 1.9 and 33.7 ± 2.1 million years (Ma). These ages fall within the range of basalt potassium–argon ages of 19 to 38 Ma and zircon fission track ages of 2 to 49 Ma for the timing of volcanism of the Central Province, NSW. These data, combined with the observation that corundum is found associated with many alkali basaltic provinces, indicate a genetic link between the growth of large corundum crystals and the processes involved in alkali basaltic magma generation. The reported failure of experimental attempts to grow corundum from a corundum-bearing basaltic composition, and more significantly, the abundance of incompatible elements such as U, Th, Zr, Nb and Ta in inclusion minerals indicate that the crystallization process is not simple. Corundum and the other minerals found as its inclusions (zircon, columbite, thorite, uranium pyrochlore, alkali feldspar etc.) could not have crystallized from most basaltic compositions. A more complex process must occur in which crystallization takes place when there are high proportions of incompatible elements and volatiles in the melt. These crystallization products are then carried to the surface by upward movement of later magmas. The extent of this process presumably determines whether a particular basaltic province carries sufficient corundum to be worked into economic concentrations of sapphire.

KEYWORDS: sapphire, uranium–lead dating, inclusions, zircon, Central Province, Australia.

Introduction

A LARGE number of important sapphire and ruby gemfields are associated with largely alkali volcanic terrains. The gem-quality corundum is commonly recovered, along with other heavy minerals such as zircon, spinel and ilmenite, from concentrations in the alluvial systems draining these terrains.

Gemfields associated with basaltic volcanism include New England and Anakie in Australia (MacNevin, 1972; Broughton, 1979); Pailin in Cambodia (Jobbins and Berange, 1981); Chanthaburi-Trat, Denchai, Bo Ploi and Khorat Plateau in Thailand (Barr and MacDonald, 1981;

Keller, 1982; Gunawardene and Chawla, 1984); Bokeo Plateau, Xuan Loc Plateau, Cardomones Massif, Solovens Plateau and Kassens Plateau in Kampuchea (Lacombe, 1969–70); Haut Chalong Plateau, Pleiku Plateau, Darlac Plateau and Djiring Plateau in South Vietnam; Kouang Tcheoci Wan and Hainan Island in Southern China; Mercaderes Rio Mayo area in Colombia (Keller *et al.*, 1985); Gimi Valley in Nigeria (Irving and Price, 1981).

A number of striking similarities between the gemfields of this group can be seen.

(a) They are associated with extrusive basaltic lava fields or eroded remnants (also laterites).

TABLE 1: Comparative ages from the Central Province, New England, N.S.W.

Age (Ma)	Description	Map Sheet	Grid Ref.	Method	Reference	
22.6 ± 2.0*	Copeton Rd, Inverell (flow)	Inverell	1:250,000	404304	K-Ar	Wellman & McDougall, 1974
19.0 ± 1.0*	11 km W of Inverell (flow)	Inverell	1:250,000	393308	K-Ar	McDougall & Wilkinson, 1967
21.2 ± 1.0*	Dangersleigh Rd, Armidale (flow)	Dorrigo	1:250,000	471215	K-Ar	McDougall & Wilkinson, 1967
34.3 ± 1.5*	42.5 km N of Guyra	Grafton	1:250,000	478299	K-Ar	McDougall & Wilkinson, 1967
32.2 ± 1.2*	1.5 km S of Armidale Airport	Dorrigo	1:250,000	463219	K-Ar	Wellman, 1971
33.8 ± 0.7*	11.4 km W of Glen Innes (flow)	Grafton	1:250,000	465319	K-Ar	Wellman, 1971
35 - 36*	Spring Mountain (flow)	Grafton	1:250,000	461303	K-Ar	Cooper <i>et al.</i> , 1963
20.6 ± 0.2	Newstead Rd (flow)	Elsmore	1:25,000	389001	K-Ar	Smith, 1988
31.9 ± 0.3	Newstead South (flow)	Elsmore	1:25,000	422004	K-Ar	Smith, 1988
32.3 ± 0.2	Tingha-Guyra Rd (flow)	Indianna	1:25,000	402889	K-Ar	Smith, 1988
23.2 ± 0.5	Braemar quarry (upper flow)	Inverell	1:100,000	349033	K-Ar	C.D. Ollier pers. comm., 1989
37.0 ± 1.5 ¹	Braemar quarry (alluvials)	Inverell	1:100,000	350034	Fission Track	Hollis & Sutherland, 1985
35.9 ± 1.6 ¹	Horse Gully (alluvials)	Inverell	1:100,000	415135	Fission Track	Hollis & Sutherland, 1985
34.7 ± 1.7 ¹	Kings Plains (alluvials)	Inverell	1:100,000	492155	Fission Track	Sutherland & Hollis unpubl. data
37.1 ± 1.9	Yolanda (diatreme)	Glen Innes	1:100,000	700825	Fission Track	Sutherland & Hollis unpubl. data
35.9 ± 1.9	Yarrow River (30/26)	Glen Innes	1:100,000	908887	U-Pb	this paper
33.7 ± 2.1	Dunvegan Lagoon (28/11)	Glen Innes	1:100,000	728212	U-Pb	this paper.

* Recalculated age using new IUGS constants (after Dalrymple, 1979). ¹ pooled age from a composite range of ages.

(b) In most cases, crater lakes, cones, cone remnants or plugs can be found.

(c) The basalt type is predominantly alkaline (with the exception of Southern China and Nigeria).

(d) Ultramafic xenoliths (predominantly mantle lherzolites) are often reported.

(e) In a number of the fields, older basalt flows are tholeiitic with later flows becoming increasingly alkaline.

(f) The corundum and associated megacrysts are corroded, suggesting disequilibrium with the carrier magma.

(g) *In-situ* corundum megacrysts are rarely seen within the basalts, apart from rare mafic dykes (such as Loch Roag; Jackson, 1984).

(h) Different areas within a gem-producing terrain may yield different coloured stones changing in spatial distances under 10 kilometres suggesting multiple sources.

(i) Crystal inclusions found within the sapphires include; apatite, columbite, rutile, pyrrhotite, boehmite, zircon, hercynite (Fe spinel), gahnospinel (Fe, Zn spinel), almandine, pyrope, hatchetelite (U, Ti pyrochlore), alkali-feldspar and plagioclase.

(j) Sapphires are almost always associated with zircon, spinel and ilmenite in placer deposits. Sometimes they may also be associated with olivine, clinopyroxene, garnet, magnetite or feldspar.

The aim of this paper is to explain the origin of corundum associated with such volcanic provinces. Is the corundum picked up at a higher level unrelated to the magmas such as a peraluminous layer in the crust, or is it formed in some

process associated with the carrier magmas themselves?

The morphology of the corundum crystals and the nature of their inclusions should help answer these questions. However the key will be the age relationship between the corundum and associated volcanic rocks.

The study area

As part of a project to determine the origin of sapphires, some 255 inclusion-bearing corundums were collected from streams and rivers draining the Central Province, NSW (The New England Gemfield).

The Central Province (Fig. 1) extends from Armidale in the south, through Glen Innes and Inverell, to the Queensland border in the north. It is part of a discontinuous band of Mesozoic-Cainozoic volcanic rocks (Wellman and McDougall, 1974; Sutherland, 1985) which extends for a north-south distance of 3500 kilometres and is about 300 kilometres wide within or adjacent to the Eastern Highlands.

The basaltic rocks of the Central Province range in age from 19 to 38 Ma based on K-Ar dating (Cooper *et al.*, 1963; McDougall and Wilkinson, 1967; Wellman and McDougall, 1974; Sutherland and Raynor, unpubl. data; C. D. Ollier, pers. comm. 1989) and from 2 to 49 Ma based on reset zircon fission track ages (Hollis and Sutherland, 1985; Sutherland and Hollis, unpubl. data). The ages available for the Central Province are shown on Fig. 1. The method used and the source of the data are listed in Table 1. The lavas are predominantly alkaline to strongly alkaline, alkali olivine basalts, basanites, hawai-

ites and nepheline hawaiites (Wilkinson, 1962, 1966, 1973; Binns, 1969; Binns *et al.*, 1970; Duggan, 1972; Wilkinson and Duggan, 1973; Street, 1974; McKay, 1975; McQueen, 1975; Barron, 1987). Theoleiites are recorded in the vicinity of Inverell by Duggan (1972). Volcaniclastic rocks have been reported by various workers (Lishmund and Oakes, 1983; Sutherland, 1985; Temby, 1986; and Barron, 1987) and mapped as occurring extensively throughout the Inverell-Glen Innes region. They are situated at or near the base of the volcanic pile (Brown and Pecover, 1986 *a, b*; Pecover and Brown, 1986; Brown, 1987; Pecover, 1987; Pecover and Coenraads, 1989). The structural control and timing of Central Province volcanism has been described by Coenraads (1988) and the province may be divided into western and eastern parts on this basis. Sapphires are found in variable quantities in almost every gully, creek and river draining the East Central Province and some of the alluvial deposits are currently being mined by large scale, mechanized operations.

The sapphires

The large-scale mining operations of T. J. & P. V. Nunan Pty. Ltd. allow the viewing of a very large number of sapphires as well as low value corundum making up the daily mine run. The stones are predominantly well formed crystals or fragments showing crystal faces. Pointed crystal-terminations, known locally as 'dogs teeth' and flat hexagonal prisms known as 'flats' are common, indicating that crystals are often broken along the basal {0001} parting plane. Large whole crystals are rare but sections bounded by crystal faces and parting planes with diameters of 3 to 4 cm indicate that some crystals grew to at least 12 cm in length.

Sharp crystal edges are not seen, crystal edges are slightly rounded and crystal faces are often smooth and glossy due to chemical corrosion (resorption due to disequilibrium with the carrier magma) whilst in transit to the surface. Zoning is common, both parallel to the prism and pyramid faces forming concentric colour bands, and parallel to the basal plane. Zoned crystals with greenish or yellowish cores or 'pipes' and blue rims may be seen. Red and pink pipes have been reported (T. Coldham, pers. comm., 1989).

Sapphires are rarely seen intergrown with other minerals. However certain evidence suggests that they were in this state during their formational history.

1) An anorthoclasite xenolith containing corundum has been reported from Ruddons Point,

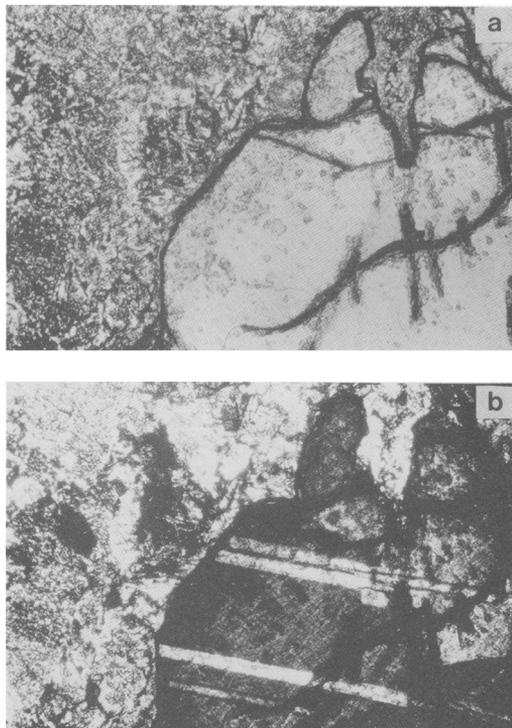


FIG. 2. Sapphire-anorthoclase xenolith from a basalt, Mt Leura, Hoy Province, Queensland. Specimen D44379 is held at the Australian Museum. The corundum is the clear, high-relief grain in the upper left of photomicrograph (a). The anorthoclase, occupying the rest of the field of view is cloudy due to incipient alteration. Under crossed polars, photomicrograph (b), the corundum shows first order grey birefringence and twinning lamellae. Examples such as these together with external crystal morphological evidence suggest that sapphires may have grown as part of a coarse mineral assemblage. The base of each photo is 3.75 mm. Photographs by R. E. Pogson.

U.K. by Upton *et al.*, 1983. In Australia, a sample of sapphire with anorthoclase from the Hoy Province is known and a piece is held by the Australian Museum (Fig. 2).

2) Sapphire crystals from the Central Province occasionally show negative crystal shapes in their faces as shown in Fig. 3. These suggest growth against other crystals, if only other sapphires.

3) Some sapphires from the Central Province show evidence of markedly uneven chemical resorption indicating that they have been partially protected at some stage, possibly by being partially included in another mineral or aggregate of minerals. Such an example is shown in Fig.

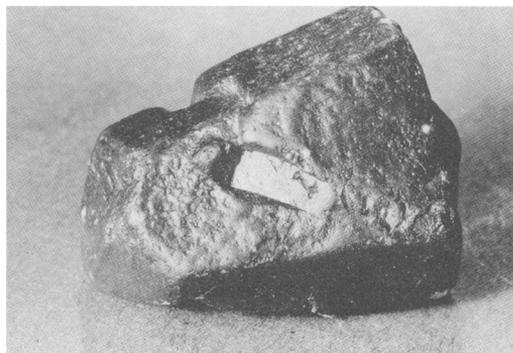


FIG. 3. Sapphire crystal from the Central Province, NSW, showing a negative crystal shape in its faces. The sapphire is 20 mm in length. Photograph by D. Barnes.

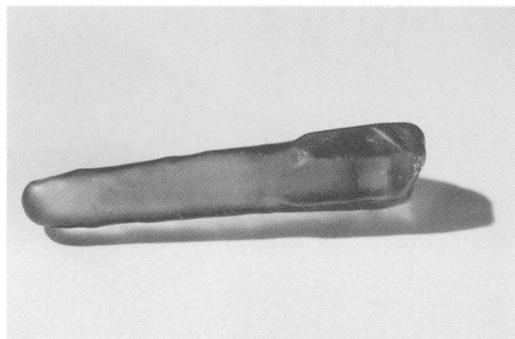


FIG. 4. Sapphire crystal from the Central province, NSW, showing uneven chemical resorption. One end of the 20 mm crystal shows markedly greater chemical resorption than the other. Fig. 5 shows a possible scenario to explain its appearance.

4, and Fig 5 shows a possible scenario explaining its morphology.

Inclusions in sapphires

In order to shed light on the origin of sapphires the inclusion minerals must be syngenetic, growing with and under the same conditions as the host sapphire. Gubelin and Koivula (1986) define certain criteria for an inclusion to be syngenetic with the host. It must be euhedral and unbroken, and show no evidence of surface etching, corrosion or oxidation which would indicate an earlier period of crystallization or derivation from the wall rocks. It must be independent from any cracks, fractures or epigenetic (later) mineraliza-

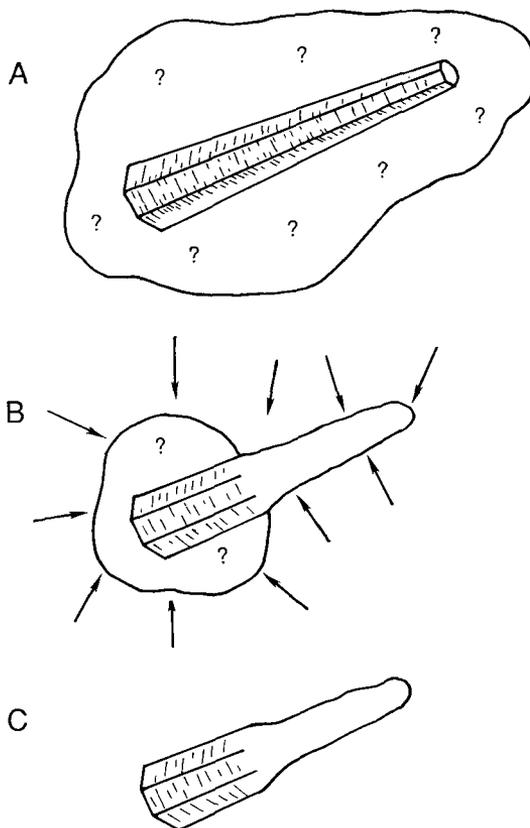


FIG. 5. Scenario explaining uneven chemical resorption such as seen on the sapphire crystal shown in Fig. 4. (a) The sapphire crystal is originally intergrown with another mineral or minerals. (b) The sapphire crystal is partially exposed. This portion of the crystal is subject to chemical corrosion due to disequilibrium with the carrier magma during its ascent to the surface. Note that alluvial abrasion cannot be evoked to explain the same scenario due to the fragility of the specimen. (c) The sapphire crystal is liberated from its host toward the end of its journey.

tion within the host. Syngenetic inclusions such as zircon, uranium pyrochlore, thorite, columbite and alkali feldspar (Gubelin and Koivula, 1986; Coenraads, in prep.) as well as abundant carbon dioxide filled fluid inclusions (R. Wilkins, pers. comm. 1989) indicate that the sapphires must have grown in an environment rich in incompatible elements (U, Th, Nb, Ta, Zr), alkalis (Na, K) and volatiles.

U-Pb dating of the zircon inclusions

Four small, transparent, euhedral zircon inclusions were identified during 1988 using the

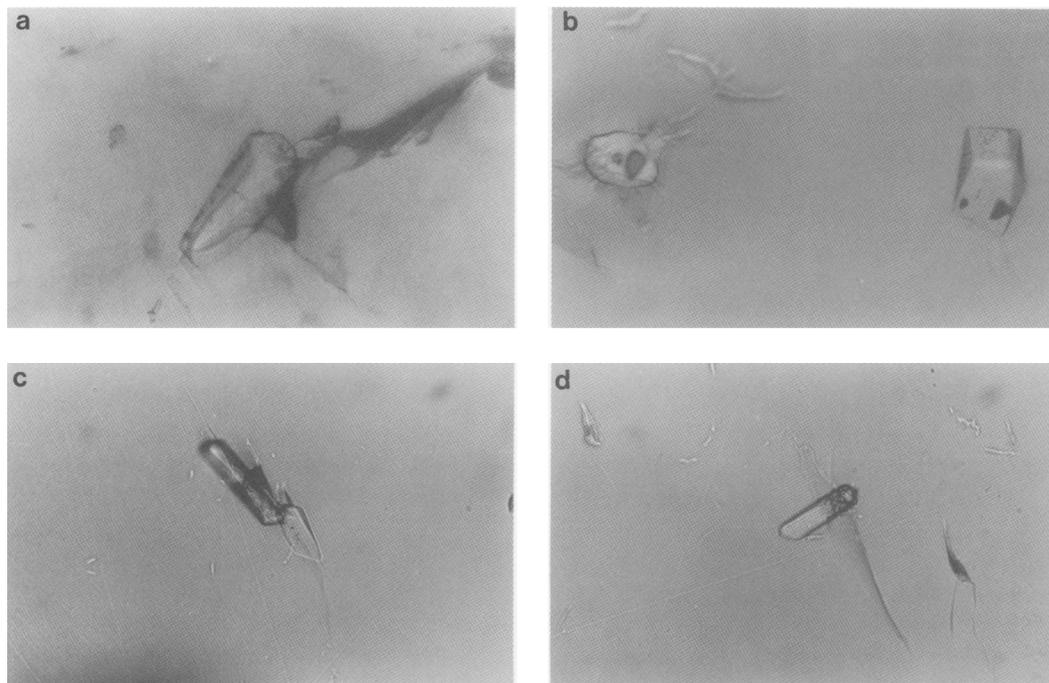


FIG. 6. Euhedral zircon crystals included in sapphire, transmitted light photomicrographs. The base of each photo is 0.22 mm. (a) Inclusion 30/26 panned from the Yarrow River, GR:908887, Glen Innes 1:100 000 sheet. (b) Inclusion 28/11 from alluvium obtained during test drilling of Dunvegan Lagoon by T. J. & P. V. Nunan Pty Ltd, GR:728212, Glen Innes 1:100 000 sheet. The inclusion on the right is the dated zircon, the inclusion on the left is a thorium silicate containing 60% ThO₂. Note the dark halo around this inclusion presumably due to radiation damage of the host sapphire. (c) Inclusion 13/1 obtained by the mining operation of T. J. & P. V. Nunan Pty Ltd. Locality unknown, either Kings Plains Creek or Reddestone Creek. (d) Inclusion 13/2, another inclusion in the same host as 13/1.

electron microprobe at Macquarie University. They are shown in Fig. 6. Zircons contain trace amounts of U isotopes taken into the lattice at the time of crystallization. By measuring the amount of breakdown to daughter Pb isotopes and knowing the rates at which this occurs, the zircons can be dated. It is clear that such inclusions are extremely valuable as a means of providing the age of formation of the host sapphires. However, it is only recently that U–Pb dating of such samples has become possible with the development of the Sensitive High Mass Resolution Ion MicroProbe (SHRIMP) at the Australian National University, which permits *in situ* isotopic analysis on a microscopic scale.

Analytical methods

The SHRIMP technique for U–Th–Pb isotopic analysis of zircons was first described by Compston *et al.* (1984) with subsequent modifications

outlined by Kinny *et al.* (1988). The electron probe mounts in which the zircon inclusions in sapphire were identified, were modified for ion probe analysis by the addition of a hole to accommodate the laboratory standard zircon, then repolished and recoated with carbon. The two largest inclusions were chosen for analysis, both being of sufficient size to accommodate the 25 µm analytical spot without overlap onto the sapphire matrix (Fig. 7) which in any case was found to contain no detectable lead or uranium. Each analysis consisted of three counting cycles of the Pb, Th, and U species of interest, after which raw Pb/Pb and Pb/U isotopic ratios, and total U and Th contents were calculated. Both analyses were ‘duplicated’ by resumed excavation of the same sputter-holes.

The young (Cenozoic) ages of the zircon inclusions necessitated special treatment of the data. For young zircons, there is little value in using the ²⁰⁷Pb/²⁰⁶Pb ratio as a measure of age, because

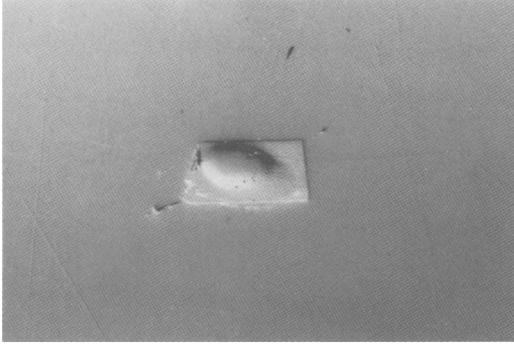


FIG. 7. Zircon inclusion 28/11, showing the sputter-hole excavated by the ion beam. The hole is approximately 25 μm across.

the majority of the measured ^{207}Pb is of non-radiogenic origin. For this reason, however, the ^{207}Pb provides the most effective means of monitoring the contribution of 'common' Pb. Assuming that the U–Pb isotopic systems are concordant and undisturbed, the measured $^{206}\text{Pb}/^{238}\text{U}$ ratio, normalised to that of the laboratory standard zircon SL3 ($^{206}\text{Pb}/^{238}\text{U} = 0.0928$), provides an initial estimate of the true age, from which the expected radiogenic $^{207}\text{Pb}/^{206}\text{Pb}$ can be calculated. Using this value together with a modelled common Pb composition (that of Broken Hill ore Pb in this instance, because the detectable common Pb was derived principally from surface contaminants rather than intrinsic non-radiogenic Pb), the proportion of non-radiogenic ^{206}Pb in the total ^{206}Pb can be accurately determined. This yields a revised estimate for the radiogenic $^{206}\text{Pb}/^{238}\text{U}$, and the corresponding age. The quoted uncertainties represent combined estimates of the errors associated with ion-counting and the uncertainty associated with normalization of the measured $^{206}\text{Pb}/^{238}\text{U}$ ratio to that of the standard zircon SL3, the latter being based upon the reproducibility of measurements of the standard zircon during the analytical session.

Results

Results are shown in Table 2. The duplicate analyses of both inclusions are each in agreement to within their assigned experimental uncertainties. Grain 30/26 gave a mean $^{206}\text{Pb}/^{238}\text{U}$ age of 35.9 ± 1.9 Ma, whereas grain 28/11 gave 33.7 ± 2.1 Ma. Since there is no significant difference between these two ages with respect to experimental error, they may be combined to yield a mean age estimate for both inclusions of 34.9 ± 1.4 Ma (2σ). Strictly speaking, this is only

a minimum estimate for the age of the inclusions, because of the possibility that at some stage of their history an unknown amount of the radiogenic Pb accumulated in the zircons may have been lost. However, we would argue against such a possibility for two reasons. Firstly, the young apparent age of the inclusions implies that up to the present time only minor damage would have been caused to the zircon structures by the *in situ* disintegration of U atoms (the principal cause of Pb loss from zircons), and secondly, being wholly enclosed within inert sapphire megacrysts, the zircon inclusions would have been protected throughout their history from interaction with permeating fluids.

Another consideration is that Pb might not have accumulated in the zircons at all prior to the time of eruption of the host basalt, owing to the high temperature of the source regions, in which case they could be much older than their indicated age. The effective 'blocking temperature' for Pb in zircon has never been established. However, Rudnick and Williams (1987) reported the preservation of Proterozoic zircon ages in lower crustal xenoliths incorporated into a Cenozoic basalt cinder cone in north Queensland, and Kinny *et al.* (1989) reported analyses of kimberlitic zircons from Botswana which were erupted in the Permian but which nevertheless preserved Archaean ages. Both examples suggest that if these zircons and their host sapphires were *in fact* ancient xenocrysts incorporated into the basaltic magmas, it is likely that the old U–Pb ages would be preserved.

On the basis of this evidence we conclude that the true age of both inclusions, and hence of the sapphires themselves, is 35 Ma.

Discussion

The 35 Ma age of the zircon inclusions in the sapphires lies within the range of K–Ar ages for the basalts of the East Central Province and within the range of zircon fission track ages for the area (Fig. 1, Table 1) thus suggesting a genetic link. We believe that the sapphires formed in some process associated with the basaltic magma generation or evolution. The surface morphology of the stones and their inclusion suite indicate however that this process is not simple. This is reinforced by the presence of reaction rims of spinel on some corundum crystals (Stephenson, 1976), by the reported failure of experimental attempts to grow corundum from a corundum-bearing basaltic composition under different pressure, temperature and hydrous conditions (Green *et al.*, 1978), and by the abundances of incompatible

TABLE 2: U–Pb ages of zircon inclusions in sapphire

Spot	$^{206}\text{Pb}/^{238}\text{U} \pm 2\sigma$	AGE $\pm 2\sigma$	U	Th	Pb*	Th/U	%com. ^{206}Pb
30/26.1	0.00562 \pm 42	36.1 \pm 2.7	984	850	6	0.864	10.4
30/26.2	0.00553 \pm 42	35.6 \pm 2.7	968	832	6	0.859	5.4
28/11.1	0.00543 \pm 48	34.9 \pm 3.1	457	178	2	0.388	21.8
28/11.2	0.00508 \pm 44	32.7 \pm 2.9	449	171	2	0.380	12.3

$^{206}\text{Pb}/^{238}\text{U}$ refers to radiogenic ^{206}Pb . Pb* refers to the total radiogenic Pb. Values for U, Th and Pb* are in parts per million. %com. ^{206}Pb is the percentage of non-radiogenic ^{206}Pb in the total.

elements such as U, Th, Zr, Nb and Ta in inclusion minerals (Gubelin and Koivula, 1986; Coenraads, in prep.).

Crystallization may be taking place as partial melting of the mantle occurs and such a process might involve generation of more evolved magmas at depth, crystallization of mineral assemblages including the sapphires and their inclusion minerals from these magmas rich in volatiles and incompatible elements, and finally the disintegration and partial resorption of the rock types during transport to the surface in later magmas. Such minerals may crystallize from liquids whose composition represents very tiny amounts of upper-mantle partial melting. As such, these crystallization products could then be expected to be out of equilibrium with basalts formed from larger proportions of melting.

Irving and Price (1981) describe a fractionation model which allows the generation of evolved magmas (phonolites) at upper-mantle depth, via the fractional crystallization of kaersutitic amphibole, olivine, iron–titanium spinel, aluminous clinopyroxene, mica and apatite. Their model is supported by the observation of lherzolite-bearing phonolitic lavas from Bokkos (Nigeria), Phonolite Hill (Australia) and Heldburg (East Germany). The crystallization products of such magmas may accumulate in pockets or fissures, or as plating on conduit walls (Irving, 1980), until such time as partial melting has generated sufficient magma to carry them to the surface.

Upton *et al.* (1983) believe that the megacrysts (anorthoclase, sanidine, clinopyroxene, kaersutite, Ti-biotite, Ti-magnetite, Mg-ilmenite, apatite, zircon and corundum) found in upper Palaeozoic alkali basalts of the British Isles are the result of the disintegration of such rock types. Evidence in the form of megacryst surface morphology and the occasional observation of corundum-bearing xenoliths discussed earlier support such a suggestion.

The growth zoning, CO₂-rich fluid inclusions and high inclusion homogenization temperatures (>685°C) observed in Queensland sapphires led

Stephenson (1976) and Irving (1986) to assign a high temperature and pressure magmatic origin to their formation. They were presumed to result from fractionation of basanitic magmas at least within the deep crust.

Sapphire-bearing rocks in the Central Province volcanic sequence

The conclusion drawn by Thompson *et al.* (1986) for the British Tertiary Volcanic Province and the observations of Pecover (1987) suggest that the following sequence of events may have occurred in the Central Province, NSW: Magma movements 'ream out' conduits containing crystallization products of the highly evolved magmas or products of metasomatizing fluids, carry the evolved megacryst assemblage upwards along fracture sets and erupt explosively onto the surface. Thompson *et al.* (1986) suggest that once the initial magmas ream out the magmatic plumbing system then the remaining batches of liquid would be able to rise without significant interruption.

The implications of this model are therefore that the volcanoclastics, or products of explosive volcanism, occurring towards the base of the volcanic pile are likely to contain the most abundant sapphires.

Lacombe (1969–70) noted that in the Bokeo Plateau region of Kampuchea, zircons, corundum, garnet, titanomagnetite, spinel and anorthoclase megacrysts, although present in the majority of basalts, are most abundant in the products of explosive eruptions and their weathering products. Similarly, Barr and MacDonald (1981) note that in the Chanthaburi-Trat area of Thailand, megacrysts of aluminous clinopyroxene, ilmenite, garnet, zircon and spinel are abundant in the pyroclastic debris in the vicinity of vents.

Variation in the fine details of such a process, such as the time elapsed between initial partial melting and megacryst formation, to their transportation to the surface, may determine the

amount of sapphires in a particular source rock and possibly their size and quality. Hence, combined with the necessary geomorphological conditions for the development of a sapphire deposit, the above-mentioned process of megacryst formation may determine whether or not a particular volcanic province has economic potential.

Conclusions

An ion microprobe age of 34.9 ± 1.4 Ma for syngenetic zircon inclusions in New England sapphires shows that the minerals originated during the Cainozoic volcanism of this area. The corundum probably formed from strongly evolved magmas held in the deep crust or mantle and was carried up in profuse abundance during the volatile-rich, explosive eruptions.

Acknowledgements

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