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Regional patterns in the paragenesis and age of inclusions in diamond, diamond composition, and the lithospheric seismic structure of Southern Africa

Steven B. Shirey[a,*], Jeffrey W. Harris[b], Stephen H. Richardson[c], Matthew Fouch[d], David E. James[a], Pierre Cartigny[e], Peter Deines[f], Fanus Viljoen[g]

[a] *Department of Terrestrial Magnetism, Carnegie Institution of Washington, 5241 Broad Branch Road, NW, Washington, DC 20015, USA*
[b] *Division of Earth Sciences, University of Glasgow, Glasgow G12 8QQ, UK*
[c] *Department of Geological Sciences, University of Cape Town, Rondebosch 7701, South Africa*
[d] *Department of Geological Sciences, Arizona State University, Box 871404, Tempe, AZ 85287-1404, USA*
[e] *Laboratoire de Geochemie des Isotopes Stables, Universite de Paris VII, Institut de Physique du Globe de Paris (IPGP), 4 Place Jussieu, 75251 Paris Cedex 05, France*
[f] *Department of Mineral Sciences, The Pennsylvania State University, University Park, PA, USA*
[g] *De Beers GeoScience Centre, PO Box 82232, Southdale, 2135, South Africa*

Abstract

The Archean lithospheric mantle beneath the Kaapvaal–Zimbabwe craton of Southern Africa shows $\pm 1\%$ variations in seismic P-wave velocity at depths within the diamond stability field (150–250 km) that correlate regionally with differences in the composition of diamonds and their syngenetic inclusions. Seismically slower mantle trends from the mantle below Swaziland to that below southeastern Botswana, roughly following the surface outcrop pattern of the Bushveld-Molopo Farms Complex. Seismically slower mantle also is evident under the southwestern side of the Zimbabwe craton below crust metamorphosed around 2 Ga. Individual eclogitic sulfide inclusions in diamonds from the Kimberley area kimberlites, Koffiefontein, Orapa, and Jwaneng have Re–Os isotopic ages that range from circa 2.9 Ga to the Proterozoic and show little correspondence with these lithospheric variations. However, silicate inclusions in diamonds and their host diamond compositions for the above kimberlites, Finsch, Jagersfontein, Roberts Victor, Premier, Venetia, and Letlhakane do show some regional relationship to the seismic velocity of the lithosphere. Mantle lithosphere with slower P-wave velocity correlates with a greater proportion of eclogitic versus peridotitic silicate inclusions in diamond, a greater incidence of younger Sm–Nd ages of silicate inclusions, a greater proportion of diamonds with lighter C isotopic composition, and a lower percentage of low-N diamonds whereas the converse is true for diamonds from higher velocity mantle. The oldest formation ages of diamonds indicate that the mantle keels which became continental nuclei were created by middle Archean (3.2–3.3 Ga) mantle depletion events with high degrees of melting and early harzburgite formation. The predominance of sulfide inclusions that are eclogitic in the 2.9 Ga age population links late Archean (2.9 Ga) subduction-accretion events involving an oceanic lithosphere component to craton stabilization. These events resulted in a widely distributed younger Archean generation of eclogitic diamonds in the lithospheric mantle. Subsequent Proterozoic tectonic and magmatic events

* Corresponding author. Fax: +1-202-478-8821.
E-mail addresses: shirey@dtm.ciw.edu (S.B. Shirey), jwh@earthsci.gla.ac.uk (J.W. Harris), shr@geology.uct.ac.za (S.H. Richardson), fouch@asu.edu (M. Fouch), cartigny@ipgp.jussieu.fr (P. Cartigny), p7d@psu.edu (P. Deines).

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altered the composition of the continental lithosphere and added new lherzolitic and eclogitic diamonds to the already extensive Archean diamond suite.

Keywords: Diamond; Eclogite; Peridotite; Inclusion; P-wave; Craton; Lithosphere

1. Introduction

A worldwide association between ancient lithospheric mantle keels beneath cratons and diamond occurrences has long been known (Boyd and Gurney, 1986; Janse, 1992). Seismic imaging of the lithospheric mantle beneath the Kaapvaal and Zimbabwe cratons and the Limpopo mobile belt known as the Southern Africa Seismic Experiment (SASE, James et al., 2001; James and Fouch, 2002) carried out during the multidisciplinary, multinational Kaapvaal Lithosphere Project (Carlson et al., 1996, 2000) has produced the first detailed picture of the lithospheric mantle at depths within the diamond stability field. It reveals prominent variations in the present seismic velocity structure of this mantle keel. Within the south African cratonic keel, Archean mantle peridotite and eclogite host multiple generations of diamonds that are both Archean and Proterozoic (Kramers, 1979; Richardson et al., 1984, 1993, 2001; Navon, 1999; Pearson and Shirey, 1999; Shirey et al., 2002) as well as less common occurrences of younger diamonds (Boyd et al., 1987; Akagi and Masuda, 1988; Navon et al., 1988; Schrauder and Navon, 1994; Pearson et al., 1998; Izraeli et al., 2001). Xenoliths of peridotite and eclogite and xenocrysts of diamond have been brought to the surface from depths as great as 150–180 km in kimberlites whose ages are typically young (65–150 Ma) but can be significantly older (240–1600 Ma; Davis et al., 1976; Davis, 1977; Smith et al., 1985). A major goal of Kaapvaal lithosphere studies is to relate current lithospheric structure to previous episodes of kimberlite eruption and to the distribution of Archean and Proterozoic peridotite, eclogite, and diamonds within the lithosphere. For the present study, we wish to put two decades of research on the geochronological age and composition of inclusion-bearing diamonds from southern Africa's major diamond deposits (Harris, 1992; Pearson and Shirey, 1999), comprising some 4000 individual diamond specimens, into regional geologic context at diamond source depths. The goal is to uncover any connection between southern African diamonds and the composition of their mantle source. We are specifically looking for any regional control of lithospheric mantle seismic structure on the formation of diamond or whether geological processes associated with diamond formation produced the extant mantle seismic structure.

2. Methods and sources of data

2.1. Seismic data

Seismic velocities in the lithospheric mantle beneath the Kaapvaal–Zimbabwe cratons have been mapped using teleseismic broadband waveform data from the period 1997–1999, gathered during the SASE (James et al., 2001; James and Fouch, 2002). For constructing tomographic images of the lithosphere used in this study, P-wave data have been used because the available data set is nearly twice as large as that available for S-waves (James and Fouch, 2002). To produce quantitative P-wave anomaly data for each diamond source area, a 50-km radius cylinder of mantle extending from 150 to 225 km depth has been averaged. In the tomographic image and the data table, seismic velocity anomalies for P-waves are represented as % deviation of this average from a cratonic reference model.

2.2. Ages on inclusions in diamond

The compositions of silicate inclusions in diamond can be grouped according to their mineralogical similarity to eclogite or peridotite xenoliths in kimberlite. Diamonds are classified as peridotitic or 'P-type' (or U-type by some workers, e.g. Gurney and Switzer, 1973; Sobolev et al., 1998) when they contain Cr-pyrope, diopside, enstatite, chromite, or olivine (Meyer and Boyd, 1972; Harris and Gurney, 1979). P-type diamonds can be further subdivided into harzburgitic or lherzolitic by the degree of depletion indicated by the Cr_2O_3 and CaO content of their included garnet and when applicable, the composition of included olivine or orthopyroxene (e.g. Sobolev et al., 1973; Harris and Gurney, 1979). Diamonds with silicate inclusions are classified as eclogitic or 'E-type' when they contain pyrope-almandine garnet, omphacite, coesite, or kyanite.

Sulfide inclusions in diamonds are chiefly intergrowths of pyrrhotite and pentlandite with subordinate chalcopyrite that have exsolved from a monosulfide solid solution. A similar E versus P paragenetic distinction, as seen for silicate inclusions, has been applied to sulfide inclusions. P-type sulfides from the Siberian craton typically contain Ni contents greater than 22.8 wt.% whereas E-type sulfides typically have less than 8 wt.% (Yefimova et al., 1983). Such a clear break between E- and P- type sulfides has not yet been established for inclusions in Kaapvaal craton diamonds (Harris and Gurney, 1979; Deines and Harris, 1995) and with the advent of Re–Os isotopic studies on individual sulfides, it has been suggested that Os content of the sulfide is a more accurate discriminant (Pearson et al., 1998; Pearson and Shirey, 1999). Thus, for the present work, if bulk sulfide inclusions have high pentlandite content (e.g. Ni equivalent to >16 wt.% in monosulfide solid solution) and 2–30 ppm Os, they are referred to as P-type and if they have low pentlandite content (e.g. Ni equivalent to < 9 wt.% in monosulfide solid solution) and less than 200–300 ppb Os, they are referred to as E-type.

Over the past two decades, ages of diamonds from the major mines have been derived from Rb–Sr and Sm–Nd studies of silicate inclusions by Richardson and coworkers (Richardson (1986); Richardson et al. (1984, 1990, 1993, 1999)). Due to the low concentrations of Rb, Sr, Sm, and Nd in garnet and clinopyroxene and the small size of such inclusions, these studies added together many inclusions of similar mineral composition to produce isochrons that represent many tens to hundreds of diamonds from any one kimberlite. Early attempts to date sulfide inclusions relied on rare large grains and the Pb–Pb isotope system (e.g. Kramers, 1979). But within the last 5 years, improvements in Re–Os analytical sensitivity have made the dating of individual sulfide inclusions in diamonds routine (Pearson et al., 1998; Pearson and Shirey, 1999; Richardson et al., 2001).

Data used in this study come from this literature and the ongoing work of Shirey et al. (Orapa) and Richardson et al. (Jwaneng). It is important to recognize the large variability of inclusion suites between kimberlites and that there may be some bias in the total data set from sample selection. For example, while E-type silicates and E-type sulfides have clear eclogitic affinities, a connection to the eclogite host rocks in the lithosphere can best be established at kimberlites where both sulfide and silicate E-type inclusions (in separate diamonds) occur commonly (e.g. Jwaneng, Orapa, and Premier). An ideal situation would be to have silicate and sulfide E-type inclusions in the same diamond, but such diamonds with inclusions large enough for analysis are exceedingly rare. In addition, to get sulfides large enough for single grain analyses of Re and Os, the larger stones are routinely sought. The abundance of E-type diamonds is observed to increase with increasing diamond size for both sulfide and silicate inclusions (Gurney, 1989; Sobolev et al., 2001; Viljoen, unpublished data), thus, in seeking larger stones, there is a greater chance that they will be E-type. This may provide an explanation for why, with the exception of one P-type diamond from Koffiefontein, all sulfide inclusions analyzed to date from southern Africa have been E-type. All available Kaapvaal inclusion ages prior to 1999 have been summarized recently in Pearson and Shirey (1999) which also contains detailed discussion of inclusion syngenecity and paragenesis.

2.3. C isotopic composition

The carbon isotopic composition of diamonds from the major mines in southern Africa has been determined over the last two decades by total combustion of inclusion-bearing diamond chips and analysis of the resultant CO_2 by Deines (1980), Deines et al. (1984, 1987, 1989, 1991a,b, 1993, 1995, 1997, 2001) and Cartigny (1998), Cartigny et al. (1998, 1999). Sample sizes for the chips in these studies ranged from 0.3 to 1.3 mg for the work of Deines et al. (e.g. 1984) and from 1 to 4 mg for that of Cartigny et al. (e.g. 1998). Based on the type of C isotopic variability of growth horizons seen in ion microprobe analyses of diamond plates (Hauri et al., 1999, 2002), bulk analyses of chips of this size suggest that any C isotopic variability associated with diamond growth zonation would have been mixed together in these isotopic analyses.

2.4. N abundance and aggregation

Fourier transform infrared spectroscopy (FTIR) studies of the abundance and crystal–chemical aggregation of trace nitrogen in the diamond lattice were also carried out on diamond chips from the same samples that were used for carbon isotopic study by Deines et al. (1984, 1987, 1989, 1991a, 1991b), Deines and Harris (1995), Cartigny (1998) and Cartigny et al. (1998, 1999). Diamonds with no detectable N are termed Type II and their percentage in a diamond population reflects low partitioning of N into diamonds during their growth in the mantle. Sufficiently old diamonds with sufficiently high N will show clustering or aggregation of N into optically active centers (see Wilks and Wilks, 1991, pp. 67–77 for a discussion of diamond types and optical centers based on N). Diamonds with enough N to detect optically (termed Type Ia diamonds) will typically progress from C-centers (Ib; single N) through A-centers (IaA; paired N) to B-centers (IaB; clusters of four N and a vacancy) in proportion with geological time (hundreds of Ma), temperature, and nitrogen content (Evans and Qi, 1982; Evans and Harris, 1989; Taylor et al., 1990; Navon, 1999). Of these three variables, N aggregation will be sensitive chiefly to temperature (Navon, 1999) but still is a function of N content. Thus, diamond populations with a lesser percentage of highly aggregated nitrogen (e.g. low % Type IaB) are more likely to have resided in cooler lithosphere whereas diamonds with a higher Type IaB nitrogen aggregation state are more likely to have resided in warmer lithosphere. As mentioned above for C isotopic studies, the diamond chips analyzed by FTIR for N are large enough to include multiple diamond growth zones.

3. Results

3.1. Seismic velocity variations

Fig. 1, showing the tomographic inversion for P-wave data, presents a picture of the current lithospheric seismic velocity structure at a depth of 150 km. The Kaapvaal–Zimbabwe craton is marked by relatively high P-wave velocity lithospheric mantle that occurs in two prominent but irregularly shaped lobes separated by a broad west-northwest trending band of relatively lower velocity mantle. The seismic array coverage crosses the margin of the craton only in the southwest (Fig. 1). There, the craton boundary as mapped at the surface is marked by a change in the regional magnetic fabric (Ayres et al., 1998), an abrupt change in Re–Os model age of mantle xenoliths (Janney et al., 1999; Carlson et al., 2000; Irvine et al., 2001), and a sharp decrease of about 1% in seismic velocity. Very low mantle velocities are evident off-craton in the far southwest underneath the western Cape Foldbelt. Kimberlites distributed across southern Africa have diamonds that derive from mantle with large differences in seismic velocity (Table 1, Fig. 1). Jwaneng, Letlhakane, Orapa, and Premier diamonds were hosted in slower lithospheric mantle with relative P-wave velocity perturbations that vary from − 0.209% to − 0.006% whereas diamonds in the Roberts Victor, Jagersfontein, Finsch,

Fig. 1. Tomographic image of the lithospheric mantle derived from seismic P-wave data at a depth of 150 km (James et al., 2001; James and Fouch, 2002). The color scheme depicts % deviation from an average cratonic lithosphere velocity model. Areal coverage spans the lithospheric mantle of the Kaapvaal (K) and Zimbabwe (Z) cratons and the Limpopo mobile belt (L; see inset, left). Bold green line indicates the outermost boundary of the Archean cratons as defined by a sharp break between Archean and Proterozoic Re–Os ages on peridotite xenoliths (Carlson et al., 1999; Janney et al., 1999; Irvine et al., 2001). The location of diamond mines is shown by colored squares. Red squares are localities whose silicate inclusion in diamond suites are predominately eclogitic (Jagersfontein = JA, Jwaneng = JW, Letlhakane = LE, Orapa = O, Premier = P) and green squares are localities whose silicate inclusion suites are predominately peridotitic (Kimberley area mines of Wesselton, Bultfontein, and Dutoitspan termed De Beers Pool = D, Finsch = F, Koffiefontein = KO, Roberts Victor = R, and Venetia = V). Mines located above red-yellowish areas referred to in text as having diamonds derived from seismically slower mantle; mines located above greenish-blue areas referred to in text as having diamonds derived from seismically faster mantle. Reprinted with permission from Shirey et al. (2002). Diamond genesis, seismic structure, and evolution of the Kaapvaal–Zimbabwe craton. Science, 297: 1683–1686. © 2002 American Association for the Advancement of Science.

Venetia, Koffiefontein, and the Kimberley area kimberlites (Bultfontein, De Beers, Dutoitspan, Wesselton; known collectively as the *De Beers Pool*) were hosted in seismically faster mantle with relative P-wave velocity perturbations that vary from +0.084% to +0.357%.

3.2. Sulfide inclusions in diamond

Data on single E-type sulfides from the De Beers Pool, Jwaneng, Koffiefontein, and Orapa form a $^{187}Re/^{188}Os$ versus $^{187}Os/^{188}Os$ array (Fig. 2) that has a circa 2.9 Ga age slope (Pearson et al., 1998;

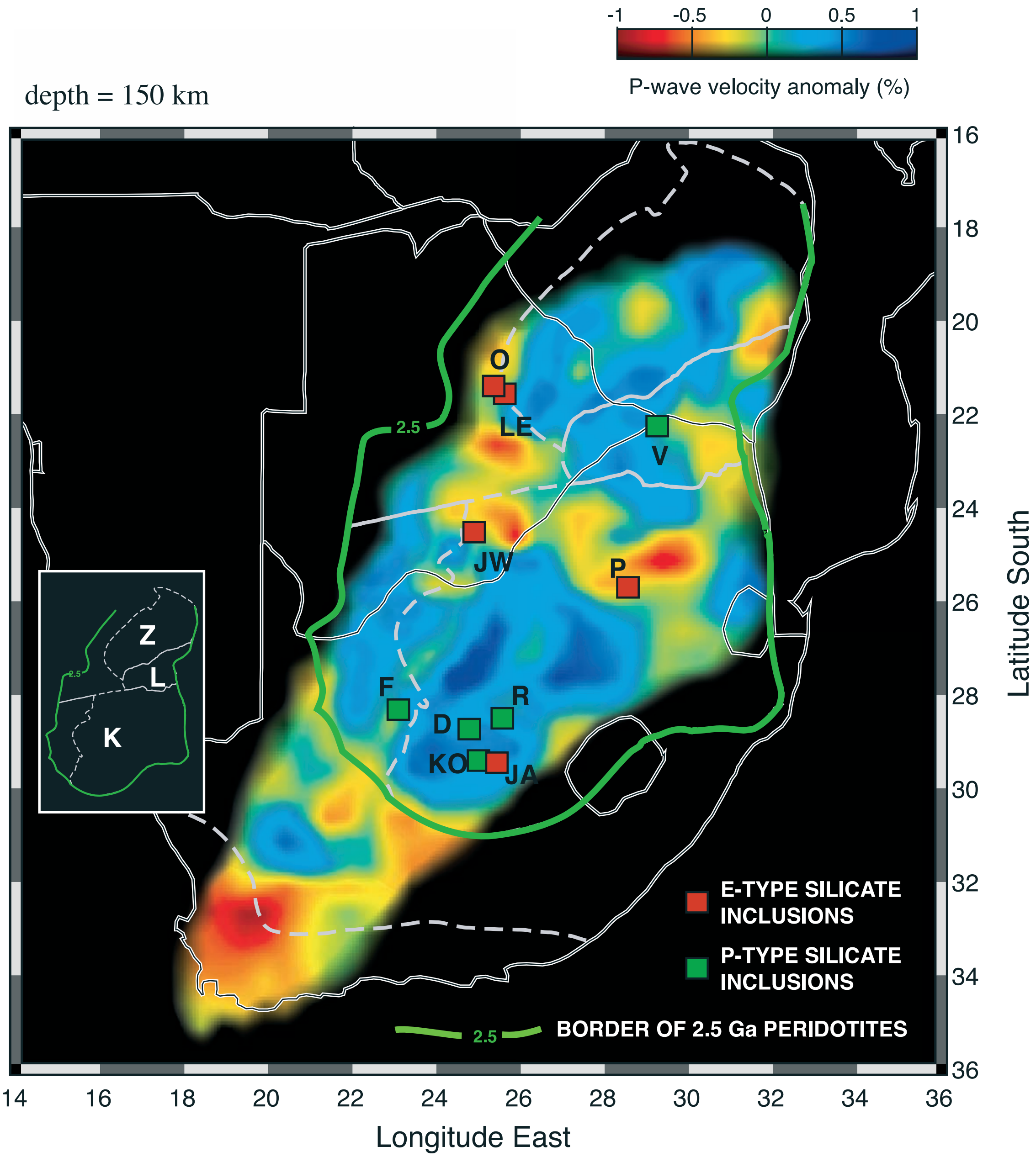

Table 1
Seismic velocity of the lithospheric mantle, inclusion Re–Os or Sm–Nd age (Ma), diamond carbon isotopic composition, nitrogen abundance and aggregation, and paragenesis compiled for Southern African diamonds and their localities

Location	Seismic velocity	Sulfide inclusion		Silicate inclusion		$\delta^{13}C$		N and N aggregation		
		Re–Os Age (Ma)	Para	Sm–Nd Age (Ma)	Para	Range (‰)	% < −9	N (ppm)	% Type Ia	Para
Jwaneng	−0.006	1500 to 2900	E	1540	E	−19 to −2	52	400	91	E,P
Letlhakane	−0.008							345	87	E,P
Orapa	−0.010	1000 to 2900	E	990	E	−26 to −3	65	478	94	E,P,(W)
Premier	−0.209			1150 to 1930	E,(P)	−12 to −2	1	413	90	E,P
Venetia	0.194					−18 to −2	11	259	77	P, (E,W)
De Beers Pool	0.245	2900	E	3200	P	−7 to −3	9	170	55	P,(E)
Finsch	0.084			1580 to 3200	P,(E)	−8 to −3	0	199	74	P,(E)
Roberts Victor	0.211					−16 to −3	71	260	73	P,(E)
Jagersfontein	0.357					−21 to −3	83	291	81	E,P
Koffiefontein	0.327	990 to 2900	E			−17 to −2	10	201	85	P,E

Average seismic velocity for P-waves in % deviation from a cratonic reference model for a 50 km radius cylinder of mantle extending from 150 to 225 km depth (James et al., 2001; James and Fouch, 2002). Parageneses (P = peridotitic, E = eclogitic, W = websteritic) are listed in relative order of abundance; subordinate parageneses are in parentheses. About 100 individual sulfide inclusions have been analyzed for Re–Os ages; about 3000 silicate inclusions (mostly composites but a few individual grains) have been analyzed for Sm–Nd ages. C isotopic and N aggregation studies have been carried out on diamonds enclosing silicate, oxide and sulfide inclusions; more than 900 individual diamonds have been studied. C isotopic composition is represented with the $\delta^{13}C$ notation in ‰ relative to the PDB standard. The percentage of diamonds with $\delta^{13}C$ less than −9‰ out of the total number of diamonds studied is shown under the column labeled '% < −9'. Nitrogen concentration is the average of the total diamond population as measured by Fourier transform infrared spectroscopy (FTIR); De Beers Pool data is by mass spectrometry. '%Type Ia' data are the percentage of diamonds in the studied population with aggregated nitrogen >20 ppm. Sources of data are from the literature as follows: De Beers Pool (Richardson et al., 1984, 2001; Cartigny, 1998; Cartigny et al., 1998); Finsch (Deines et al., 1984, 1989; Richardson et al., 1984, 1990; Smith et al., 1991); Jagersfontein (Deines et al., 1991a), Jwaneng (Cartigny et al., 1998; Richardson, unpublished data; Richardson et al., 1999); Koffiefontein (Deines et al., 1991a; Pearson et al., 1998); Orapa (Richardson et al., 1990; Shirey, unpublished data; Deines et al., 1991b, 1993); Premier (Milledge et al., 1983; Deines et al., 1984, 1989; Richardson, 1986; Richardson et al., 1993); Roberts Victor (Deines et al., 1987); Venetia (Deines et al., 2001; Viljoen, 2002). Note that the sulfide, silicate, and C–N studies were all carried out on separate suites of inclusion-bearing diamonds.

Richardson et al., 2001; Shirey et al., 2001). For each of these four inclusion suites individually, most of the inclusions have Re–Os systematics that approximate a 2.9 Ga age slope. De Beers Pool sulfide inclusions shows the tightest array whereas Jwaneng, Orapa, and Koffiefontein have a significant percentage of sulfide inclusions that plot at clearly younger ages ranging from 1 to 1.5 Ga. There is no obvious correspondence of sulfide inclusion age with lithospheric seismic velocity as both fast and slow mantle apparently hosted diamonds with circa 2.9 Ga ages. However, younger ages may be more frequent for diamonds from localities in slower mantle (e.g. Jwaneng for which more radiogenic data is not shown in Fig. 2). The circa 2.9 Ga age is not resolvable from the Re–Os model ages obtained on mantle peridotites whose median can be either 2.7 Ga if using a time of *Re depletion* (T_{rd}) approach or 3.1 Ga if using a time of *mantle reservoir separation* (T_{ma}) approach (Carlson et al., 1999, 2000; Irvine et al., 2001).

3.3. Silicate inclusions in diamond

Sm–Nd isochron and mantle model ages for silicate inclusion suites from the Kaapvaal–Zimbabwe craton are shown in Fig. 3. P-type (harzburgitic) garnets from Finsch and De Beers Pool (Richardson et al., 1984) have the oldest model ages yet recorded for inclusions in diamond from southern Africa. Premier P-type (lherzolitic) garnets have a much younger isochron age (1.9 Ga, Richardson et al., 1993) that, with reasonable assumptions of protolith Sm/Nd (see Fig. 3, caption) would lead to a younger 3.0 Ga depleted mantle model age compared to the De Beers Pool inclusions. All other southern African silicate inclusion suites dated with the Sm–Nd system (Richardson, 1986, 1990; Smith et al., 1991, 1999) are

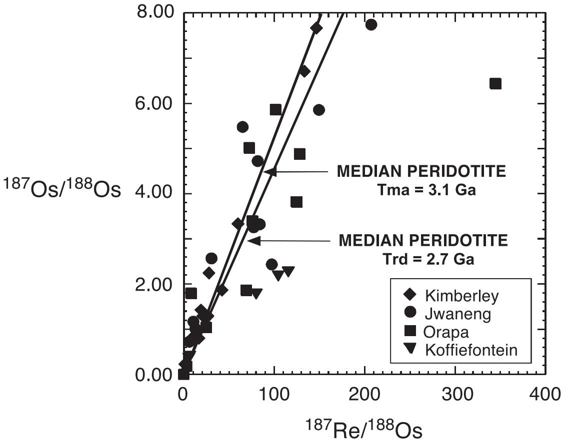

Fig. 2. Re–Os isotopic array for individual sulfide inclusions in single diamonds compared to typical Re–Os model ages on peridotites from the Kaapvaal–Zimbabwe craton. Figure modified after Shirey et al. (2001). Data sources are as follows: De Beers Pool (Richardson et al., 2001), Jwaneng (Richardson et al., unpublished), Koffiefontein (Pearson et al., 1998), and Orapa (Shirey et al., unpublished). Peridotite Re–Os model ages from Irvine et al. (2001) and Carlson et al. (1999).

E-types and yield Proterozoic isochron or model ages. P-type silicate inclusion model ages in the 3.2–3.3 Ga range overlap some of the older crustal U–Pb ages obtained (Moser et al., 2000; Schmitz, 2002), but Fig. 3 shows that they predate the Re–Os model ages of many peridotite xenoliths (Carlson et al., 1999; Irvine et al., 2001), sulfide inclusions in diamond (see Fig. 2), diamondiferous eclogite xenoliths (Shirey et al., 2001; Menzies et al., 2003), and the U–Pb ages of crustal metamorphism and plutonism (Schmitz, 2002). A common history for these diverse lithospheric components could be postulated from circa 2.9 Ga onwards.

Most diamondiferous kimberlites have diamond populations with silicate inclusions from both P-type and E-type parageneses (Table 1). It is whether the majority of the inclusions are eclogitic or peridotitic that shows a correspondence with slow versus fast lithospheric mantle (Fig. 1). Jwaneng, Letlhakane, Orapa, and Premier have inclusion-bearing diamond populations where E-type inclusions predominate; these kimberlites have penetrated seismically slower mantle. De Beers Pool, Finsch, Koffiefontein, Roberts Victor, and Venetia have inclusion-bearing diamond populations where P-type inclusions predominate; these kimberlites have penetrated seismically fast mantle. Only Jagersfontein does not fit this pattern, having penetrated seismically faster mantle but containing an E-type dominant silicate inclusion suite. Jagersfontein diamonds may be exceptional because of the evident fertilization and metasomatism of Jagersfontein xenoliths (Haggerty, 1983; Winterburn et al., 1990; Pyle and Haggerty, 1998) and the presence of sublithospheric xenoliths (Haggerty and Sautter, 1990) and diamonds (Deines et al., 1991b).

3.4. C isotopic compositions

It has long been known (Sobolev et al., 1979) that diamonds of both P-type and E-type paragenesis show a prevalent mantle-like carbon isotopic composition ($\delta^{13}C = -3$‰ to -7‰) with an isotopically light sub-population ($\delta^{13}C = -10$‰ to -34‰),

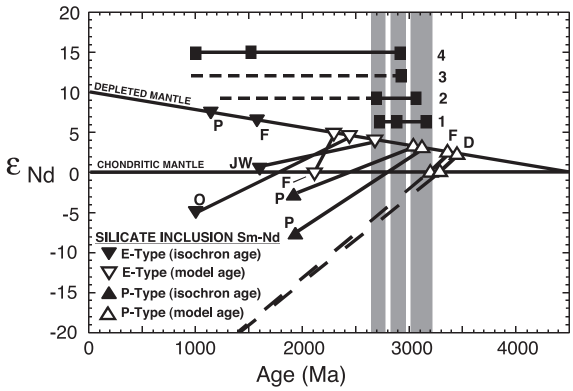

Fig. 3. Sm–Nd isochron and model ages of silicate inclusions in diamond (sources, Table 1 caption) compared to the ages of major crust forming events in the Kaapvaal craton (1; Schmitz, 2002), peridotite xenolith Re–Os model ages (2; sources, Fig. 2 caption), diamondiferous eclogite xenoliths (3; Shirey et al., 2001; Menzies et al., 2003), and sulfide inclusions in diamonds (4; sources, Fig. 2 caption). Growth curves from the isochron age (crystallization) of silicate inclusions to their reservoir separation from depleted mantle are estimated by assuming that the measured Sm/Nd of the clinopyroxene associated with lherzolitic garnet at Premier and the clinopyroxene associated with eclogitic garnet at Finsch, Jwaneng and Orapa represent a maximum (and perhaps typical) Sm/Nd of the protolith. These assumptions can be supported because most of the light REE in an eclogite will reside in clinopyroxene (e.g. Taylor et al., 1996) and lherzolitic garnet and associated clinopyroxene show regular REE patterns (e.g. non-sinusoidal; Stachel and Harris, 1997; Stachel et al., 1998).

dominated by diamonds of E-type paragenesis (e.g. Gurney, 1989; Galimov, 1991; Kirkley et al., 1991). If the C isotopic composition of all southern African diamonds studied is broken down by either paragenesis or the seismic velocity of the source mantle, it can be seen that the distribution of C isotope composition for E-type diamonds (Fig. 4A) appears nearly identical to that for diamonds from the seismically slow lithospheric mantle (Fig. 4B). But if the eclogitic diamonds with a C isotopic composition less than -9‰ (e.g. those that comprise the isotopically light tail of the δ^{13}C distribution) are treated mine by mine (Table 1), it can be seen that the large number of specimens from Jwaneng and Orapa dominate the isotopically light population and its correlation with seismically slow mantle. Premier, Jagersfontein, and Roberts Victor also directly contraindicate such a correlation: Premier lies above seismically slow mantle but has no isotopically light diamonds in its E-type population whereas Jagersfontein and Roberts Victor lie above seismically fast mantle but have their E-type populations dominated by diamonds with isotopically light carbon (Table 1). Therefore, while paragenesis is the controlling factor in the differences in C isotopic composition of diamonds from seismically fast versus slow mantle, the cratonwide distribution of E-type diamonds with isotopically light C is not straightforward.

3.5. N abundance and N aggregation

Nitrogen abundance shows a correspondence with lithospheric seismic structure that is linked to paragenesis (Fig. 5A and B; Table 1), the statistically significantly higher N content of E-type diamonds noted previously for Finsch, Jwaneng, Orapa, and Premier by Deines et al. (1989, 1993, 1997). Diamonds from slower lithospheric mantle (Jwaneng, Letlhakane, Orapa, and Premier localities) have a higher percentage of Type Ia diamonds and an average N content above 300 ppm to accompany their greater percentage of E-type inclusions (Fig. 5A and B). Except for Jagersfontein, diamonds from faster lithospheric mantle (Koffiefontein, Finsch, Roberts Victor, Venetia, and the De Beers Pool) have a lower percentage of Type Ia diamonds and an average N content below 300 ppm to accompany their greater percentage of P-type inclusions. The aggregation state

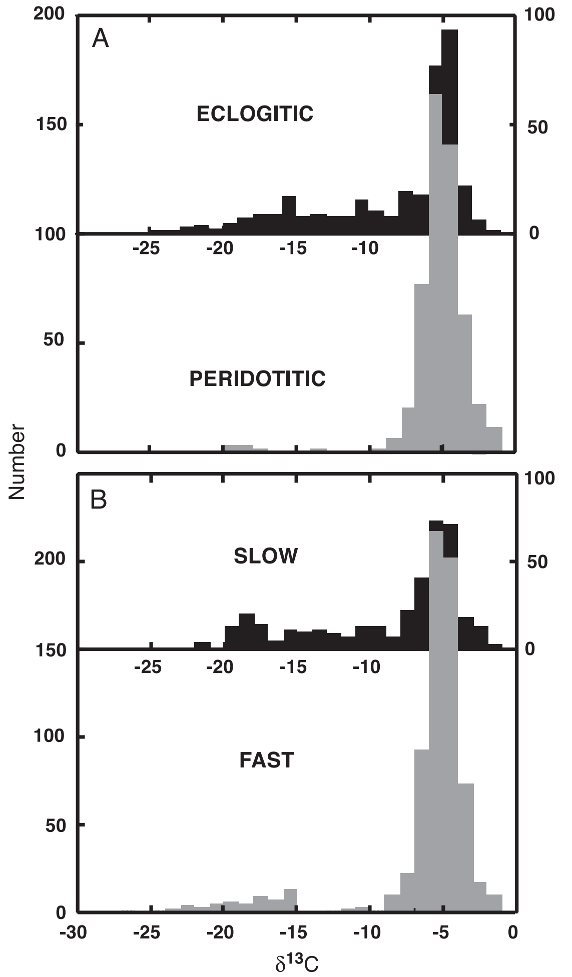

Fig. 4. (A) Comparison of the carbon isotopic composition of individual diamond analyses grouped according to E-type or P-type paragenesis. Note that both E-type and P-type histograms include diamonds from all nine localities. (B) Comparison of the carbon isotopic composition of southern African diamonds, grouped according to their derivation from a locality in seismically slower (Jwaneng, Letlhakane, Orapa, and Premier) or seismically faster lithospheric mantle (Venetia, De Beers Pool, Finsch, Roberts Victor, Jagersfontein and Koffiefontein). The similarities in the histograms in A and B occur because a greater proportion of diamonds of E-type paragenesis and isotopically light carbon derive from localities occurring in seismically slower lithospheric mantle. See text and Table 1 for sources of data. Reprinted with permission from Shirey et al. (2002). Diamond genesis, seismic structure, and evolution of the Kaapvaal–Zimbabwe craton. Science, 297: 1683–1686. © 2002 American Association for the Advancement of Science.

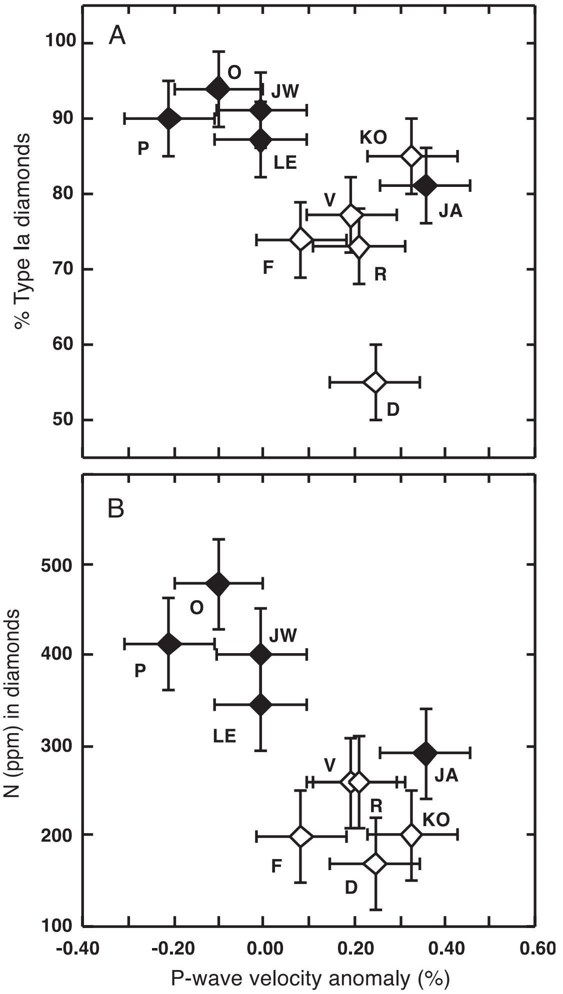

Fig. 5. Percentage of Type Ia diamonds (5A) and average N abundance in diamonds (5B) versus P-wave velocity anomaly. Error bars have been set at the ± 5 % level for percentage of Type Ia diamonds, ± 50 ppm for average N abundance, and ± 0.1 % for P-wave velocity anomaly. Type Ia diamonds are the total of IaA, IaA/B and IaB diamonds (Table 1). Same lettering scheme as in Fig. 1. Peridotitic = open symbols; eclogitic = closed symbols. Reprinted with permission from Shirey et al. (2002). Diamond genesis, seismic structure, and evolution of the Kaapvaal–Zimbabwe craton. Science, 297: 1683–1686. © 2002 American Association for the Advancement of Science.

of N in the Ia diamonds (not shown) displays no systematic variation with lithospheric seismic velocity although De Beers Pool, Finsch, and Roberts Victor diamonds from seismically fast mantle display the lowest percentages of aggregation to B-centers.

4. Discussion

4.1. Cratonic keel structure and diamond age distribution

With the advent of new age information on mantle lithologies and diamonds from the Re–Os system, it is interesting to re-evaluate the timing of diamond formation in the mantle beneath southern Africa and its relationship to seismic structure. There was a long-standing suspicion that cratonic mantle keels are old (Holmes and Paneth, 1936; Kramers, 1979). This initially was confirmed for the Kaapvaal craton by Richardson et al. (1984) using the Sm–Nd system on harzburgitic garnets included in diamonds from Finsch and the De Beers Pool that gave mid-Archean ages. Subsequent work of Richardson et al. (1986), Richardson et al. (1990, 1993, 1999) on diverse eclogitic and lherzolitic silicate inclusions in diamonds produced Proterozoic ages. Recent Re–Os data shows a preponderance of old eclogitic sulfide inclusion ages and cratonic peridotite mantle model ages of late Archean age (Carlson et al., 1999; Irvine et al., 2001; Shirey et al., 2001). These ages confirm that widespread diamond formation took place in a cratonic root that was being created or stabilized in the late Archean.

In detail, harzburgitic silicate inclusions in diamonds from the De Beers Pool and Finsch that give the oldest mid-Archean model ages (3.2–3.3 Ga; Richardson et al., 1984) currently have been found only above seismically faster lithospheric mantle. All other silicate inclusion suites dated so far are either lherzolitic or eclogitic, Proterozoic in age (Richardson, 1986, 1990, 1993, 1999) and occur above either seismically fast or slow mantle. Eclogitic sulfide inclusions from the four localities studied so far have all given late Archean ages (Pearson et al., 1998; Richardson et al., 2001; Shirey et al., 2001) as well as occasional Proterozoic ages that match the Proterozoic ages of the eclogitic silicate inclusions. Eclogitic sulfide inclusions are found above either seismically slow or fast mantle.

The connection between inclusion paragenesis and lithospheric seismic structure (Fig. 1) is clear only for silicate inclusion suites in diamonds. Most of these suites have not had age determinations because their diamonds were studied chiefly for C isotopic composition, N abundance and N aggregation (Table 1). It is noted that dated P-type silicate inclusions yield Archean (model) ages (Finsch, DeBeers Pool, Premier; Richardson et al., 1984, 1993, Fig. 3) and E-type silicate inclusions give Proterozoic model ages (Jwaneng, Orapa, Premier, Finsch; Richardson, 1986; Richardson et al., 1990, 1999). The best explanation for the complexities in the range and distribution of diamond ages and the distribution of diamond inclusion types lies in the uneven make-up of the overall diamond sample and the non-overlapping nature of the extant diamond studies in addition to the episodic nature by which the cratonic keel was generated, stabilized, and modified. The picture emerging from the diamond data is of a keel that has retained distinct populations of diamonds that were formed during each of the major tectonothermal episodes to have affected the cratonic keel (Shirey et al., 2002).

4.2. Age of the South African lithospheric seismic velocity structure

Several lines of evidence suggest that the current lithospheric seismic structure of the craton is a mid-Proterozoic overprint to this predominantly 2.9–3.3 Ga keel. Proterozoic E-type, silicate inclusion-bearing diamonds from seismically slow lithosphere (Figs. 1 and 3; Table 1) occur in the same kimberlites that contain Archean E-type sulfide inclusions (e.g. Orapa and Jwaneng) or peridotites with Archean Re–Os model ages (e.g. Letlhakane, Irvine et al., 2001). The Premier kimberlite, penetrating seismically slow mantle, not only contains peridotite xenoliths with Archean Re–Os model ages but also the clearest example of peridotite overprinted in the Proterozoic (Carlson et al., 1999; Irvine et al., 2001). Premier P-type silicate inclusions (lherzolitic garnet and clinopyroxene) that were equilibrated at the 1.9 Ga isochron age which represents the time of encapsulating diamond growth (Richardson et al., 1993) have Sm–Nd mantle model ages that are Archean (Fig. 3).

Modification of the cratonic root, which was originally thought to affect chiefly the Premier locality on the basis of Re–Os studies of lithospheric peridotite (Carlson et al., 1999; Irvine et al., 2001), is apparently more widespread than originally thought, extending across the northern Kaapvaal craton and to the south of the Zimbabwe craton–Limpopo Belt some hundreds of kilometers from the craton edge. Correlation of the seismically slow regions of the northern Kaapvaal Craton that trend ESE–WNW south of the Limpopo belt (Fig. 1) with surface outcrop of the 2.05 Ga Bushveld Complex in South Africa and Molopo Farms Complex in Botswana suggests that the modification may be closely related to Bushveld–Molopo magmatism. For the seismically slow mantle that trends N–S on the west side of the Zimbabwe craton (Fig. 1), regional metamorphism that created the Magondi–Okwa terranes was likely to have been the surface manifestation of the tectonism that modified the craton on this margin.

4.3. Lithospheric mantle composition, diamond growth and storage

Recent thermal structure of the Kaapvaal lithospheric mantle from surface heat flow (Jones, 1988) and cratonic geotherms from xenolith geothermobarometry (Danchin, 1979) show that even in the seismically slow areas, such as near Premier, a normal cratonic geotherm has existed since at least the Premier eruption age of 1.2 Ga. This is evidence that the lithosphere is seismically slower chiefly because it is compositionally different, not hotter. The seismically slow region of the lithosphere is likely higher in basaltic components (Fe, Ca, clinopyroxene) and metasomatizing veins which hydrate and alter the vein wall of the host peridotite. These are the main petrological differences that would be expected to account for the 1% difference in P-wave velocity seen in Fig. 1.

Differences in silicate inclusion paragenesis and their diamond host composition correlate with the compositional differences recorded in the seismic structure of the lithosphere. Diamond suites from seismically slower lithosphere have a greater percentage of eclogitic inclusions which is in direct agreement with regions of the lithosphere that would have a higher proportion of basaltic components. Also, diamond suites from these regions have the eclogitic subset with isotopically light $\delta^{13}C$, the highest percentage of Type Ia stones and a higher average N content (>300 ppm). It is not clear whether higher average N incorporation during growth of these diamonds was due to a diamond-forming fluid with higher N content or due to faster growth. The lack of an obvious difference in N aggregation characteristics between diamonds from seismically slow and fast mantle (Table 1) indicates that diamonds from both types of mantle were stored in the lithosphere at hot enough temperatures (e.g. 1150 ± 50 °C) for long enough to allow substantial aggregation of N to B centers (Evans and Harris, 1989; Navon, 1999). In this case, the low IaB % of De Beers Pool, Finsch, and Roberts Victor diamonds (Table 1) would be related perhaps more to the lower average N content of these diamond populations than to temperature. Simple heating of the lithosphere in the seismically slow regions, as might be suggested by the resetting of U–Pb ages in low-closure-temperature minerals such as rutile found in lower crustal granulites elsewhere (Schmitz, 2002) is not recorded in the N aggregation data. This could be because any thermal pulse was too short-lived (Danchin, 1979) for substantial N aggregation to occur in a short period (Richardson and Harris, 1997; Navon, 1999). The current N aggregation data also are not detailed enough to resolve any systematic temperature or depth differences between E-type and P-type diamonds.

Diamonds and their inclusions examined on the regional scale of this study are surprisingly good at retaining evidence of lithospheric processes and craton-scale compositional effects. Craton-wide patterns in xenolith and megacryst suites that correlate with lithospheric seismic structure are subtler perhaps because silicate mineral assemblages not protected by encapsulating diamond continue to equilibrate and are subject to infiltrating metasomatic fluids. Some Premier peridotites, for example, have younger Re–Os model ages attributable to interaction with Bushveld age melts (Carlson et al., 1999) and the Cr/Cr + Al of spinel megacrysts is lower (Schulze, 2001) in kimberlites from seismically slower lithosphere. These examples provide further evidence that the current physiochemical features that produce the observed seismic structure were created during Proterozoic modification of Archean lithosphere.

4.4. Creation, stabilization, and modification of lithospheric mantle

Constraints on lithosphere creation come from those diamonds that are Archean: the four suites with E-type sulfides that have been dated using the Re–Os system at around 2.9 Ga (Pearson and Shirey, 1999; Richardson et al., 2001, Richardson et al., unpublished data) and two suites with P-type silicates that have been dated using the Sm–Nd system at 3.2–3.3 Ga (Richardson et al., 1984). Until the advent of Re–Os analyses on sulfide inclusions, the prevalent model for lithosphere creation and stabilization involved degrees of melting high enough to create komatiite (e.g. Richardson et al., 1984; Walker et al., 1989; Boyd et al., 1999). Although this model failed to account for the high orthopyroxene (e.g. silica content) of the lithospheric mantle, it could simultaneously account for the high Mg# of peridotitic olivine, the bouyancy (and hence preservation) of the lithospheric mantle, its low heat production, its highly unradiogenic Os isotopic composition, and the harzburgitic composition of the 3.2–3.3 Ga garnet inclusions. Surprisingly, *none* of the Archean sulfide inclusions analyzed yet for Re–Os are P-type, they are all E-type. As seen in Figs. 2 and 3, typical ages do not cluster around the oldest P-type silicate age of 3.2–3.3 Ga (Richardson et al., 1984) or the dominant crust-forming age of 3.1 Ga (Moser et al., 2000; Schmitz, 2002) but have a typical age varying around a younger 2.9 Ga age. Nowhere is this age distinction clearer than with the De Beers Pool samples from the Kimberley area kimberlites. Here, 3.2–3.3 Ga P-type, depleted (harzburgitic) garnets coexist in the same lithospheric mantle section sampled by kimberlitic volcanism as 2.9 Ga E-type sulfides that have elevated, enriched initial Os isotopic composition.

All indications are that diamond formation in Archean cratonic mantle is episodic and such episodicity may apply to the formation and stabilization of the cratonic mantle itself. The occurrence of 3.2–3.3 Ga diamonds with depleted harzburgitic silicate inclusions (Sm–Nd, Rb–Sr model ages; Richardson et al., 1984) and 2.9 Ga diamonds with enriched eclogitic sulfide inclusions (Re–Os isochron age; Richardson et al., 2001) in the same Kimberley kimberlites (De Beers Pool) indicates that creation and assembly of the craton was *at least* a two-stage process (Shirey et al., 2002). Considering Re–Os, Sm–Nd, and Rb–Sr model age relationships, a time gap of 300 ± 200 million years is required between the two Archean diamond formation events recorded in De Beers Pool diamonds. Cratonic nuclei were first created by mantle-melting processes that produced severe depletion.

This could have occurred by vertical underplating (Haggerty, 1986) above a mantle plume (Arndt et al., 1997) although recent experimental and trace element work suggests that such depletion might have occurred initially at shallower depths (Canil and Wei, 1992; Stachel et al., 1998; Walter, 1998) or in a hot and wet subduction setting (Parman et al., 2001). Melting within a thickening volcanic pile led to crustal differentiation, the production of sial, and the first phase of crustal preservation. Preservation of the first-stage depleted mantle in the lithosphere may have been partial due to the magmatism and delamination required by the crustal differentiation process, an idea supported by the restricted distribution of 3.2–3.3 Ga harzburgitic silicate inclusions that thus far have only been documented from the Kimberley–Finsch area. It must be noted that the spatial distribution of extensively studied diamond-bearing kimberlites in southern Africa limit the above model for episodic creation and stabilization of the Kaapvaal–Zimbabwe craton chiefly to its western part. However, mid-Archean Re–Os model ages on lithospheric mantle peridotites, which have a slightly wider geographic distribution than the diamonds presently studied, are proposed similarly to reflect the existence of early depleted cratonic nuclei (Pearson et al., 2002).

The existence of these early cratonic nuclei provided a locus against which to accrete cratonic lithosphere created in the second step of the process (Shirey et al., 2002). In this phase, subduction of depleted oceanic harzburgite, (e.g. Helmstaedt and Schulze, 1989) which may have included the roots of Archean oceanic plateaus, built the rest of the lithospheric mantle. This second phase of lithosphere creation could account for widespread circa 2.9 Ga age Archean E-type sulfide inclusions in diamonds (Table 1; Shirey et al., 2001), their enriched initial Os isotopic composition (Richardson et al., 2001), the presence of enriched and depleted Archean inclusion chemistry in the same kimberlite, the younger age of these E-type diamonds compared to harzburgitic P-Type diamonds, the typical Re–Os model age for mantle peridotite (Carlson et al., 1999; Irvine et al., 2001), and the occurrence of 2.9 Ga diamondiferous eclogite with basaltic komatiitic compositions (Shirey et al., 2001; Menzies et al., 2003). This two-step lithosphere creation model also fits the detailed geochronological record for the lower and upper crust of the western Kaapvaal craton in which the earliest crustal components are formed from 3.20 to 3.26 Ga and the craton is sutured together by subduction convergence at 2.88–2.94 Ga (Schmitz, 2002).

Subduction for the western craton is supported by stable isotopic data from Orapa and Jwaneng. Orapa E-Type diamonds have light C and heavy N isotopic compositions (Cartigny et al., 1999) and sulfide inclusions with mass-independent sulfur isotopic fractionations (Farquhar et al., 2002) that are consistent with incorporation of C, N, and S from surficial sedimentary endmember reservoirs (Navon, 1999; Farquhar et al., 2002). Also, some Archean Jwaneng megacrystic zircon has low O isotope compositions suggesting that the zircon host lithologies were once hydrothermally altered as oceanic crust (Valley et al., 1998). Subduction is likely not to be the only source of C and N isotopic variability, however, because of the difficulties of finding subducted materials with appropriately high C/N ratios (e.g. Cartigny et al., 1998, 1999). Furthermore, intra-mantle processes (e.g. Cartigny et al., 2001) that must have been changing C and N isotopic composition of fluids during diamond growth are required by the isotopic composition differences observed in the growth horizons of some diamonds (Hauri et al., 1999; Hauri et al., 2002). The lack of complete C and N isotopic data sets from all but a few of south Africa's diamond suites means that currently it is not possible to discuss a cratonwide picture of intra-mantle isotopic fractionation of C and N in diamonds.

The near one-to-one correspondence of Proterozoic diamond suites having a majority of E-type silicate inclusions with seismically slow mantle suggests that craton modification and Proterozoic diamond formation were part of the same process (Shirey et al., 2002). Proterozoic craton modification that reduced the seismic velocity of the lithosphere did not apparently involve the addition of new mantle from the asthenosphere to the lithosphere. This is indicated by a lack of dominant Proterozoic Re–Os model ages on peridotites from the seismically slower portions of the lithosphere. Since the paragenesis of the Proterozoic silicate inclusions is chiefly E-type, it is likely that this process involved basaltic components generated in conjunction with Bushveld-Molopo Farms magmatism under the center of the northern Kaapvaal craton or in conjunction with some form of subduction along the western Kaapvaal–Zimbabwe craton margin. Both sublithospheric magmatism and western craton margin subduction were tectonothermal events that altered the composition of the lithosphere and added new diamonds to an already extensive Archean diamond population resident in the lithosphere.

5. Conclusions

Regional patterns of diamond composition, inclusion age, and paragenesis were derived from a data set comprising measurements on some 4000 individual stones or their inclusions. When these patterns are compared to the seismic velocity structure of the lithosphere in the diamond stability field, they lead to new insights on the creation of the lithospheric seismic velocity anomaly, the formation of various diamond generations, and the formation of continental lithosphere itself. Mantle lithosphere with slow P-wave velocity correlates with a greater proportion of eclogitic versus peridotitic silicate inclusions in diamond, a greater range in Sm–Nd age of silicate inclusions, and diamond suites that have a greater proportion of lighter C isotopic composition diamonds, a higher average N content, and more Type Ia diamonds. Mantle lithosphere with high P-wave velocity is typified by a higher proportion of peridotitic versus eclogitic silicate inclusions, the oldest Sm–Nd model ages, and diamond suites that have a lower average N content and fewer Type Ia diamonds.

The oldest formation ages of diamonds indicate that the mantle keels which became continental nuclei were created by middle Archean (3.2–3.3 Ga) mantle depletion events with high degrees of melting and early harzburgite formation. The predominance of sulfide inclusions that are eclogitic in the 2.9 Ga age population links late Archean subduction-accretion events involving an oceanic lithosphere component to craton stabilization which resulted in a widely distributed, late-Archean generation of eclogitic diamonds. Subsequent Proterozoic tectonic and magmatic events altered the composition of the continental lithosphere, produced the seismic velocity variations, and added new lherzolitic and eclogitic diamonds to the already extensive Archean diamond suite.

Tests of these ideas will come with future studies, including geographically more complete, overlapping data sets from individual mines, studies of sulfide inclusion composition (Fe, Ni, and Cu content) and C and N isotopic and FTIR studies on individual diamonds that have been dated with the Re–Os system. These need to be pursued in order to establish the relative importance of intra-mantle processes and regional variability on the isotopic composition of diamonds and the ultimate source of diamond-forming fluids.

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