

The significance of mineral inclusions in large diamonds from Yakutia, Russia

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ABSTRACT

Large diamonds (10–200 carats) from Udachnaya, Mir, and Aikhal kimberlite pipes of Yakutia contain variable mineral inclusions typical of diamonds of smaller size. Among these inclusions, extracted from outer portions of these diamonds, the majority are garnets of harzburgitic paragenesis, with a small number of inclusions belonging to lherzolitic and eclogitic groups, in addition to chromite diamond inclusions (DIs). The most striking feature of the garnet DIs is the sinusoidal rare-earth elements (REE) patterns observed for all harzburgitic, and some lherzolitic garnets, whereas eclogitic garnets form normal LREE-depleted patterns. These sinusoidal REE patterns are indicative of a complex petrogenesis for the garnets, involving partial-melting, followed by metasomatic enrichment, particularly of the LREEs/MREEs. The metasomatic agents were probably not those of carbonatitic fluids, but rather C-O-H-N-S fluids, as indicated by the negative anomalies of Ba and Sr in the garnets.

The general axiom among investigators that most, if not all, DIs are syngenetic with their diamonds is seriously questioned. The petrogenetic history of the harzburgitic garnet DIs involved complex processing *prior* to their diamond encapsulation. That is, the garnet DIs retained a signature of their earlier, pre-diamond encapsulation history. This result is taken as proof that basically all harzburgitic garnet DIs are non-syngenetic (i.e., protogenetic) with their host diamonds.

INTRODUCTION

Diamond inclusions (DIs) provide a unique set of mantle data because of their potentially pristine nature, with isolation from *P-T-X* changes subsequent to their encapsulation. One of the most important applications of research on DIs has been to establish the nature of the formation of diamonds, based upon the assumption of *syngenetic* relationship between the inclusions and their host diamonds. A series of investigations (e.g., Sobolev et al. 1998a, 1998b; Taylor et al. 1998, 2000) have revealed extreme chemical variations among inclusions within a single diamond, thereby addressing the co-genesis of the diamonds and their DIs. Thus, the “frozen-in” chemistry of these DIs may record their evolutionary history prior to their moment of “capture” by the diamonds. This concept of syngensis of the diamonds and their DIs is at the heart of determinations of the ages of diamonds, for example, and is seriously questioned in the present study.

Extraordinary large gem-quality diamonds (10–200 carats) from kimberlites of Yakutia are stored in the diamond collections of the Kremlin in Moscow. We were fortunate to be able to examine these diamonds and to obtain samples of DIs from the Udachnaya, Mir, and Aikhal kimberlite pipes. In this report, we present detailed major- and trace-element chemistry of a special set of garnet and chromite DIs from this sampling. This study is directed at the petrogenesis of these DIs, as this

may address the paradigm of syngenetic relationships between DIs and their host diamonds.

ANALYTICAL METHODS

The rarity and high economic values of large diamonds normally limits their study to only nondestructive methods. Therefore, mineral inclusions were extracted only when they were located at or near the edge of a diamond. Of over 2300 large diamonds investigated visually, 56 revealed garnet and chromite inclusions that could be sampled effectively without causing much damage to the host diamonds. A sharp steel needle was used to remove these DIs; unfortunately, observations on the morphologies of the inclusions were not recorded. The sampling procedure and visual estimation of abundance of inclusions, in addition to a preliminary report, were presented by Sobolev et al. (2001). In the present investigation, fragments of 16 garnets and 8 chromites were selected, based mainly on their relatively large sizes ($\geq 100 \mu\text{m}$), for detailed study. The inclusions are mono-mineralic, with no signs of alteration—e.g., chemical zonation, as determined by electron microprobe analyses. We believe that the chemical characteristics of the DIs reported here are those at the instant of the diamond capture and encapsulation—i.e., the DIs are pristine.

Major-element compositions of DIs were determined with a fully automated CAMECA SX-50 electron microprobe (EMP) at the University of Tennessee. The EMP analytical conditions employed an accelerating potential of 15 kV, with 30 nA beam

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current, 5 μm beam size, and standard PAP procedures. Counting times were 20 s for the majority of elements, except for Na and P in garnet (100 s), and V, Ni, and Zn in chromite (60 s).

Trace-element analyses of the diamond inclusions were performed with the modified CAMECA IMS-3f ion microprobe at Washington University. Details of the experimental procedures are described by Zinner and Crozaz (1986a, 1986b), Alexander (1994), Hsu (1995), and Fahey et al. (1987). Detection limits are variable, depending on the element and phase being analyzed, but may be as low as a few ppb in favorable cases.

CHEMISTRY OF DIAMOND INCLUSIONS

Garnet

Most garnet inclusions in the present study contain high MgO contents (19–25 wt%; Table 1), belonging to the p-type or peridotitic paragenesis (e.g., G-10 garnets of Dawson and Stephens 1975). Only two low-MgO (10–12 wt%) inclusions of e-type or eclogitic paragenesis are present, as depicted in Figure 1. Regardless of the sizes of the host diamonds, the compositional ranges of the garnets DIs reported here are similar to those of Siberian garnet DIs from previous reports (Griffin et al. 1993; Sobolev et al. 1997, 1998a, 1998b; Taylor et al. 1996, 2000). In the commonly used plot of Cr_2O_3 vs. CaO contents (Fig. 2 and Sobolev 1977), the high- Cr_2O_3 (>6 wt%) p-type garnets are of harzburgitic paragenesis, whereas the low- Cr_2O_3 garnets (1.4–3.4 wt%) belong to lherzolitic paragenesis. However, Griffin et al. (1999) suggested that the position of the harzburgite-lherzolite transition should be represented by an approximately ~1 wt% broad band, because of its dependency on pressure, temperature, and bulk composition. Based upon this and its REE characteristics, sample Ud-1689 is considered as harzburgitic garnet in this study.

In Figure 3, there are definitive, decreasing trends of Yb, Ho, and Na upon increasing Cr/Al values, taken as a depletion index. The two garnets with the highest Na contents and lowest Cr/Al ratios are from eclogites. The next three lowest Cr/Al values are in garnets from lherzolites. The remaining garnets are harzburgitic. Because Yb, Ho, and Na are less likely to be affected by metasomatism, the trends in the latter two groups appear to indicate a history of partial melting experienced by these garnets. These correlations, however, are not observed in some harzburgitic DIs from the African and Chinese Cratons (Stachel and Harris 1997a, 1997b; Wang et al. 2000, respectively). In contrast, those elements that could be modified easily by metasomatic enrichment, e.g., La and Nd, show crude positive relationships with the Cr/Al ratio. These enrichments are more pronounced in the Chinese and African data. The Sr contents in garnet inclusions from Yakutian diamonds, however, show no obvious correlation with the depletion index.

The harzburgitic garnets exhibit characteristic *sinusoidal chondrite-normalized REE patterns* that peak between Nd and Gd, and reach minima at either Dy or Er, as shown in Figure 4a. Concentrations of REE among inclusions vary considerably, over two orders of magnitude for the LREE (La to Nd), but variations become more limited toward HREE (Ho to Lu). Inclusion Ud-1592 is the most depleted in LREEs, and also

has the lowest FeO content of the garnets (Table 1). Its sinuous REE shape reaches a maximum at Gd.

Both the lherzolitic and eclogitic DI garnets possess “normal” REE patterns that are flat at HREE but decrease sharply toward the LREE, except in the case of SA-21, which shows relatively less LREE depletion compared with other garnets (Fig. 4b). In addition, the eclogitic garnets are generally more enriched in the REEs as compared with the lherzolitic garnets. The REE patterns of these large-diamond garnets are in agreement with those previously reported for garnets included in smaller Yakutian diamonds (Shimizu et al. 1997; Sobolev et al. 1997, 1998a, 1998b; Taylor et al. 2000).

Chromite

With one exception, all the chromite DIs have high- Cr_2O_3 contents in the range ~64 to 68 wt% (Table 2; Figure 5), which is within the field defined by chromite DIs worldwide (Meyer 1987). Furthermore, as shown in Figure 5a, most chromites have TiO_2 contents of <0.7 wt%, further supporting the “diamond indicator range” proposed by Daniels (1995).

In the majority of samples, it is apparent that $Cr\#$ [$100 Cr / (Cr + Al)$] is correlated negatively with $Mg\#$ [$100 Mg / (Mg + Fe^{2+})$], as shown in Figure 5b. Bulanova (1995) observed similar correlations in Yakutian chromite DIs and stated this as a result of magmatic differentiation. Three high- Cr_2O_3 samples have appreciably higher $Mg\#$ (>60) than most samples within the range of other Siberian chromite DIs (Bulanova 1995; Griffin et al. 1993). The well-defined positive trend apparent in the Zr vs. $Cr\#$ plot of Figure 5c may indicate magmatic differentiation, without metasomatic overprinting by Zr-rich fluids.

DISCUSSIONS

Petrogenesis of harzburgitic garnet DIs

The sinusoidal REE patterns of the garnet DIs are the most prominent feature from the large diamonds of this study. In fact, these unusual REE patterns are characteristic of virtually all harzburgitic garnets that occur both as DIs and in xenoliths/megacrysts in kimberlites from cratonic lithospheres worldwide (e.g., Shimizu 1975; Shimizu and Richardson 1987; Stachel et al. 1998). Although the proposed schemes for the formation of these REE patterns vary somewhat, all investigators agree that the sinusoidal-REE-patterned garnets have experienced complex evolutionary histories (e.g., Griffin et al. 1999; Hoal et al. 1994; Stachel and Harris 1997a, 1997b; Stachel et al. 1998). Such garnet genesis involved an initial depletion event (A in Fig. 6), followed by mantle metasomatism (B of Fig. 6), culminating in a possible late-stage depletion episode (C of Fig. 6).

The initial, ancient melting (A of Fig. 6) is evident as the depletion of HREE (Ho-Lu), common in these and other harzburgitic garnet DIs (Fig. 4a). It has been suggested, based on chemical and radiometric data of both DIs and mantle xenoliths from cratonic lithospheres, that this depletion episode corresponds with komatiite magmatism as a major crust-forming event in the Archean (Boyd and Gurney 1986; Boyd et al. 1997; Pearson 1999; Pearson et al. 1995; Richardson et al. 1984, 1993; Snyder et al. 1995; Takahashi 1990). Walker (1998) dem-

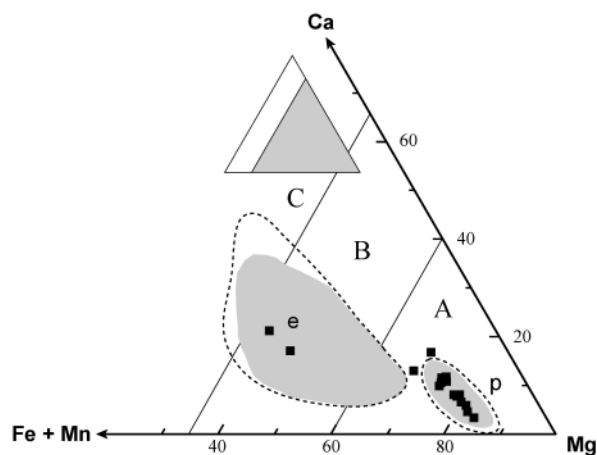


FIGURE 1. Compositions of garnet inclusions in large diamonds from Siberia. Most inclusions fall into peridotitic paragenesis (p-type), with two samples being eclogitic (e-type). Shaded areas represent Siberian DI data from other studies (Griffin et al. 1993; Sobolev et al. 1998a; Taylor et al. 1996, 2000). Dashed envelopes represent worldwide garnet inclusions in diamonds (Sobolev et al. 1998b). The group boundaries of eclogites (A-B-C) are from Coleman et al. (1965).

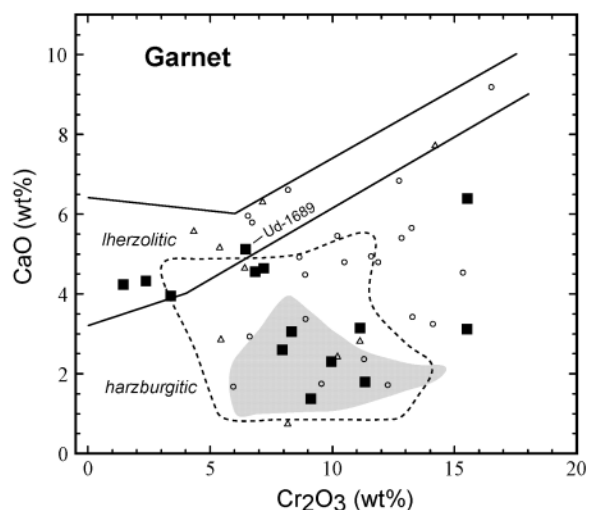


FIGURE 2. Cr_2O_3 -CaO plot of garnet inclusions in large diamonds from Siberia (solid squares). Gray-shaded area and dashed-line envelope represent Siberian garnet DIs from Shimizu et al. (1997) and Griffin et al. (1993), respectively. Garnet DIs from other localities: (1) China (open circles; Wang et al. 2000), (2) Western Africa (open triangles; Stachel and Harris 1997b). Boundaries of different paragenetic types are from Sobolev (1977). Sample Ud-1689, which lies near the boundary, is grouped into harzburgitic type based upon its REE characteristics (see text).

TABLE 1. Major- (wt%) and trace-element (ppm) compositions of garnet inclusions in large diamonds from Yakutia*

Paragen.	Harzburgitic										
	Ud-1592	Ud-2260	Ud-2157	Ud-2237	Ud-1878	Ud-2138	Ud-145	Ud-1689	Ud-2182	Ud-2158	Ud-2178
P ₂ O ₅	0.04	<0.02	<0.02	<0.02	0.03	<0.02	0.03	0.04	0.03	<0.02	0.04
SiO ₂	41.4	41.4	41.1	41.0	41.6	41.3	40.9	40.8	41.2	39.9	40.6
TiO ₂	0.05	0.06	0.10	0.08	<0.03	<0.03	<0.03	0.07	0.11	0.10	0.03
Al ₂ O ₃	16.0	18.0	15.3	18.1	17.8	18.0	16.7	18.2	15.5	11.3	11.8
Cr ₂ O ₃	9.95	7.19	11.1	6.84	8.33	7.96	9.12	6.45	11.3	15.5	15.5
MgO	23.0	21.3	22.4	21.2	22.3	22.8	24.6	19.4	23.2	19.1	21.7
CaO	2.30	4.64	3.14	4.56	3.05	2.60	1.37	5.12	1.79	6.39	3.11
MnO	0.30	0.30	0.29	0.34	0.38	0.33	0.33	0.47	0.31	0.30	0.32
FeO	6.27	6.62	6.29	7.10	6.56	6.53	6.42	9.06	6.42	6.46	6.66
Na ₂ O	0.03	0.02	0.02	0.03	0.02	0.03	<0.02	0.03	0.03	<0.02	0.02
Total	99.40	99.51	99.72	99.24	100.04	99.56	99.47	99.69	99.90	99.20	99.73
Na	204	171	179	219	121	247	41	215	252	86	167
K	16.4	27.3	22.5	8.21	16.9	23.0	13.7	21.6	87.8	9.59	22.8
Sc	169	141	171	138	166	172	214	90	196	163	285
Ti	256	356	512	344	56	75	104	355	542	543	202
Rb	0.32	0.28	0.30	0.21	0.19	0.31	0.41	0.45	0.25	0.36	0.44
Sr	0.09	0.12	0.15	0.28	0.97	0.81	2.08	0.71	1.42	1.40	1.71
Y	0.58	1.16	1.70	1.16	0.70	0.54	0.35	2.60	1.75	0.97	1.54
Zr	11.4	6.30	24.1	5.93	0.77	0.91	2.91	18.2	16.8	18.5	13.7
Nb	0.29	0.73	0.81	0.57	0.33	0.21	0.27	0.64	0.63	2.70	2.02
Ba	0.064	0.035	0.073	0.021	0.066	0.076	0.87	0.090	0.15	0.029	0.052
La	0.001	0.004	0.006	0.008	0.010	0.016	0.036	0.069	0.067	0.080	0.33
Ce	0.017	0.19	0.11	0.22	0.19	0.23	0.57	0.84	0.79	1.56	2.75
Pr	0.010	0.14	0.079	0.19	0.082	0.075	0.25	0.29	0.33	0.61	0.70
Nd	0.10	1.34	0.95	1.31	0.46	0.48	1.13	1.90	3.02	3.88	4.66
Sm	0.24	0.31	0.81	0.28	0.082	0.10	0.14	0.67	0.89	1.27	1.61
Eu	0.13	0.078	0.19	0.081	0.014	0.024	0.030	0.18	0.18	0.30	0.42
Gd	0.63	0.30	0.75	0.29	0.093	0.086	0.064	0.35	0.55	0.86	1.04
Tb	0.062	0.031	0.096	0.045	0.010	0.012	0.009	0.059	0.067	0.067	0.076
Dy	0.16	0.22	0.43	0.18	0.055	0.055	0.029	0.45	0.27	0.24	0.31
Ho	0.021	0.046	0.065	0.036	0.019	0.014	0.008	0.090	0.056	0.028	0.038
Er	0.048	0.15	0.16	0.11	0.12	0.084	0.066	0.21	0.16	0.056	0.12
Tm	0.012	0.040	0.033	0.023	0.025	0.029	0.021	0.029	0.020	0.009	0.017
Yb	0.16	0.33	0.17	0.29	0.36	0.32	0.26	0.29	0.15	0.075	0.13
Lu	0.039	0.10	0.052	0.091	0.11	0.10	0.095	0.041	0.042	0.019	0.039

*Detection limits are 0.02 wt% for P₂O₅ and Na₂O, and 0.03 wt% for other major oxides.

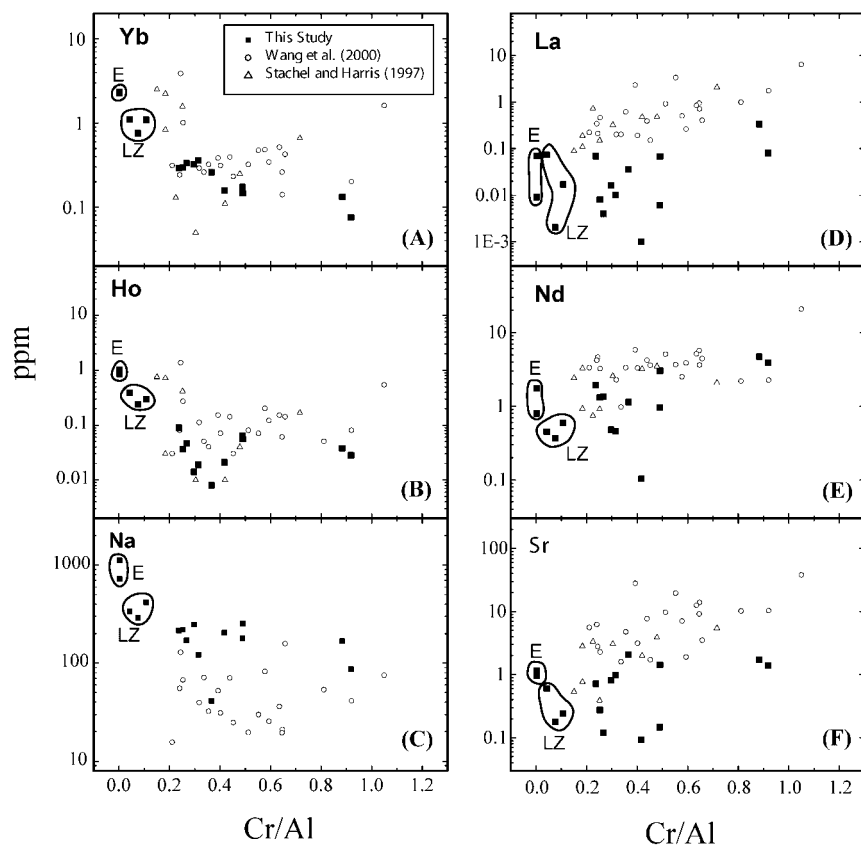


FIGURE 3. Plots of Cr/Al vs. selected trace-element concentrations of garnet DIIs. Negative correlations observed in Yb, Ho, Na (a–c) suggest a depletion event. Positive correlations observed in La (d) and Nd (e) may indicate an enrichment that is more intense in depleted samples. No clear correlation is displayed by Sr (f). The two eclogites and three lherzolites are labeled in the plots; all other samples are harzburgites.

TABLE 1—Extended

	Lherzolithic			Eclogitic	
	Mr-38	Mr-1	SA-21	Ud-2045	SA-1
0.05	0.04	0.03	0.08	0.09	
42.2	41.9	42.2	39.8	39.6	
0.29	0.28	0.36	0.43	0.21	
21.1	21.1	22.5	22.1	22.0	
2.37	3.37	1.44	0.08	0.08	
21.8	21.3	21.6	12.1	10.5	
4.33	3.95	4.23	6.44	7.97	
0.33	0.35	0.31	0.32	0.37	
6.94	7.69	7.54	18.4	19.1	
0.04	0.05	0.04	0.14	0.10	
99.45	100.03	100.25	99.99	99.98	
290	417	335	1115	725	
16.3	21.5	20.7	11.4	21.3	
91	94	86	49	50	
1329	1291	1569	2039	1018	
0.26	0.31	0.14	0.58	0.37	
0.18	0.24	0.60	0.94	1.15	
8.17	11.2	10.6	30.3	32.5	
30.2	13.1	38.8	17.3	24.9	
0.26	0.24	0.17	0.039	0.14	
0.099	0.097	0.13	0.032	0.18	
0.002	0.017	0.073	0.009	0.070	
0.041	0.18	0.38	0.13	0.63	
0.023	0.066	0.079	0.055	0.23	
0.37	0.59	0.45	0.79	1.74	
0.51	0.40	0.31	0.81	1.42	
0.20	0.14	0.14	0.46	0.76	
0.83	0.81	0.74	2.12	2.77	
0.16	0.21	0.18	0.49	0.65	
1.10	1.44	1.46	3.92	5.04	
0.24	0.30	0.38	0.86	1.01	
0.80	1.12	1.19	2.77	2.74	
0.11	0.18	0.16	0.39	0.36	
0.75	1.09	1.10	2.31	2.25	
0.13	0.28	0.24	0.41	0.30	

onstrated experimentally that peridotite from the Siberian craton can be generated as a residue of komatiite melt extraction at ~6 GPa and 40% melting. Stachel et al. (1998), however, argued against such high-pressure komatiite magmatism based upon the high-Cr contents in garnets that could only be produced from a protolith of spinel-lherzolite composition that experienced partial melting at low pressure. In fact, the requirement of low-pressure melting could be achieved when a komatiite-generating plume reached the low-pressure regimes before being recycled back into the mantle through Archean or pre-Archean subduction zones (Walker 1998). Wang (1998) also favored plume activity in the Archean to explain the coexistence of both peridotitic and eclogitic inclusions within the same diamond.

That it was metasomatism (B in Fig. 6) that modified the initially depleted REE patterns in harzburgitic, and certain lherzolithic, garnets is agreed upon by all. However, the details of the process (es) to form the MREE (B) and LREE (C in Fig. 5) portions of the sinusoidal shapes in response to metasomatic enrichment are understood only qualitatively. Griffin et al. (1999) presented an interpretation of these REE patterns from garnets of harzburgite xenoliths from the Wesselton kimberlite of South Africa. They suggested that the enrichment in MREEs reflected the REE patterns of metasomatic melts (e.g., carbonatites), whereas the low LREEs were due to their progressively misfit in the low-Ca garnet structure. This model, however, probably is not applicable to the garnet DIIs of the present study due to the lack of a clear positive correlation of

CaO contents and the LREEs, thereby not supporting carbonatitic metasomatism. Trace-element signatures do not support this.

The formation of the sinuous REE patterns for harzburgitic garnets has been suggested to be the result of some sort of disequilibrium metasomatic processes (Hoal et al. 1994; Shimizu et al. 1997). However, this explanation is not considered to be valid; the observation of such REE patterns in harzburgitic garnets worldwide, both as DIs and as xenoliths, argues against such “consistent” disequilibrium. In the disequilibrium scenario, the LREEs and MREEs in garnets initially mimic the steep negative slope of metasomatic fluids (B of Fig. 6), probably due to the large difference between the LREE contents in the fluid and garnet. The high levels of LREEs, which deviate most from the restraints of the garnet crystal-chemistry, subsequently re-equilibrate, thereby forming a positive slope (C of Fig. 6). Although this interpretation is consistent with the large variation for the LREEs, along with limited variation for the HREEs in harzburgitic DI garnets in our study, the kinetics of “re-equilibration” (i.e., diffusion rates) of the REEs are such that they cannot account for the sinusoidal patterns (Van Orman et al. 2002). In fact, if such extensive re-equilibration did occur, the garnet REE patterns would become “normal” with all-positive slopes.

At present, we do not have a satisfactory explanation for the LREE portion (C of Fig. 6) of the harzburgitic garnet REE patterns. Such typical positive slopes of REEs with negative-sloped MREE patterns are usually interpreted as the result of a depletion episode, such as one that accompanies a partial-melting event. If this is the case here, it can be said that the event that last occurred before encapsulation of the garnets in the diamonds was this partial melting. This melting would appear to remove the earlier metasomatism from the diamond formation, which occurred only afterward.

Although there may be alternate interpretations for attaining the final shape of these sinusoidal REE patterns, it can be

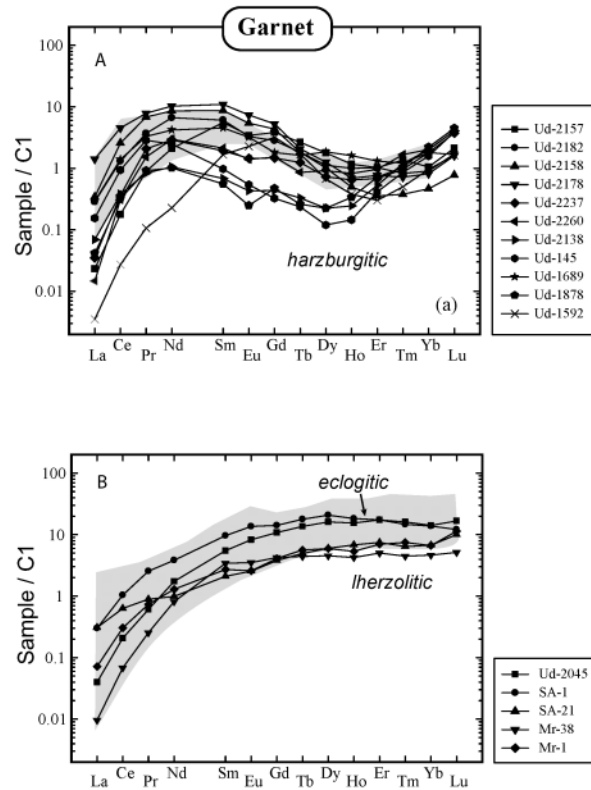


FIGURE 4. Chondrite-normalized (e.g., Anders and Grevesse 1989) REE patterns of garnet inclusions in large diamonds from Siberia. (a) Sinusoidal REE patterns of harzburgitic garnets are probably due to an initial partial melting followed by LREE-enriched metasomatism with a subsequent LREE-depletion event. (b) “Normal” LREE-depleted patterns shown by lherzolitic and eclogitic garnets. Shaded areas in Figures 4a and 4b are data of Siberian garnet diamond inclusions from Shimizu et al. (1997) and Taylor et al. (1996, 2000), respectively.

TABLE 2. Major- (wt%) and trace-element (ppm) compositions of chromite inclusions in large diamonds from Yakutia

Sample:	Ud-2043	Ud-1995	Ud-100	Ud-105	Ud-187	Ud-1696	Ud-1848	M-2-1	M-97
SiO ₂	0.06	<0.03	0.09	<0.03	0.06	0.04	0.05	0.04	0.05
TiO ₂	0.42	0.18	0.04	0.03	0.78	0.03	0.52	0.07	1.21
Al ₂ O ₃	4.00	1.96	6.22	2.82	4.13	5.47	4.63	5.62	17.4
V ₂ O ₃	0.23	0.17	0.30	0.18	0.24	0.33	0.29	0.33	0.31
Cr ₂ O ₃	65.9	68.2	64.2	67.6	64.0	63.9	64.5	63.8	46.5
MgO	12.7	8.80	12.4	9.80	10.8	11.0	12.2	10.8	14.2
MnO	0.17	0.23	0.20	0.21	0.21	0.22	0.21	0.18	0.16
FeO	15.5	19.3	15.8	18.3	18.9	17.8	16.0	18.1	18.9
NiO	0.12	0.06	0.09	0.06	0.13	0.08	0.11	0.09	0.17
ZnO	0.03	0.09	0.04	0.05	0.06	0.07	0.07	0.11	0.10
Total	99.15	98.91	99.32	99.09	99.32	98.92	98.51	99.12	98.85
Na	40.0	85.8	30.5	110	47.3	50.0	74.1	23.7	418
Si	174	137	295	201	186	202	1443	125	7521
P	2.24	4.74	9.32	1.66	3.44	10.7	32.4	3.30	228
K	8.39	20.0	31.1	8.62	16.6	52.1	62.4	18.0	451
Ca	17.1	56.4	72.2	43.1	22.1	76.9	307	23.4	4923
Sc	3.79	1.43	1.88	1.07	2.09	1.35	2.31	1.42	4.29
Co	254	389	308	356	345	359	317	345	392
Ni	530	330	491	284	637	401	504	420	1048
Cu	102	87.0	56.6	35.1	212	104	207	44.2	1193
Rb	0.13	0.076	0.12	0.046	0.14	0.14	0.11	0.11	0.47
Sr	0.073	0.40	0.50	0.24	0.19	0.35	0.64	0.17	10.4
Y	0.018	0.012	0.017	0.008	0.012	0.017	0.034	0.011	0.46
Zr	1.92	4.12	0.81	2.28	1.56	0.98	3.66	0.79	12.7
Nb	n.a.*	n.a.	2.11	1.18	6.16	1.86	2.92	0.94	1.20

* Not analyzed.

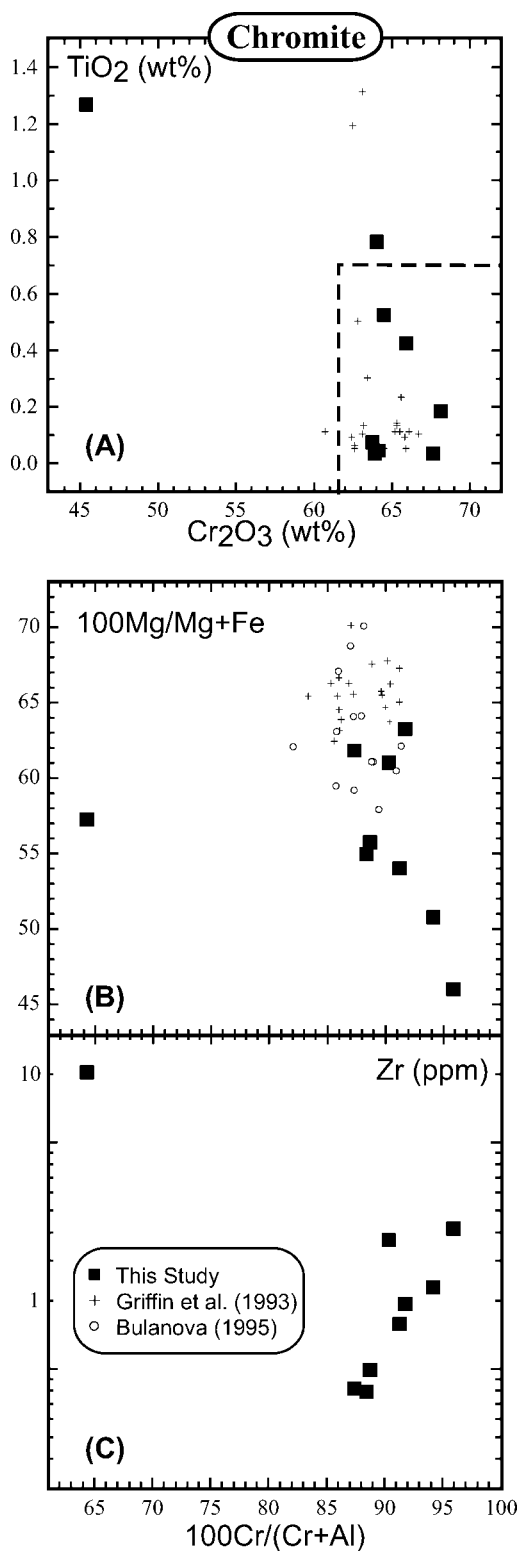


FIGURE 5. Compositions of chromites in large diamonds from Yakutia, compared with literature data from the same region (Bulanova 1995; Griffin et al. 1992). Dashed lines in (a) represent the “diamond inclusion field” (Daniels 1995), where high Cr_2O_3 contents of chromites are used as “indicator minerals” in diamond exploration.

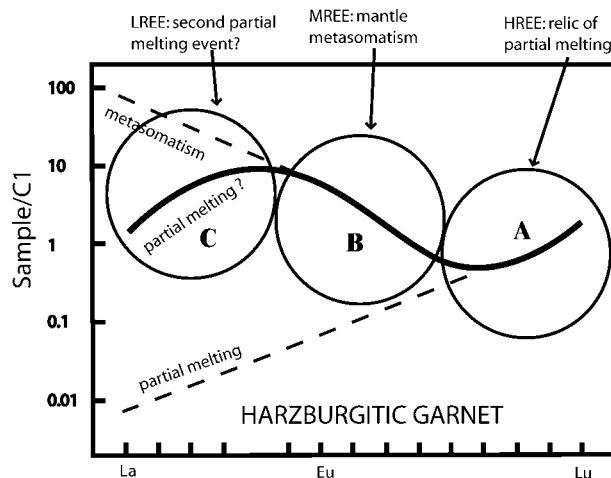


FIGURE 6. Diagram illustrating schematic interpretation of sinusoidal-shaped REE pattern, typical in harzburgitic-garnet diamond inclusions. The REE pattern suggests multiple processes that begin with a partial melting event (A), followed by metasomatic enrichment (B), and with a possible late-stage episode of partial melting (C).

safely assumed that harzburgitic garnets as DIs have experienced a complex genesis entirely prior to their encapsulation in their host diamonds. An increasing amount of evidence has been reported for unusual effects of metasomatism, particularly for eclogitic DIs, which often possess tremendous chemical and/or isotopic heterogeneity (e.g., Otter and Gurney 1989; Ruzicka et al. 1998; Sobolev et al. 1998a; Spetsius and Taylor 2002; Stachel et al. 1998; Taylor et al. 2000; Wang and Gasparik 2001).

Nature of the metasomatic fluids

The chemistry of the metasomatic agents, that modified the chemistry of DIs and possibly precipitated the diamonds can only be constrained qualitatively. The depletion of high-field strength elements (HFSE), such as Ti, Zr, and Hf (not shown) observed in harzburgitic garnets of the present study, rules out silicate melts as the metasomatic agent, as they are enriched in these elements. Carbonatitic melts, typically low in HFSEs, have been commonly proposed as the metasomatizing media (Griffin et al. 1993, 1999; Wang and Gasparik 2001; Wang et al. 2000; Taylor et al., 2003b). However, the scarcity of carbonate minerals (magnesite, calcite) as inclusions in diamonds seems to weaken this possibility, as well as depletion in Ba and Sr (Stachel et al. 1998). Although a recent investigation by Israeli et al. (2001) indicates the presence of carbonates along with silicates and brine as inclusions in cloudy diamonds from Koffiefontein, South Africa, these inclusion assemblages only occur in unusual cloudy and possibly fibrous-coated diamonds, probably resulting from late-stage kimberlitic formation. In addition, the reaction of garnets with carbonatitic melts would probably result in an enrichment of certain incompatible elements, such as Ba and Sr. However, pronounced depletions of these elements are evident in the trace-element diagrams for the present garnet DIs (Fig. 7).

It appears that garnet inclusions from the large diamonds of Yakutia were not subjected to any appreciable carbonatitic metasomatism. Indeed, the enrichment signature for these gar-

net DIs is more typical of mantle metasomatism (e.g., Kopylova et al. 1997; Stachel and Harris 1997b; Stachel et al. 1998; Taylor et al. 1998), entirely pre-dating diamond formation. It is also probable that the fluids that facilitated the actual diamond growth were C-O-H-N-S rich (e.g., Stachel et al. 1998; Taylor et al. 2000).

Nor have the diamonds themselves experienced any carbonatic fluids. McCammon et al. (2001) recently reported on the survival of diamonds during metasomatism and suggested that increasing carbonate activities would enhance resorption of the diamonds, even at low oxygen fugacities. These extraordinary large diamonds have resorption-free surfaces—indeed, they are perfect octahedra and of gem quality. This feature indicates little to no contact with corrosive, carbonatic fluids, both during and after diamond formation.

Syngenetic diamond inclusions

Virtually all papers on diamond inclusions start with the major premise that the diamonds and their inclusions are *syn-*

genetic—i.e., formed simultaneously under the same *P-T-X* conditions and by the same chemical/physical process. Yet, supporting evidence for the assumption of syngenesity usually is omitted (e.g., Griffin et al. 1993; Richardson and Harris 1997; Stachel et al. 1998; Taylor et al. 1996; Wang and Gasparik 2001; Wang et al. 2000). However, such criteria for the identification of syngenetic inclusions have been discussed by Harris (1968) and Sobolev (1977), for example. The most commonly used criterion is the imposition of the morphology of the host diamond on the inclusion (Meyer 1987). However, such morphologies are not always present or apparent. Indeed, crystallographic data, even if available, may not be unequivocal for syngenetic growth of garnets and diamonds (Taylor et al. 2003a). It is imperative, therefore, to investigate the syngenetic theory based on the chemistry of the garnet DIs from which we address the paradigm of syngenesity. Inherent here is *the requirement for co-crystallization of diamonds with their varieties of mineral inclusions.*

Recent studies by several authors (e.g., Harte et al. 1999; Sobolev et al. 1998a; Spetsius et al. 2002; Taylor et al. 1998) have questioned the significance of chemical heterogeneity of multiple inclusions in diamonds, as these relate to a common heritage for the host diamonds and their inclusions. For example, Sobolev et al. (1998a, 1998b) reported 35 garnets and 5 clinopyroxenes within a single diamond from the Mir pipe of Yakutia. Their major- and trace-element chemistry covers almost the entire compositional range of that obtained from mantle eclogites from all of Yakutia. Such *extreme chemical variations* in the eclogitic DIs of one diamond cannot be adequately explained by any syngenesity scenario with diamond crystallization. Sobolev et al. (1998a) proposed a model wherein metasomatic fronts, whose compositions vary drastically over time, precipitate diamonds either as rims on pre-existing “seed” crystals or as separate crystals. These pre-existing seed minerals, therefore, are more appropriately considered as *non-syngenetic* diamond inclusions. A recent study by Spetsius et al. (2002) of sulfide inclusions in zircon has been extended to suggest that diamonds may also capture and preserve older sulfides during their growth or regrowth in mantle rocks, and the inclusions therefore do not necessarily date the formation of the diamond, i.e., these inclusions are *non-syngenetic* with respect to the host diamond.

The compositions of the harzburgitic and lherzolitic garnet DIs in the present study are a case for this protogenetic (i.e., non-syngenetic) origin. The complicated genesis and evolution of the garnet DIs, which ultimately resulted in the sinusoidal REE patterns described above, are impossible to explain within the context of a syngenetic relationship between the inclusions and their encapsulating diamonds. This is because such REE patterns would require a depletion event followed by metasomatic enrichment of the garnets, and a depletion of LREEs. The unique REE patterns of the harzburgitic garnet DIs has been described by numerous authors (Griffin et al. 1999; Hoal et al. 1994; Stachel and Harris 1997a, 1997b; Stachel et al. 1998). An important implication from the complex petrogenetic history of the garnet DIs is the requirement that the garnets underwent extensive processing *prior* to diamond encapsulation—i.e., proof of the non-syngenetic formation of diamond and their DIs. Granted, a scenario can be imagined

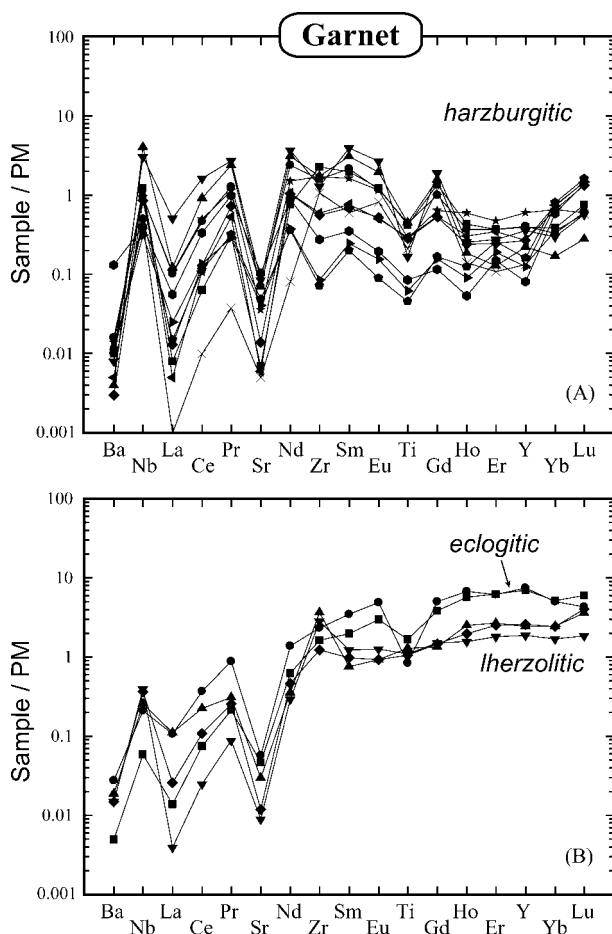


FIGURE 7. Trace-element patterns, normalized to primitive mantle, of garnet inclusions in large diamonds from Siberia. Negative anomalies for Ba, Sr, Ti, and Zr observed in harzburgitic garnet DIs suggest C-O-H-N-S fluids, rather than carbonatic fluids, as the metasomatic agents. Lherzolitic garnet DIs show no depletion of Zr and Ti, which may indicate an influence of a silicate melt. Symbols are as in Figure 4.

whereby the garnets have been metasomatized by the same fluids that precipitated the diamonds. However, the entire chemistry of the garnets was not modified at that time, making for “new” garnets, especially because they retained a signature of the early partial-melting event. Furthermore, there would appear to have been a late-stage depletion event (partial melting?) preceding the capture of the garnets by the diamond. Additional evidence has been presented by Harte et al. (1999), in a study of DIs from São Luiz, Brazil, that supports our contention that DIs predate the formation of host diamonds. In summation, the general axiom in the diamond community that DIs are syngenetic with their diamonds is seriously questioned. This is a subject that merits careful consideration for the study of diamonds and their mineral inclusions.

CONCLUDING REMARKS

In this scenario, the already-grown and evolved garnets were accidentally encapsulated by the diamonds from the surrounding environment. The nature of the sinusoidal-REE patterns for virtually all harzburgitic DI garnets world-wide is confirming evidence that these garnets and their host diamonds were not syngenetic. In fact, the garnet and diamond may have had entirely different geneses. Such non-syngenetic relationships can have important and highly significant ramifications for interpretations of the *P-T-X* conditions and age of diamond formation. The chemistry of the harzburgitic garnets is very revealing of this non-syngeneses, and we insist that the general axiom of syngeneses of diamonds and their DIs must be seriously considered for all such mineral inclusion studies.

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