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Integrated models of diamond formation and craton evolution

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Abstract

Two decades of diamond research in southern Africa allow the age, average N content and carbon composition of diamonds, and the dominant paragenesis of their syngenetic silicate and sulfide inclusions to be integrated on a cratonwide scale with a model of craton formation. 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 mid-Proterozoic and display little correspondence with the prominent variations in the P-wave velocity ($\pm 1\%$) that the mantle lithosphere shows at depths within the diamond stability field (150–225 km). Silicate inclusions in diamonds and their host diamond compositions for the above kimberlites, Finsch, Jagersfontein, Roberts Victor, Premier, Venetia, and Letlhakane show a regional relationship to the seismic velocity of the lithosphere. Mantle lithosphere with slower P-wave velocity relative to the craton average correlates with a greater proportion of eclogitic vs. 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. The oldest formation ages of diamonds support a model whereby mantle that became part of the continental keel of cratonic nuclei first was created by middle Archean (3.2–3.3 Ga or older) mantle depletion events with high degrees of melting and early harzburgite formation. The predominance of eclogitic sulfide inclusions in the 2.9 Ga age population links late Archean (2.9 Ga) subduction-accretion events to craton stabilization. These events resulted in a widely distributed, late Archean generation of eclogitic diamonds in an amalgamated craton. Subsequent Proterozoic tectonic and magmatic events altered the composition of the continental lithosphere and added new lherzolitic and eclogitic diamonds to the already extensive Archean diamond suite. Similar age/paragenesis systematics are seen for the more limited data sets from the Slave and Siberian cratons. © 2004 Elsevier B.V. All rights reserved.

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

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1. Introduction

A worldwide association between ancient cratons and diamond occurrences has long been known (e.g., Clifford, 1966; Gurney and Switzer, 1973; Boyd and Gurney, 1986; Janse, 1992). This association, plus the intersection of mantle xenolith-derived geotherms

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with the diamond stability field and the Archean ages of inclusions in diamonds led to the suggestion that diamonds reside in Archean lithospheric mantle keels beneath cratons (e.g., Richardson et al., 1984; Boyd et al., 1985; Boyd and Gurney, 1986; Haggerty, 1986). Within the southern African cratonic keel (the conjoined Kaapvaal-Zimbabwe cratons and the Limpopo mobile belt), Archean mantle peridotite and eclogite host multiple generations of diamonds that are both Archean and Proterozoic in age (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, mostly cubic and fibrous 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 studies of Archean lithosphere is to relate lithospheric structure to the age and chemical variation of peridotite, eclogite, and diamonds and the geological processes that created and assembled the cratons in the hope of arriving at a comprehensive model for diamond formation. This goal is now attainable for a number of reasons: (1) more than two decades of research on the composition and age of diamonds from southern Africa's major diamond deposits (comprising some 4000 individual diamond specimens in more than 30 research papers) have been carried out, (2) new advances have been made over the last decade in applying the Re-Os isotopic system to age determinations of key samples erupted in kimberlites (peridotites, eclogites, and sulfide inclusions in diamond), (3) high-precision U-Pb geochronology and thermochronology of the upper and lower crust are now available, and (4) the Kaapvaal Lithosphere Project has produced a new seismic picture of the lithosphere. In this synthesis, we focus on how the composition of southern Africa's diamonds and the various episodes of diamond growth fit with ideas of cratonic lithosphere creation, stabilization and modification.

2. Methods and materials studied

2.1. Seismic imaging

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 a picture of the lithospheric mantle at depths within the diamond stability field. To produce quantitative P-wave anomaly data for each diamond source area in the lithosphere, a 50 km radius cylinder of mantle has been averaged. This cylinder is centered below each diamond mine and extends from 150 to 225 km depth. Seismic data (Table 1) is presented as the P-wave velocity anomaly in % deviation from an average cratonic reference model (James et al., 2001; James and Fouch, 2002).

2.2. Inclusion mineralogy and sample distribution

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 when they contain Cr-pyrope, diopside, enstatite, chromite, or olivine (Meyer and Boyd, 1972; Gurney and Switzer, 1973; Harris and Gurney, 1979) or Fe-sulfide with pentlandite component making up >22 wt.% Ni in the bulk sulfide (Yefimova et al., 1983). Peridotitic diamonds can be further subdivided into harzburgitic or lherzolitic by the degree of depletion indicated by the Cr₂O₃ and CaO content of their included garnet, the absence or presence of diopside, and when applicable, the composition of included olivine or orthopyroxene (e.g., Sobolev et al., 1973; Harris and Gurney, 1979). Diamonds are classified as eclogitic when they contain pyropealmandine garnet, omphacite, coesite, kyanite or Fe sulfide with pentlandite component making up less than 8 wt.% of the bulk sulfide. Based on the strong correlation between Ni and Os content in sulfide inclusions, Pearson has suggested Os concentration is a good peridotitic vs. eclogitic discriminant (Pearson et al., 1998; Pearson and Shirey, 1999). Thus, South African sulfide inclusions with bulk Ni content >16 wt.% have 2-30 ppm Os and can be classified as peridotitic

| Seismic velocity | Locality paragenesis | Re-Os age (Ma) | Re-Os suite paragenesis | Sm–Nd age (Ma) | Sm-Nd suite paragenesis |
|---------------------|---|---|--|---|---|
| -0.006 | E, P | 1500-2900 | Е | 1540 | Е |
| -0.008 | E, P | | | | |
| -0.010 | E, P, (W) | 1000 - 2900 | Е | 990 | Е |
| -0.209 | E, P | | | 1150-1930 | E, (P) |
| 0.194 | P, (E, W) | | | | |
| 0.245 | P, (E) | 2900 | Е | 3200 | Р |
| 0.084 | P, (E) | | | 1580-3200 | P, (E) |
| 0.211 | P, (E) | | | | |
| 0.357 | E, P | | | | |
| 0.327 | Р, Е | 990-2900 | Е | | |
| | Seismic velocity - 0.006 - 0.008 - 0.010 - 0.209 0.194 0.245 0.084 0.211 0.357 0.327 | Seismic velocity Locality paragenesis - 0.006 E, P - 0.008 E, P - 0.010 E, P, (W) - 0.209 E, P 0.194 P, (E, W) 0.245 P, (E) 0.084 P, (E) 0.211 P, (E) 0.357 E, P 0.327 P, E | Seismic velocity Locality paragenesis Re-Os age (Ma) - 0.006 E, P 1500-2900 - 0.008 E, P - - 0.010 E, P, (W) 1000-2900 - 0.209 E, P - 0.194 P, (E, W) - 0.245 P, (E) 2900 0.084 P, (E) - 0.357 E, P - 0.327 P, E 990-2900 | Seismic velocity Locality paragenesis Re–Os age (Ma) Re–Os suite paragenesis - 0.006 E, P 1500–2900 E - 0.008 E, P 1000–2900 E - 0.010 E, P, (W) 1000–2900 E - 0.209 E, P 0.194 P, (E, W) 0.245 P, (E) 2900 E 0.084 P, (E) 0.211 P, (E) 0.357 E, P 990–2900 E | Seismic velocity Locality paragenesis Re-Os age (Ma) Re-Os suite paragenesis Sm-Nd age (Ma) - 0.006 E, P 1500-2900 E 1540 - 0.008 E, P - - - - 0.010 E, P, (W) 1000-2900 E 990 - 0.209 E, P 1150-1930 - 1150-1930 0.194 P, (E, W) 2900 E 3200 0.245 P, (E) 2900 E 3200 0.084 P, (E) 1580-3200 0.211 P, (E) 0.357 E, P 990-2900 E - |

Seismic velocity of the lithospheric mantle, locality paragenesis, inclusion Re-Os or Sm-Nd age (Ma) and paragenesis of inclusions for southern African diamonds

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). Locality parageneses [peridotitic (P), eclogitic (E), websteritic (W)] are listed in relative order of abundance. Subordinate parageneses are in parentheses. These data are from the work of Deines and Harris and Cartigny. 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. The ages given in the table have been generalized to reflect the major episodes of diamond formation as discussed in the text. 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 et al., 1999, 2004); Koffiefontein (Deines et al., 1991a; Pearson et al., 1984, 1989; Richardson et al., 1990; Deines et al., 1991b, 1993; Shirey et al., unpublished data); 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).

whereas sulfide inclusions with bulk Ni ≤ 9 wt.% have 200–300 ppb Os and can be classified as eclogitic.

Table 1

Inclusion suites display significant variability from kimberlite to kimberlite which can lead to sample selection bias in the data set. Also, age determinations from multiple isotopic systems generally have not been made on inclusions in diamonds from the same kimberlite (e.g., Table 1) usually because silicate and sulfide inclusions occur with different frequency in separate diamonds. In fact, eclogitic sulfides and eclogitic silicates occur commonly in only three of the major mines studied so far: Jwaneng, Orapa, and Premier. An ideal situation would be to have silicate and sulfide inclusions in the same diamond, but such diamonds with inclusions large enough for analysis are exceedingly rare in most mines. In addition, larger stones (3-4 mm in diameter) are routinely sought to get sulfides large enough for single grain Re and Os isotopic analysis. The relative abundance of eclogitic diamonds increases with increasing diamond size for both sulfide and silicate inclusions in some of the southern African mines (Gurney, 1989; Sobolev et al., 2001; Viljoen, unpublished data). Thus, in seeking larger stones there can be a greater chance that they will be eclogitic. This may provide an explanation for why,

with the exception of one peridotitic diamond from Koffiefontein, all sulfide inclusions analyzed for Re– Os to date from southern Africa have been eclogitic. All available Kaapvaal inclusion ages obtained prior to 1999 have been summarized recently in Pearson and Shirey (1999) which also contains detailed discussion of inclusion syngenesis and paragenesis.

2.3. Age-dating methods

The dating of diamonds relies on the successful application of radioisotope methods to their mineral inclusions and the indication that the inclusions are syngenetic with respect to diamond crystallization. The question of syngenesis hast been hotly debated over the years (c.f. Pidgeon, 1989; Richardson, 1989; Shimizu and Sobolev, 1995; Navon, 1999; Spetsius et al., 2002) and has recently been reviewed (Pearson and Shirey, 1999; Richardson et al., 2004). The diamond ages used in this paper are based on the conclusion that the dated inclusions are syngenetic. Over the past two decades, ages of diamonds from the major mines have been derived from Sm–Nd and Rb–Sr isotopic studies of silicate inclusions chiefly by Richardson et al. (1984, 1986, 1990, 1993, 1999). Due to the low concentra-

tions of Sm, Nd, Rb, and Sr 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 occasional large grains and the U–Pb isotope system (e.g., Kramers, 1979). But within the last 5 years,



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 differences in crustal lithologies, a change in aeromagnetic fabric (Ayres et al., 1998), and a break between Archean and Proterozoic Re–Os ages on peridotite xenoliths (Carlson et al., 1999; Janney et al., 1999; Irvine et al., 2001). The locations of diamond mines are 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 Bultfontein, De Beers, Dutoitspan, and Wesselton 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 slower mantle; mines located above greenish-blue areas referred to in text as having diamonds derived from seismically slower from Shirey et al. (2002). Diamond genesis, seismic structure, and evolution of the Kaapvaal–Zimbabwe craton. (Science, 297: 1683–1686). Copyright 2002 American Association for the Advancement of Science.

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). Diamond ages used in this study come from this more recent literature and ongoing work (Orapa; Shirey et al., unpublished data; Jwaneng; Richardson et al., 2004). Ar-Ar laser probe age measurements can be obtained on single eclogitic and peridotitic clinopyroxene inclusions (e.g., Burgess et al., 1989, 2004; Phillips et al., 1989). These results are not incorporated in this synthesis however, because complexities due to diffusional Ar loss, excess radiogenic Ar, or radiogenic Ar inhomogeneities (Burgess et al., 1992, 1998) have led in some cases to anomalously young ages.

2.4. Direct determinations on diamonds

The carbon isotopic compositions (δ^{13} C) of diamonds from the major mines in southern Africa have been determined over the last two decades by total combustion of inclusion-bearing diamond chips and analysis of the resultant CO₂ by Deines (1980), Deines et al. (1984, 1987, 1989, 1991a,b, 1993, 1997, 2001), Deines and Harris (1995) and Cartigny (1998) and Cartigny et al. (1998, 1999). Infrared spectroscopic (IR) studies of the abundance and crystal-chemical aggregation of trace nitrogen in the diamond structure were carried out on diamond chips from the same samples that were used for carbon isotopic study and Cartigny also has added N isotopes to the C isotopic studies. 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 would potentially mix together any C isotopic variability associated with diamond growth zonation. Data from these studies have been summarized previously.

3. Results and analyses

3.1. Seismic structure

Fig. 1, showing the tomographic inversion for Pwave 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, therefore, have diamonds that derive from mantle with 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, 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



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., 2004), 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).

with relative P-wave velocity perturbations that vary from +0.084% to +0.357%.

3.2. Diamond ages from inclusions

Data on single eclogitic sulfides from the De Beers Pool, Jwaneng, Koffiefontein, and Orapa form a ¹⁸⁷Re/¹⁸⁸Os vs. ¹⁸⁷Os/¹⁸⁸Os data array (Fig. 2) that clearly includes diamonds of different ages. The data are dominated by the greater number of inclusions analyzed from the De Beers Pool, Jwaneng and Orapa. Those from the De Beers Pool show a 2.9 Ga isochron (Richardson et al., 2001) while the other diamond suites have populations that fall around this age but with more scatter (Fig. 2; Shirey et al., 2001). For Koffiefontein, the first sulfide inclusion suite studied with the Re–Os system, only two inclusions out of the six studied correspond to a circa 2.9 Ga age and the majority give a Proterozoic (1 Ga) age (Pearson et al., 1998). Jwaneng and Orapa each have a number of their analyzed sulfide inclusions which plot at younger ages that match (respectively) the ages obtained from Sm– Nd studies of silicate inclusions. The DeBeers Pool sulfide suite only has one such grain. Thus, in all localities, there appear to be at least two generations of sulfide inclusions: an Archean, circa 2.9 Ga age suite and a Proterozoic suite of an age specific to each locality. The circa 2.9 Ga age of the older suite 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 <u>*Re depletion*</u>' (Trd) approach or 3.1 Ga if using a 'time of <u>mantle reservoir separation</u>' (Tma) approach (Fig. 2; Carlson et al., 1999, 2000; Irvine et al., 2001).

Sm-Nd isochron and mantle model ages for silicate inclusion suites from the Kaapvaal-Zimbabwe craton are summarized in Fig. 3. Peridotitic (harzburgitic) garnets from Finsch and De Beers Pool (Richardson



Fig. 3. Sm–Nd isochron and model ages of silicate inclusions in diamond (sources in Table 1 caption) compared to the ages of major crust forming events in the Kaapvaal craton depicted as thick, grey vertical bands (1) (Schmitz, 2002; Schmitz et al., 2004), peridotite xenolith Re–Os model ages (2) (sources in Fig. 2 caption), diamondiferous eclogite xenoliths (3) (Shirey et al., 2001; Menzies et al., 2003), and sulfide inclusions in diamonds (4) (sources in Fig. 2 caption). Horizontal lines that extend to the Proterozoic are dashed to indicate that younger ages on eclogites and peridotites exist but evidence for specific ages are unclear. The two peridotite ages correspond to the dominant Trd (younger) and Tma (older) ages. 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., nonsinusiodal; Stachel and Harris, 1997; Stachel et al., 1998). Note that depleted mantle growth is shown here schematically only as a straight line. Other, more geologically based models for early Archean depleted mantle evolution will lead to slight differences in depleted mantle model ages from those depicted here.



Fig. 4. (A) Comparison of the carbon isotopic composition of individual diamond analyses grouped according to eclogitic or peridotitic paragenesis. Note that both eclogitic and peridotitic 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 panels A and B occur because a greater proportion of diamonds of eclogitic 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). Copyright 2002 American Association for the Advancement of Science.

et al., 1984) have the oldest model ages yet recorded for inclusions in diamond from southern Africa. Premier peridotitic (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) still would lead to a mid-Archean (3.1 Ga) depleted mantle model age. All other southern African silicate inclusion suites dated with the Sm–Nd system (Richardson, 1986; Richardson et al., 1990, 1999; Smith et al., 1991) are eclogitic and yield Proterozoic isochron or model ages.

Fig. 5. Percentage of Type Ia diamonds (A) and average N abundance in diamonds (B) vs. P-wave velocity anomaly. Error bars have been set at the \pm 5% level for percentage of Type Ia diamonds, \pm 50 ppm for average N abundance, and \pm 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= \diamondsuit , eclogitic= \blacklozenge . Reprinted with permission from Shirey et al. (2002). Diamond genesis, seismic structure, and evolution of the Kaapvaal–Zimbabwe craton. (Science, 297: 1683–1686). Copyright 2002 American Association for the Advancement of Science.

3.3. Diamond isotopic compositions

Diamonds selected for C isotopic and N studies perhaps provide a larger diamond sample than those selected for age studies because they include both the peridotitic and eclogitic parageneses that are typically present at each locality. It has long been known (Sobolev et al., 1979) that diamonds of both peridotitic and eclogitic paragenesis show a prevalent mantle-like carbon isotopic composition ($\delta^{13}C = -3\%$ to -7%) with an isotopically light subpopulation ($\delta^{13}C = -10\%$ to -34%), dominated by diamonds of eclogitic paragenesis (e.g., Gurney, 1989; Galimov, 1991; Kirkley et al., 1991). Compiled data for the Kaapvaal–Zimbabwe cratons (Table 1 and refs. therein, Fig. 4; Shirey et al., 2002, 2003) adhere to this pattern.

Deines et al. (1989, 1993, 1997) previously noted the significantly higher N content of eclogitic diamonds from Finsch, Jwaneng, Orapa, and Premier compared to their peridotitic counterparts from the same kimberlite. Fig. 5 shows that the paragenesis vs. diamond N content relationship extends to all 10 kimberlites with wellstudied diamond populations. The kimberlites whose diamonds are mostly eclogitic have diamonds with an average N content above 290 ppm and a percentage of Type Ia diamonds greater than 80% compared to the kimberlites whose diamonds are mostly peridotitic which are lower in both respects. This N content difference has not resulted in a resolvable difference in the aggregation state of N in the Type Ia diamonds (Shirey et al., 2002, 2003) perhaps because of differences in mantle residence times of eclogitic vs. peridotitic diamonds or because N spectra come from multiple growth zones in rough diamonds or chips. The large range in C isotopic composition and N abundance unfortunately cannot be directly correlated with diamond ages because these diamonds are undated.

4. Discussion and conclusion

4.1. Patterns of cratonic keel structure, diamond age, and paragenesis

New age information from the Re–Os system on mantle lithologies and diamonds allows the relationship among diamond ages, paragenesis, and cratonic seismic structure to be explored. The early work of Holmes and Paneth (1936) demonstrated that eclogite xenoliths could be much older than the kimberlites that carried them. Kramers (1979), using common Pb in sulfide inclusions, and Richardson et al. (1984), using Sm-Nd on harzburgitic garnet inclusions, showed that diamonds are early Proterozoic to mid-Archean. These results indicated that at least parts of the craton were underlain by a mantle keel that could be as old as the oldest crust, a result now generally confirmed by Re-Os model ages on whole rock peridotite xenoliths (e.g., Walker et al., 1989; Carlson et al., 1999, 2000) and in situ Re-Os model ages on sulfides hosted in primary silicates (Griffin et al., 2003a,b). Additional work of Richardson et al. (1986, 1990, 1993, 1999) on diverse eclogitic and lherzolitic silicate inclusions in diamonds produced Proterozoic ages suggesting a progression to more fertile silicate inclusion compositions with time. However, recent Re-Os data show that all four of the eclogitic sulfide inclusion suites studied include late Archean ages that overlap the average late Archean Re-Os mantle model ages on cratonic peridotite (Carlson et al., 1999; Irvine et al., 2001; Shirey et al., 2001). Furthermore, the peridotitic silicate inclusion model ages in the 3.2-3.3 Ga range now appear to overlap some of the oldest crustal U-Pb ages obtained in the western Kaapvaal craton (Moser et al., 2000; Schmitz, 2002; Schmitz et al., 2004) and predate the ages of many other materials in the lithosphere (Fig. 3) as suggested by the Re-Os model ages of whole-rock peridotite xenoliths (Carlson et al., 1999; Irvine et al., 2001) and their sulfides (Griffin et al., 2003a,b), 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; Schmitz et al., 2004).

The distribution of diamond ages and paragenesis with respect to seismic structure reveals that peridotitic silicate (harzburgitic) inclusions in diamonds from the De Beers Pool and Finsch which give the oldest mid-Archean model ages (Richardson et al., 1984), were derived from seismically faster lithospheric mantle. The other peridotitic silicate (lherzolitic) inclusion suite dated (Premier; Table 1) is Proterozoic (Richardson et al., 1993) and was derived from seismically slower mantle. All the other silicate inclusion suites dated so far are eclogitic, Proterozoic (Richardson, 1986; Richardson et al., 1990, 1993, 1999; Smith et al., 1991) and were derived from seismically slower mantle. In comparison, sulfide inclusions from the four localities studied so far are eclogitic, have an Archean age population (Pearson et al., 1998; Richardson et al., 2001; Shirey et al., 2001) and were derived from both fast and slow mantle. Of these four localities, De Beers Pool, whose diamonds were derived from seismically faster mantle, has the lowest number of Proterozoic inclusions.

The distribution of C isotope composition for eclogitic diamonds (Fig. 4A) appears nearly identical to that for diamonds from the seismically slow lithospheric mantle (Fig. 4B) but this is chiefly controlled by paragenesis. There are additional complexities, however. The eclogitic diamonds with δ^{13} C values less than -9% (e.g., those that comprise the isotopically light tail of the δ^{13} C distribution) are dominated by the large number of specimens from Jwaneng, and Orapa (Shirey et al., 2003). Furthermore, Premier, Jagersfontein, and Roberts Victor directly contradict the isotopically light eclogitic diamond to seismically slow lithosphere correlation; Premier lies above seismically slow mantle but has no isotopically light diamonds in its eclogitic population whereas Jagersfontein and Roberts Victor lie above seismically fast mantle but have their eclogitic populations dominated by diamonds with isotopically light carbon (Shirey et al., 2002, 2003). Therefore, while paragenesis is the controlling factor in the differences in C isotopic composition of diamonds from seismically fast vs. slow mantle, the cratonwide distribution of eclogitic diamonds with isotopically light C is not straightforward, perhaps because of the irregular distribution of petrologically diverse eclogite types.

Nitrogen abundance too, shows some correspondence with lithospheric seismic structure that is linked to paragenesis (Fig. 5A and B; Shirey et al., 2002, 2003). Diamonds from Jwaneng, Letlhakane, Orapa, and Premier which are dominantly eclogitic, have a higher percentage of Ia types, an average N content above 290 ppm, and were derived from seismically slower mantle (Fig. 5A and B). Diamonds from Koffiefontein, Finsch, Roberts Victor, Venetia and the De Beers Pool which are dominantly peridotitic, have a lower percentage of Ia types, an average N content below 290 ppm, and were derived from seismically faster lithospheric mantle. The aggregation state of N in the Ia diamonds (not shown) displays no clear systematic variation with lithospheric seismic velocity although De Beers Pool, Finsch and Roberts Victor diamonds from seismically fast mantle do display the lowest percentages of aggregation to B-centers (Shirey et al., 2002, 2003).

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, the variability of eclogitic xenolith suites and the nonoverlapping nature of the extant diamond studies. These complexities are in addition to the episodic nature by which the cratonic keel was generated, stabilized and modified. Table 2 shows the emerging picture from the diamond data of a mantle keel that has retained distinct populations of diamonds that were formed during each of the major tectonothermal episodes to have affected the cratonic keel (e.g., Shirey et al., 2002).

4.2. Age of and compositional constraints on 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 eclogitic (silicate) inclusionbearing diamonds from seismically slow lithosphere (Figs. 1 and 3; Table 1) occur in the same kimberlites that contain Archean eclogitic 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, 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 peridotitic silicate inclusions (lherzolitic garnet and clinopyroxene) that were equilibrated at the 1.9 Ga isochron age representing 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 keel, 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, extending across the northern Kaapvaal craton and to the Table 2

Summary of time periods, inclusions studied and their ages and parageneses, geologic settings proposed for craton formation, and petrologic observations that can be explained by this craton formation model

| Time (Ga) | Age, parageneses, locality | Geologic event | Geologic settings proposed | Observations explained by model |
|-----------|---|---|---|--|
| 3.2-3.7 | 3.2 Ga; peridotitic (hz) silicates; D, F | Creation of earliest cratonic nuclei | Midocean ridges; Oceanic subduction zones | Silicic, differentiated crust, 3.5–3.7 Ga age hz gt inclusions with depletion and enrichment Formation of SCLM at low pressure Wet, pyroxene spinifex komatiites |
| 2.9-3.2 | 2.9 Ga; eclogitic sulfides; D, JW, K, O | Craton assembly into present form | Microcontinent collision; Closing of Archean ocean basins; Marginal subduction | Thermochronology of crustal rocks Geologic pattern of terrane accretion Differentiated crustal rocks (e.g., TTG suites) Widely distributed diamonds with eclogitic sulfides Re-Os ages of peridotites and eclogites Silica (opx) enrichment of the lithosphere |
| <2.1 | 1.9 Ga; peridotitic (lhz) silicates; P 1–2 Ga; eclogitic silicates; F, JW, O, P 1–2 Ga; eclogitic sulfides; JW, K, O | Modification of the craton | Subcratonic magmatism; Marginal subduction | Proterozoic diamond ages Silicate inclusions from both DM and SCLM Seismic structure (tomography) of lithosphere Correspondence of tomography and inclusions Reworked peridotites (Premier, N Lesotho) Bushveld–Molopo Farms Complexes |

Locality letter descriptions follow Fig. 1 (see caption). Acronyms used in observations are as follows: harzburgite (HZ), subcontinental lithospheric mantle (SCLM), tonalite-trondhjemite-granodiorite (TTG), orthopyroxene (opx), depleted mantle (DM), Iherzolite (Ihz).

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 (Carney et al., 1994) is likely to be the surface manifestation of the tectonism that modified the craton on this margin.

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 now 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 that 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 Pwave velocity seen in Fig. 1.

Diamond suites from seismically slower lithosphere have a greater percentage of eclogitic and lherzolitic inclusions which matches regions of the lithosphere that would have a higher proportion of basaltic components. Also, some diamond suites from these regions with eclogitic inclusions have the isotopically light carbon, the highest percentage of Type Ia stones and a higher average N content (>290 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 (Shirey et al., 2002) indicates that diamonds from both types of mantle were stored in the lithosphere at high 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 percentage of IaB diamonds in the De Beers Pool, Finsch, and Roberts Victor diamond populations would be related more to the lower average N content of these diamond populations and their perhaps slower growth than to temperature (Shirey et al., 2002, 2003). Simple heating of the lithosphere in the

seismically slow regions, as might be suggested by the resetting of U–Pb ages in low-closuretemperature 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

Fig. 6. Proposed model for the creation, stabilization and modification the Archean Kaapvaal–Zimbabwe craton by largely subduction-related processes. In the 3.7–3.2 Ga period, oceanic spreading ridges formed on an Earth hotter than today producing a thicker komatilitic to basaltic– komatilitic oceanic crust and a thicker layer of depleted harzburgite. This early crust was recycled by some form of oceanic subduction which allowed the access of water to the melting of the mantle and began the process of crustal differentiation. The products of this period were differentiated cratonic nuclei with abundant sialic component, a distinct upper and lower crust and an attendant cratonic keel. In the period from 3.2 to 2.9 Ga, amalgamation of cratonic nuclei constructed a composite craton with eastern and western domains sutured along the N–S Colesburg Lineament (vertical black line middle right) and other major features similar to the craton as preserved today. Because the closure of ocean basins floored by high-temperature Archean oceanic lithosphere with attendant subduction was the major way to bring cratonic nuclei together, the process led to abundant C, O, H, S fluids for diamond formation and the emplacement of peridotitic and eclogitic protoliths into the lithospheric mantle while preserving age/terrane differences. In the period after 2 Ga, the composite craton was subject chiefly to modification from the margin by subduction and from below by magmatism and metasomatism. This produced multiple generations of new diamonds with different source characteristics and a correspondence between the diamonds and the seismic structure of the lithospheric mantle. Komatile (K), basaltic komatile (BK), harzburgite (HZ), upper crust (UC), lower crust (LC), subcontinental lithospheric mantle (SCLM).

detailed enough to resolve any systematic temperature or depth differences between eclogitic and peridotitic diamonds.

4.3. Model for diamond growth during creation, stabilization, and modification of the craton

A summary of geologic events for the Kaapvaal-Zimbabwe craton and the diamond ages and localities we associate with these events is presented in Table 2 and Fig. 6. Any comprehensive model for diamond formation and craton evolution must explain the ages of diamonds, the distribution of ages relative to paragenesis and craton geology, and the concurrent production of crustal and mantle lithologies. Constraints on lithosphere creation/stabilization come from those diamonds that are Archean: the four suites with eclogitic sulfides that have been dated using the Re-Os system at around 2.9 Ga (Pearson and Shirey, 1999; Richardson et al., 2001, Richardson, Shirey et al., unpublished data) and two suites with peridotitic silicates that have been dated using the Sm-Nd and Rb-Sr 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 depletion caused by 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 (i.e., 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. None of the Archean sulfide inclusions yet analyzed for Re-Os are peridotitic, they are all eclogitic, perhaps due to sampling bias as discussed above. As seen in Figs. 2 and 3, typical sulfide ages do not cluster around the oldest peridotitic 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; Schmitz et al., 2004) but scatter 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 peridotitic, depleted harzburgitic garnets (SmNd, Rb–Sr model ages; Richardson et al., 1984) coexist in the same lithospheric mantle section with 2.9 Ga eclogitic sulfides (Re–Os isochron age; Richardson et al., 2001) that have an elevated, enriched initial Os isotopic composition.

Regarding the reliability of these ages, the point has often been made (e.g., Navon, 1999) that Sm-Nd model ages alone do not preclude the formation of the harzburgitic diamonds at some time later than 3.2 Ga which might allow for coeval crystallization of peridotitic and eclogitic diamonds at 2.9 Ga. This is indeed the case since the Sm-Nd system dates the metasomatism producing the low Sm/Nd in the protolith (3.4-3.5 Ga based on chondritic or depleted mantle precursors respectively; Fig. 3) rather than diamond crystallization (3.2-3.3 Ga based on combined Sm-Nd and Rb-Sr model age arguments). However, comparison of the radiogenic Sr isotope signatures of inclusion (encapsulated) and macrocryst (unencapsulated) garnets from disaggregated harzburgitic diamond host rocks indicates that a 2.9 Ga or younger age for the diamonds is unlikely. The key to this argument is that coupled low Sm/Nd and high Rb/ Sr are typically produced in the same metasomatic event. Yet these harzburgitic garnets have negligible Rb contents and hence very low Rb/Sr ratios. Both inclusion and macrocryst garnets have low Sm/Nd but very low Rb/Sr because garnet includes Sm, Nd, and Sr, but excludes Rb. However, the host rock Rb/Sr remains high beyond diamond crystallization at 3.2-3.3 Ga. Thus, the inclusion and macrocryst garnets inherit their respective Sr isotope signatures by (re)crystallization at 3.2-3.3 Ga and, in the latter case, continuing diffusive exchange with the radiogenic Sr produced in high Rb/Sr harzburgitic diamond host rocks during lithospheric mantle storage prior to sampling by kimberlite. In particular, Kimberley macrocryst garnets have highly radiogenic present-day Sr isotope compositions (⁸⁷Sr/⁸⁶Sr>0.755) as compared to that measured in the corresponding inclusion garnets $({}^{87}\text{Sr}/{}^{86}\text{Sr}=0.706)$ which were isolated from the high Rb/Sr host after diamond crystallization. If the inclusion garnets were only encapsulated in diamond at 2.9 Ga or later, they would be expected to inherit a much more radiogenic Sr isotope composition $(^{87}\text{Sr}/^{86}\text{Sr}>0.710)$ from the high Rb/Sr host than that actually observed (0.706; see Fig. 4 in Richardson et al., 1984).

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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 and 2.9 Ga diamonds with enriched eclogitic sulfide inclusions in the same Kimberley kimberlites indicates that creation and assembly of the craton was a multistage process (Shirey et al., 2002). The Re–Os, Sm–Nd, and Rb–Sr model age relationships indicate a time gap of 300 ± 200 million years between the two Archean diamond formation events.

4.3.1. Craton nuclei creation

Cratonic nuclei initially were created by mantle melting processes that produced severe depletion in the cratonic mantle keel as evidenced by their highly forsteritic olivine (Boyd et al., 1999), high Cr/low Ca garnet (Gurney and Switzer, 1973), and low time averaged Re/Os (Walker et al., 1989). Komatiitic volcanism involving a high degree of mantle melting usually has been implicated in producing this depletion (e.g., Richardson et al., 1984; Walker et al., 1989; Wilson et al., 2003). Classically, komatiite production was thought to involve high degrees of melting above a mantle plume (e.g., Arndt et al., 1997) and in this tectonic setting the cratonic keel could have been produced by vertical underplating of deep mantle (e.g., Haggerty, 1986; Griffin et al., 1999, 2003b), perhaps from transition zone depths. However, more recent experimental and trace element work suggests that such depletion, while still involving high extents of melting, could have occurred at shallower depths (Ringwood, 1977; Canil and Wei, 1992; Stachel et al., 1998; Walter, 1998) perhaps in a hot and wet subduction setting (Parman et al., 2001; Wilson et al., 2003).

A subduction setting for creation of the early cratonic nuclei is advocated (Fig. 6) although its form (if it existed) in the mid-Archean may have been different because plate tectonics then may have been significantly different (e.g., de Wit, 1998). Some form of subduction, nonetheless, is favored over a plume setting for other important reasons (e.g., Table 2). Chief among them is that a subduction setting can produce the ancient rock record of differentiated silicic crust along with the necessary characteristics of the mantle keel. Evolved calc-alkaline plutonic or volcanic rocks such as dacites or members of the tonalite-trondhjemite-granite suite are found as key components of both the eastern (Hunter, 1974; Kroner et al., 1989) and western (Drennan et al., 1990; Anhaeusser and Walraven, 1999; Schmitz, 2002; Schmitz et al., 2004) domains of the Kaapvaal craton and indeed the crust of all of the earliest cratonic nuclei on Earth (Martin, 1993, 1994; Windley, 1995). Furthermore, they occur in most identified oceanic or continental convergent margin settings from the Archean to the present (Barker, 1979; Gill, 1981). The presence of a highly depleted, seismically fast continental keel under the Superior Province (Grand, 1987; van der Lee and Nolet, 1997), a craton formed by arc accretion (Card, 1990) and comprised of more than 50% differentiated crust, demonstrates that Archean subduction is effective at making depleted mantle keels. Indeed, in originally characterizing the properties and evolution of the continental tectosphare, Jordan (1978, 1981, 1988) advocated a subduction model for making the mantle keel that involves depletion in the montle wedge followed by consolidation and thickening during subsequent collisional orogenesis. In addition, a subduction model for early cratonic nuclei provides a mechanism for producing the light REE enrichment juxtaposed onto the oldest harzburgitic garnet inclusions (Richardson et al., 1984). Mantle plumes could succeed as a way to make cratonic nuclei because they make sufficiently buoyant or depleted mantle residua and can even produce some limited amount of rhyolite if hydrothermally altered basalt is buried to wet-melting depths (e.g., Iceland). However, they fail because they do not provide an efficient mechanism to make an extensive (more than 50% of a crustal section) differentiated silicic crust of the type seen in continents.

Crustal differentiation, the production of sial, and the first phase of crustal preservation may have resulted in only partial preservation of the earliest depleted mantle in the lithosphere. This is hypothesized because there is no seismological evidence for extensive ultramafic residues in the lower crust (James and Fouch, 2002) as required by early crustal differentiation (Martin, 1993). The removal of ultramafic residues, if it occurred by delamination, must have occurred at depths shallower than the lithospheric keel and could not have occurred with a stable keel in place. Thus, the restricted distribution of 3.2-3.3 Ga harzburgitic silicate inclusions (documented so far only from the Kimberley-Finsch area) may be explained partially as a preservation feature. It must be noted that the spatial distribution of extensively studied diamond-bearing kimberlites in southern Africa limit the diamond-based constraints on the above model for episodic creation and stabilization of the Kaapvaal-Zimbabwe craton chiefly to its western part. However, mid-Archean Re-Os model and isochron ages on whole-rock peridotites (Pearson, D.G. et al., 2002; Carlson and Moore, 2004), sulfides hosted in silicate mineral concentrates (Griffin et al., 2003a) and komatiites (Wilson et al., 2003) which have a slightly wider geographic distribution than the diamonds presently studied are proposed similarly to reflect the existence of an older depleted cratonic nucleus in the eastern part of the Kaapvaal craton.

4.3.2. Craton amalgamation and stabilization

In the second phase of the envisioned process (Fig. 6b), early cratonic nuclei were accreted along with Archean oceanic lithosphere to create a larger, composite craton similar to that which is preserved today. In this phase, subduction, which included depleted oceanic harzburgite and perhaps the roots of Archean oceanic plateaus, built the rest of the lithospheric mantle (Shirey et al., 2002). The process envisioned is a geologically more complex version of the 'stacked slab' model proposed by Helmstaedt and Schulze (1989). This would involve processes more akin to typical modern subduction (Richardson et al., 2001) where fluid fluxing, melting, and severe depletion of the deeper mantle wedge (Ringwood, 1977; Kesson and Ringwood, 1989; Wilson et al., 2003) could accompany metasomatism and Si enrichment of the shallower mantle already in place as continental lithosphere. Both processes are as much a part of stable lithosphere creation as are the shallower processes of intracrustal melting, metamorphism and compressional tectonics (Schmitz and Bowring, 2001; Schmitz, 2002; Schmitz et al., 2004). A second phase of lithosphere creation (Table 2) could account for the widespread distribution of circa 2.9 Ga age Archean eclogitic 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

diamonds from the same kimberlite, the younger age of these eclogitic diamonds compared to peridotitic (harzburgitic) 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 xenoliths with basaltic komatiitic compositions (Shirey et al., 2001; Menzies et al., 2003). This two-step lithosphere creation model 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 3.20–3.26 Ga and the eastern and western domains (now separated by the N–S Colesberg Lineament) of the craton are sutured together by subduction convergence at 2.88– 2.94 Ga (Schmitz, 2002; Schmitz et al., 2004).

Subduction affecting the western margin of the Kaapvaal-Zimbabwe craton during craton amalgamation is supported by stable isotopic data for Orapa and Jwaneng diamonds. Orapa eclogitic 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 have light 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, intramantle processes (e.g., Cartigny et al., 2001) that changed the 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, 2002). The lack of complete C and N isotopic data sets from all but a few of southern Africa's diamond suites means that it is not possible currently to discuss a cratonwide picture of intramantle isotopic fractionation of C and N in diamonds.

4.3.3. Craton modification

The near one-to-one correspondence of Proterozoic diamond suites having a majority of eclogitic silicate inclusions with seismically slow mantle suggests that

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craton modification and Proterozoic diamond formation were closely linked (Shirey et al., 2002). Proterozoic craton modification that reduced the seismic velocity of the lithosphere did not apparently result in the growth of the lithospheric mantle. This is indicated by a lack of dominant Proterozoic Re-Os model ages on peridotites from the seismically slower portions of the lithosphere. In the Proterozoic silicate inclusion suite, eclogitic inclusions predominate over peridotitic. Thus, it is likely that the modification process was more closely linked to eclogitic and lherzolitic lithologies, perhaps associated with the mafic to ultramafic 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 (Fig. 6). Deep crustal evidence for this thermal event is found in upper U-Pb concordia intercepts of near Bushveld age in garnet-biotite granulite xenoliths from the Orapa kimberlite (Schmitz and Bowring, 2003). 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.

The diamond data do not carry any sign of the young (circa 90 Ma) replacement of the base of the lithosphere below 160-180 km by asthenospheric or metasomatic components as proposed by Griffin et al. (1999, 2002a) from garnet concentrate geothermobarometry and trace element compositions. The chief reason for this is that the nearly all diamonds, being Archean or Proterozoic are too old to carry information on this young event. Furthermore, if the replacement scenario is correct, then most diamonds must originate from depths in the lithosphere shallower than 160-180 km since there is no systematic difference between the diamond content of Group 1 vs. Group 2 kimberlites.

4.4. Secular trends, new south African studies and other cratons

4.4.1. Secular variation of Ni and Re/Os in sulfide inclusions

In the model for craton evolution outlined above, there is a major difference in the composition of mantle melts associated with Archean processes of craton nuclei creation and craton amalgamation vs. the Proterozoic processes of cratonic lithosphere modification. The Archean processes call for highertemperature, komatiite to basaltic komatiite magma production to establish the extreme lithosphere depletion and create the oceanic lithosphere that was consumed during craton amalgamation (Fig. 6). Proterozoic processes call for lower temperature, basaltic magmatism to modify the cratonic lithosphere in the Proterozoic. Thus, the diamond-forming fluids/melts would have a greater chance to be in equilibrium with some ultramafic lithologies in the Archean and mafic lithologies in the Proterozoic. Indeed, acknowledging that silicate inclusion suites plotted in Fig. 1 are not dated and that the dated silicate inclusion suites plotted in Fig. 3 have been subjected to the selection bias introduced by the requirement for datable inclusion suites, the much greater proportion of peridotitic silicate inclusions in the Archean and eclogitic silicate inclusions in the Proterozoic (Fig. 3) fits the pattern expected from the model. Eclogitic sulfide inclusions allow an additional test of these ideas because both Archean and Proterozoic inclusions occur in the same study population at each kimberlite (e.g., De Beers Pool, Jwaneng, Orapa, Koffiefontein), each inclusion can be dated separately, and the Re/Os and bulk Ni contents should vary with ultramafic versus basaltic lithologies.

Although there is overlap between the Archean and Proterozoic eclogitic sulfide inclusion populations, Fig. 7A and B confirms a systematic difference between the two age groups. Archean inclusions cover the whole range seen in Os, Ni, and Re content whereas Proterozoic inclusions are largely confined to the lower Os and Ni contents. This follows the observed pattern of Precambrian magmatism on Earth. Komatiites and basalts are formed in the Archean but by the mid- to late-Proterozoic basaltic magmatism dominates.

4.4.2. Predictions testable with new studies

The correlation of existing diamond age, composition and inclusion paragenesis data with lithospheric seismic structure leads to a set of predictions. These can be applied to future studies of new diamond suites at little-studied kimberlites in areas of the craton, such as the eastern or northern Kaapvaal, that have received

Fig. 7. Re vs. Os for mafic to ultramafic rocks (A) and dated sulfide inclusions (B) grouped according to their Archean or Proterozoic age. Note the lack of low Re/Os sulfide inclusions of Proterozoic age. Panel A is modified from Shirey and Walker (1998) using data from the literature. Data for panel B are from Koffiefontein (Pearson et al., 1998), De Beers Pool (Richardson et al., 2001), Jwaneng (Richardson et al., 2004) and Orapa (Shirey et al., unpublished data).

little previous attention (Table 3). For example, if the western and eastern domains of the Kaapvaal were created separately, the eastern domain, containing older crustal components than the western domain, should have its own clustering of diamond ages that are systematically older than those in the western domain. If diamonds with depleted peridotitic (harzburgitic) inclusions represent portions of the mantle keel that are relict components after protocraton creation, then newly analyzed suites of peridotitic (harzburgitic) inclusions should be 3.2 Ga old or greater and be rare outside of seismically fast lithosphere. If the major process of craton stabilization was the coalescence of earlier protocratons with accompanying consumption of oceanic lithosphere at about

| Table | 3 |
|-------|---|
| 10010 | ~ |

| Predictions | for | future | study | from | new | suites | of | diamonds |
|-------------|-----|--------|-------|------|-----|--------|----|----------|
|-------------|-----|--------|-------|------|-----|--------|----|----------|

| Data and/or | Implication | Test and/or prediction |
|---|---|--|
| observation | | |
| Eastern cratonic domain has older crust and lithosphere dating back as far as 3.5 Ga | Eastern cratonic domain and western cratonic domain were created separately | Eastern domain should have a separate clustering of diamond ages/ parageneses Ages should be systematically older in eastern domain vs. western domain |
| Where dated, peridotitic inclusions with depleted compositions are the oldest and are not widespread | Peridotitic inclusions occur in relict mantle keels left from protocraton creation | New suites of peridotitic, depleted inclusions will be rare out side of fast lithosphere, should be 3.2 Ga or older |
| Eclogitic sulfides inclusions occur in both seismically slow and fast lithosphere and are widely distributed | Assembly by cratonic nuclei accretion made eclogitic diamonds Possible emplacement of eclogite into lithosphere along with subduction | New suites of old (ca. 2.9 Ga) eclogitic diamonds should involve subducted components |
| Silicate inclusion paragenesis and diamond composition (C and N) correspond with lithospheric type Nd isotopic composition of silicate inclusions show asthenospheric and lithospheric sources | Magmatism and metasomatism around 2 Ga changed the lithospheric composition | Diamonds in the seismically slow regions will have greatest diversity of composition |

2.9–3.0 Ga, then additional suites of eclogitic diamonds should be widely distributed in all domains of the craton. Diamond suites as yet comprising small numbers of specimens (e.g., Dokolwayo, Koffiefontein, and Klipspringer) may be part of this late Archean diamond-forming episode or may, with the determination of more precise isochron ages, show fine-scale discrimination that can relate diamond formation to slightly younger geological events such as Ventersdorp magmatism (e.g., Westerlund et al., 2004). If metasomatism of the craton accompanying its modification in the Proterozoic is the cause of the lithospheric seismic structure, then Proterozoic diamonds from the seismically slower part of the lithosphere would typically be expected to have the

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, N, and S isotopic and IR 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 intramantle processes and regional variability on the isotopic composition of diamonds and the ultimate source of diamond-forming fluids.

greatest range in age and composition.

4.4.3. Global patterns in age and paragenesis

There is an interesting parallel to the age distribution of diamonds seen in the Kaapvaal-Zimbabwe craton and other cratons such as the Siberian, Slave and Australian. Presently, other cratons do not have a comparable number of studied diamond or xenolith suites. However, it is significant that those localities where diamond suites have been dated fit the overall pattern shown by the Kaapvaal-Zimbabwe craton. Udachnaya, which is known for its abundant eclogite xenoliths and diamonds with sulfide inclusions, presents the clearest example of this parallel. Peridotitic sulfides from Udachnaya diamonds have been dated with the Re-Os system at 3.2 Ga (Pearson et al., 1999a,b) and sulfide inclusions in olivine in Udachnaya peridotite xenoliths have yielded 3.2 Ga in situ laser ablation ICPMS Re-Os ages (Griffin et al., 2002b; Pearson N.J. et al., 2002). Although no 2.9 Ga diamonds at Udachnaya have been found yet or dated, 2.9 Ga Re-Os whole rock ages have been

| Table 4 | | | | | | |
|---------|---------|----------|--------|------|-----------|---------|
| Summary | of dian | nond age | s from | four | different | cratons |

| Diamond paragenesis, age | Kaapvaal | Siberian | Slave | Australian | |
|---|--------------------------|-----------|-------|------------|--|
| 3.2 Ga, peridotitic (hz) silicate or sulfide inclusions | D, F | Udachnaya | Panda | | |
| 2.9 Ga, eclogitic sulfide inclusions or diamondiferous eclogites | D, JW, KO, N, O, R | Udachnaya | | | |
| 1.9 Ga, peridotitic (lhz) silicate 1-2 Ga, eclogitic silicates 1-2 Ga, eclogitic sulfides | F, JW, KO, O, P | Udachnaya | | Argyle | |

Age references from caption to Table 1 plus additional references for Argyle (Richardson, 1986), Newlands (N; Menzies et al., 2003) Panda (Westerlund et al., 2003b), Premier (Kramers, 1979), Roberts Victor (Shirey et al., 1998, 2001), Udachnaya (Pearson et al., 1995, 1999a,b; Richardson and Harris, 1997). Locality letter designations follow Fig. 1 (see caption).

obtained on diamondiferous Udachnaya eclogite xenoliths (Pearson et al., 1995). If one assumes that the 2.9 Ga diamondiferous eclogites have 2.9 Ga eclogitic diamonds, then this is similar to the distribution of Kaapvaal-Zimbabwe craton Archean diamond ages (Table 4). That is, the Udachnaya peridotitic sulfides match the De Beers Pool and Finsch peridotitic silicates in 3.2 Ga age and depleted harzburgitic paragenesis and the Udachnaya eclogites, presumably dating sulfides in eclogite xenoliths, match eclogitic sulfides from De Beers Pool, Jwaneng, Koffiefontein and Orapa in 2.9 Ga age and paragenesis. Udachnaya even has a Proterozoic, lherzolitic silicate inclusion suite that has negative initial epsilon Nd values yielding an Archean model age (Richardson and Harris, 1997) that is strikingly similar to the lherzolitic silicate inclusion suite from Premier (Richardson et al., 1993). Note, however, that so far only the Kaapvaal cratonic lithosphere records an extended history of Proterozoic diamond formation (Table 4). Both the Slave and Australian cratons have only one diamond suite that has been studied geochronologically but they also roughly fit the Kaapval-Zimbabwe craton age distribution. Peridotitic sulfides in diamonds from the Panda pipe

give a 3.2–3.4 Ga isochron age (Westerlund et al., 2003b) and eclogitic silicate inclusions from Argyle give a mid-Proterozoic age (Richardson, 1986). These similarities in age and paragenesis of diamond suites among cratons that are now widely separated may indicate that diamond formation on the Archean Earth was controlled by global scale events in mantle/crustal evolution and/or that the Kaapvaal/Zimbabwe, Siberian, Slave, and Australian cratons were situated close together in an Archean supercontinent (e.g., Zegers et al., 1998) that may have become disassembled by the mid-Proterozoic.

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