SHORT COMMUNICATIONS

Chloride–Carbonate Nodules in Kimberlites from the Udachnaya Pipe: Alternative Approach to the Evolution of Kimberlite Magmas

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Group-I kimberlites are the crystallization products of low-degree partial melts derived from the convective mantle [1, 2]. Since kimberlites are the main economic source of diamonds, the evolution of primary kimberlite melts is of great importance for deciphering mantle compositions, melting conditions, and diamond genesis.

Most kimberlites consist of a brecciated mixture of olivine phenocrysts and xenogenic mantle and crustal material embedded in a fine- or coarse-grained groundmass, which is dominated by serpentine, calcite, and, occasionally, dolomite [3–5]. Thus, the evolutionary trend of kimberlite magmas is controlled by Mg silicates (high contents of MgO and SiO_2) and carbonates (high contents of CaO and CO_2). However, this trend can be intensely affected by interactions during ascent, the assimilation of mantle and crustal xenoliths, and the syn- and postmagmatic alteration of most kimberlite rocks. Almost all studied kimberlites around the world, even aphanitic [6, 7] or "uncontaminated" or "fresh" ones, were variably affected by these processes [7–10].

In the studying kimberlites, special attention was given to one aspect of the evolution of kimberlite magmas in mantle and crustal conditions: the entrapment of country rocks and interaction with them [3, 11]. Nodules in kimberlites (peridotites, eclogites, metamorphic and sedimentary rocks, as well as diamondiferous rocks) are usually used to estimate physicochemical and structural conditions in the subcontinental lithosphere, the properties of kimberlite magma, and the crystallization conditions of diamonds. It is convenient to study these processes in the Udachnaya kimberlite pipe in Yakutia because of a large number, diversity, and remarkable freshness of its nodules [11, 12].

Our study was focused on a new (carbonate–chloride) type of nodules recently found in kimberlites at deep levels (>350 km) of the open-pit mine at the Udachnaya–Vostochnaya pipes. Host kimberlites are also unique in chemical and mineral composition. The groundmass is predominantly olivine–carbonate–chloride in composition and shows almost no evidence of replacement [13, 14]. The presence of such nodules in unaltered kimberlites seems not to be coincidental and provides new insight into the evolution of kimberlite magmas.

KIMBERLITES OF THE UDACHNAYA–VOSTOCHNAYA PIPES

The studied kimberlites of the Udachnaya–Vostochnaya pipe (Daldyn field, Yakutia, 347 Ma) have mineralogic–petrographic, geochemical, and Sr–Nd–Pb isotopic ($^{87}Sr/^{86}Sr \approx 0.7047$; $\varepsilon Nd \approx +4$) characteristics [13, 14] typical of Group-I kimberlites [1, 2]. However, unlike most other kimberlites from elsewhere worldwide, the rocks of the Udachnaya–Vostochnaya pipe from a depth of 350 m are remarkably fresh, in particular, are free of primary/secondary serpentine and other hydrous phases, with the only exception of accessory phlogopite in the groundmass. They show extremely low water contents (<0.5 wt %) [13] and high contents of alkali elements (4.3–5.7 wt % Na + K) and Cl (2.3–3.2 wt %). Such an enrichment is caused by the presence of halite NaCl, sylvite KCl, Na–K–Ca carbonates (shortite $Na_2Ca_2(CO_3)_3$ and zemkorite $(Na, K)_2Ca(CO_3)_2$ [15, 16], as well as accessory K–Na sulfates, sodalite $Na_8Al_6Si_6O_{24}Cl$, djerfisherite $K_6(Fe,Cu,Ni)_{25}S_{26}Cl)$ and rasvumite KFe_2S_3 [13, 17–20].

CARBONATE–CHLORIDE NODULES

A remarkable feature of deep-seated (400–500 m) kimberlites from the Udachnaya pipe is the presence of rounded or angular nodules (5–30 cm), which consist
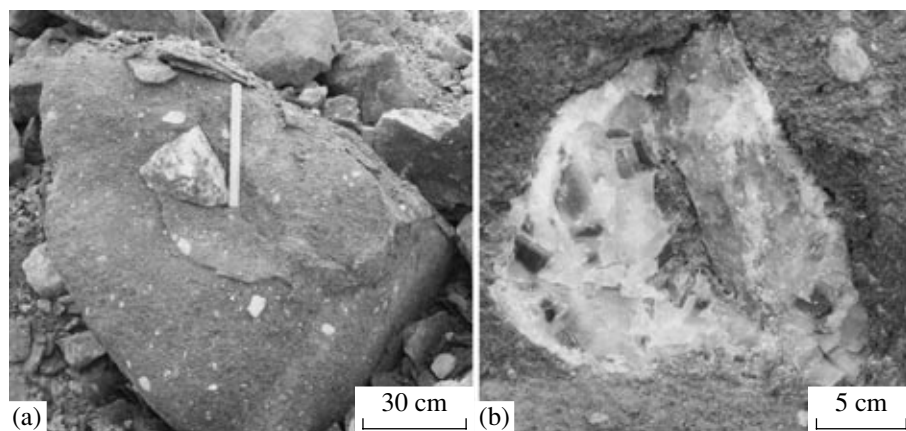
Fig. 1. Chloride nodule in kimberlite from the Udachnaya–Vostochnaya pipe (sample UV-1-05, prior to and after recovery).
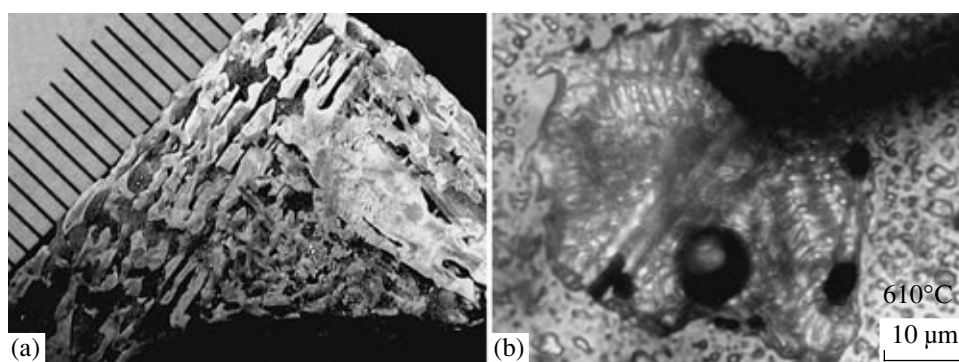

Fig. 2. Typical immiscibility textures in (a) the chloride–carbonate nodule UV-5a-03 and (b) the melt inclusion in groundmass olivine from the unaltered kimberlite of the Udachnaya–Vostochnaya pipe, which were formed owing to exsolution into alkaline–carbonate and alkaline–chloride liquids.

of variable proportions of chlorides and carbonates (Figs. 1, 2a). Nodules are in sharp contact with the kimberlites and display no evidence of thermal alterations (Fig. 1). The contact zone, up to 1 mm thick, is made up of a breccia-like rock consisting of olivine, calcite, sodalite, phlogopite–tetraferriphlogopite, humite–clinohumite, Fe–Mg carbonates, perovskite, apatite, magnetite, djerfisherite, and alkali sulfates in a chloride matrix.

The structure of carbonate–chloride nodules is defined by the intercalation of systematically oriented carbonate and chloride layers (Fig. 2a). The layers of oriented carbonates (2–2.5 cm) are isosceles triangles in cross sections or skeletal three-dimensional rhombohedra. The individual carbonate layers typically have a rough to pitted surface, but, in places, smooth surface. They often give boudin-like lenses up to 1–1.5 mm across, with symmetrical zoning in swells. Chloride forms sugar-like aggregates that fill interstices between carbonate layers and fissures in carbonates. Carbonate layers in individual nodules strongly differ in phase composition. They consist of either shortite–northupite–calcite assemblages (sample UV-2-03) or zoned

aggregates of K-rich nyererite in the core and shortite in the rim (samples UV-1-03 and UV-5a-03).

The cores of zoned carbonate layers (Figs. 3a, 3b, 3c, samples UV-1-03 and UV-5a-03) are made up of nyererite strongly varying in K_2O and SO_3 contents (up to 12.7 and 13.3 wt %, respectively, table). Carbonates of the same composition were previously found in the groundmass of host kimberlites and in melt inclusions in olivine [13, 20]. The strong correlation between K_2O and SO_3 ($r^2 = 0.94$) suggests that the nyererite contains submicron grains of aphthitalite, a K–Na sulfate. This is confirmed by the presence of aphthitalite peaks in the XRD patterns of this nyererite. Aphthitalite occasionally forms larger (up to 20–50 μm) inclusions that are oriented parallel to the elongation of the host nyerereite and therefore can be readily identified optically. The porous rim of zoned carbonate layers consists of predominant shortite, which sharply differs from nyererite in the absence of K and S (table, Figs. 3a, 3b). However, in this case, aphthitalite $K_3Na(SO_4)_2$ increases in amount and forms larger grains both in shortite and at the contact of shortite with chloride (Fig. 3c). This indicates that the earlier S-rich alkaline carbonates were
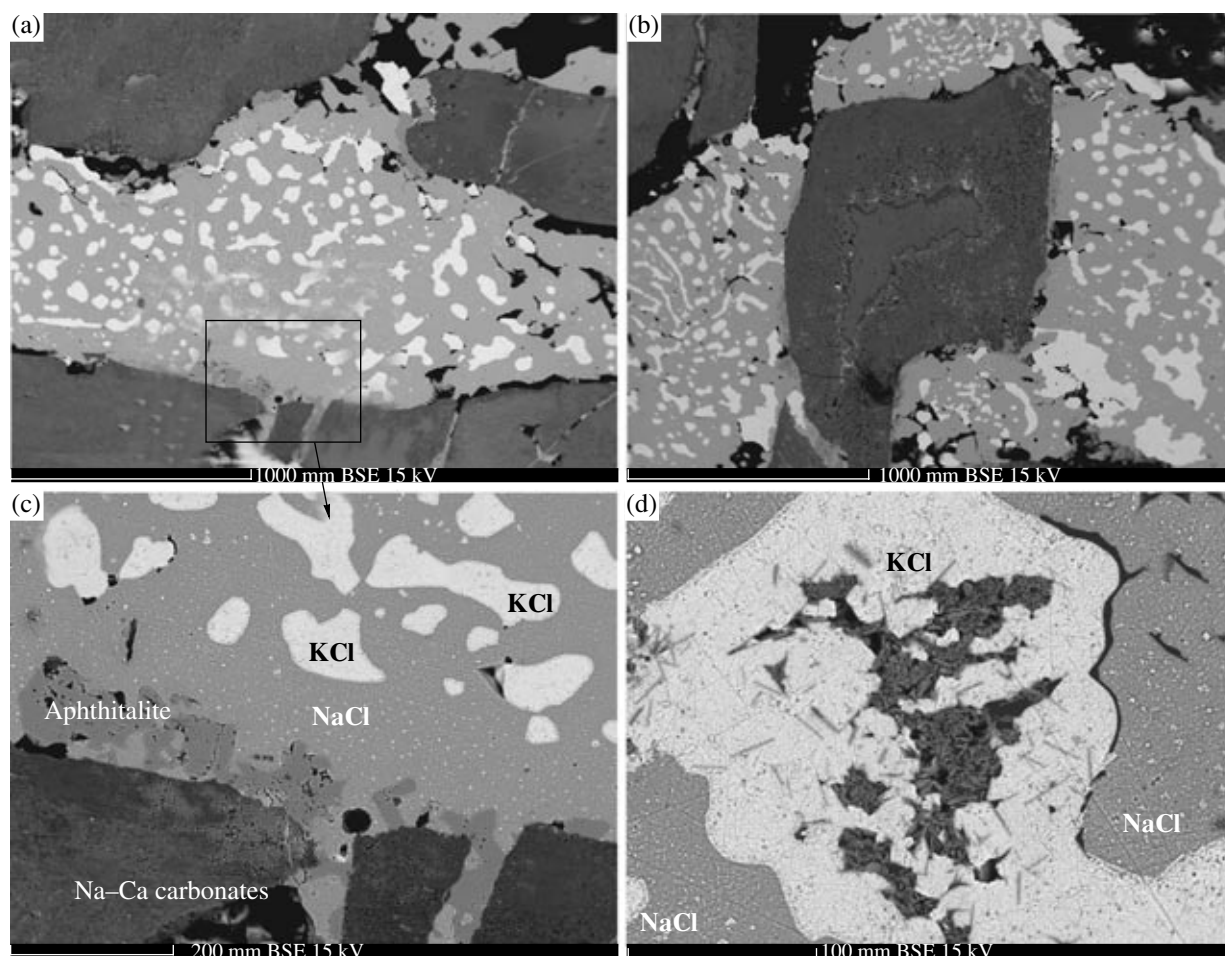
Fig. 3. BSE images of polished chloride–carbonate nodule UV-5a-03 from the Udachnaya–Vostochnaya pipe. (a, b) Relations between alkali–carbonate layers (dark gray) and halite–sylvite intergrowths (light gray and white, respectively). Carbonates demonstrate chemical heterogeneity: lighter nyererite (table, an. 3–4) and darker shortite (table, an. 5). The morphology and distribution of sylvite in halite imply the unmixing of the chloride liquid; (c) aphthitalite (table, an. 7), located at the contact between carbonates and chlorides (magnified fragment in Fig. 3a); (d) equant sylvite in halite with a druse filled with fine-grained phlogopite. This suggests the presence of aluminosilicate in the residual chloride–carbonate melt.

decomposed to form Na–Ca carbonates and oriented aphthitalite grains. The chloride component is mainly halite, whereas sylvite usually forms equant or ameboidal droplets in halite. Such textures (Figs. 3a, 3b, 3c) resemble melt immiscibility textures rather than exsolution lamellae.

Northupite $Na_3Mg(CO_3)_2Cl$, calcite, and various Ba–Sr–Ca carbonates usually occur as subordinate phases among carbonates. The carbonates (mainly, shortite, sample UV-2-03) occasionally contain a great number of equant inclusions of K–Na and Ba–Sr sulfates, apatite, phlogopite–tetraferriphlogopite, and sulfides. The chloride constituent often contains aluminosilicate (fine-grained phlogopite) (Fig. 3d), as well as potassium sulfides, rasvumite, and djerfisherite [19].

DISCUSSION

Since chlorides, alkali sulfates, and, to some extent, alkali carbonates are water-soluble phases, their presence in the groundmass of unaltered kimberlites of the Udachnaya–Vostochnaya pipe is not typical of magmatic rocks. The active Oldoinyo Lengai natrocarbonatite volcano in Tanzania is the only example where the same unusual minerals crystallized from melts and precipitated from fumarole gases [21–23]. However, chlorides, sulfates, and carbonates formed at Oldoinyo Lengai are hygroscopic minerals and are not preserved for a long time. They disappear within a few hours or days after their formation owing to dissolution in meteoric waters, and their preservation in geological history seems to be highly improbable [24]. The presence of these ephemeral minerals in the kimberlites of the Udachnaya–Vostochnaya pipe could be explained by two mechanisms: (1) precipitation from modern plat-

Chemical composition of kimberlite groundmass, chloride–carbonate nodule, and nodule minerals

Component	1	2	3	4	5	6	7	8	9	10
SiO_2	26.12	1.32	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
TiO_2	1.25	0.05	–	–	–	–	–	–	–	–
Al_2O_3	1.99	0.09	–	–	–	–	–	0.00	0.00	0.00
FeO_t	7.24	0.37	0.00	0.00	0.00	0.00	0.02	0.00	0.00	0.00
MgO	28.93	2.47	0.00	0.00	0.00	0.03	15.83	0.01	0.19	0.03
CaO	12.38	31.9	31.68	28.75	37.16	0.07	0.30	35.89	56.02	0.04
Na_2O	4.25	18.87	17.75	17.26	19.44	10.33	36.91	20.04	0.00	10.15
K_2O	2.23	2.41	5.19	11.43	0.09	41.24	0.62	0.32	0.00	41.45
P_2O_5	0.44	0.23	0.06	0.22	0.02	0.01	0.00	0.00	0.00	–
SO_3	0.68	1.45	5.39	9.32	0.02	48.31	0.06	0.00	0.00	48.31
Cl	2.84	4.32	0.36	0.66	0.00	0.01	14.26	–	–	–
SrO	0.10	0.25	0.70	0.60	0.35	0.01	0.04	0.45	0.47	0.04
BaO	0.15	0.05	0.14	0.11	0.05	0.04	0.08	0.01	0.00	0.03
CO_2	10.27	35.27	–	–	–	–	–	–	–	–
H_2O	0.51	0.10	–	–	–	–	–	–	–	–
Total	99.38	99.15	61.26	68.36	57.15	100.05	68.12	56.70	56.68	100.05

Note: (1) Groundmass of unaltered kimberlite from the Udachnaya-Vostochnaya pipe ($n = 3$); (2) chloride–carbonate nodule, sample UV-2-03; (3–5) zoned carbonate aggregate (Fig. 3b, sample UV-5a-03): (3, 4) core and rim of nyererite; (5) shortite from rim; (6) aphthitalite at the carbonate–chloride contact (Fig. 3c); (7–10) minerals from nodule, sample UV-2-03: (7) northupite; (8) shortite; (9) calcite; (10) aphthitalite. FeO_t all iron as FeO; dash means not analyzed. Rocks and minerals were analyzed on a Phillips 1400 by RFA and on a CamecaSX100 microprobe, respectively, at the Tasmanian University. CO_2 and H_2O (Fig. 4) were analyzed on a Carlo Erba CHNS-O.

formal brines present in the quarry at the pipe; (2) the complete absence of postmagmatic "drainage" of kimberlite rocks by aqueous fluids (in the case of magmatic origin).

There are a few lines of evidence that the mineral assemblage of kimberlites in the Udachnaya–Vostochnaya pipe, which is not typical of magmatic rocks and is unstable in subsurface conditions, is of primary magmatic origin and has not experienced any significant alterations since the pipe formation (~350 Ma). First, the Sr, Nd, and Pb systematics of the kimberlites proper, as well as the water soluble groundmass minerals, salt aggregates, and brines from the quarry at this pipe, indicate that the system was closed [14]. The brines are enriched in Sr but have a much more radiogenic $^{87}Sr/^{86}Sr$ composition (0.70885–0.70897) than the kimberlites (0.7047) [14]. It should be noted that no mineralogical data are available as yet for the crystallization of alkali carbonates from platformal brines in the kimberlites from the Udachnaya pipe or other Siberian pipes. Second, a strong gas flow (hydrogen and methane, 10^5 m^3/day) with a pressure of 50–70 atm was detected when this unaltered kimberlite block was penetrated by boreholes [25]. Third, the kimberlites have a massive structure, with no visible fissures and deformations, while petrographic observations testify to a close paragenetic assemblage of chlorides, sulfates, and alkali carbonates with groundmass minerals (olivine,

phlogopite, calcite) [13]. A very important fact is that serpentine, a typical mineral of kimberlites around the world, was not found in the kimberlites of the Udachnaya–Vostochnaya pipe, indicating an extremely low water content in the rocks (table).

The above data indicate that the rocks in question were not affected by postmagmatic alterations. This is independently supported by the similar chemical and mineral compositions of carbonate–chloride nodules and melt inclusions in the groundmass olivine [13, 20]. The melt inclusions are homogenized at temperatures of 660–760°C, and these temperatures can be used to estimate the entrapment temperatures of olivine inclusions. On cooling, the melt inclusions remain essentially liquid until 600°C, when chloride-rich and carbonate-rich liquids spontaneously unmix. The spatial distribution of the two immiscible phases that occurred during unmixing (Fig. 2b) closely resembles the structural features of our chloride–carbonate nodules (Fig. 2a). This indicates that the nodules were formed from melt droplets that were rich in alkali carbonate and chloride components but depleted in the aluminosilicate component.

The Udachnaya–Vostochnaya kimberlite groundmass (table) is abundant in the same carbonate–chloride component. Hence, the nodules of relevant composition were formed by the segregation of a carbonate–chloride melt from the almost solidified kimberlite at
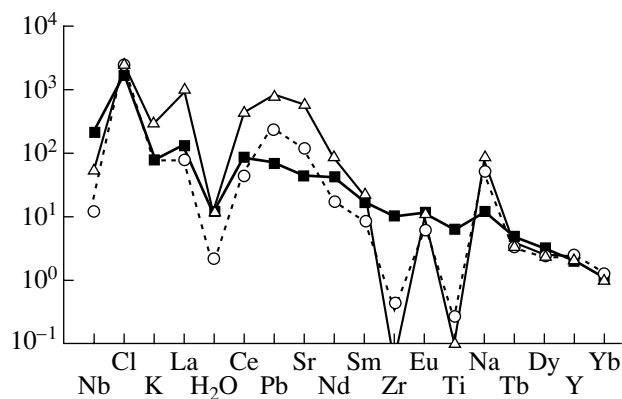
Fig. 4. Pyrolite-normalized [35] compositions of the Udachnaya–Vostochnaya kimberlite groundmass (squares, *n* = 3), chloride–carbonate nodule UV-2-03 from kimberlite (circle), and natrocarbonatite from Oldoinyo Lengai (triangles, samples OL102 and OL105) [36]. The normalizing value of 420 ppm for H_2O is based on $H_2O/Ce = 250$.

temperatures higher than the immiscibility point (~600°C) but no lower than the entrapment temperature of melt inclusions in the olivine (660–760°C).

The presence of primary magmatic alkali carbonates in other kimberlites [26, 27] and geochemical similarity between the Udachnaya–Vostochnaya kimberlites and Oldoinyo Lengai natrocarbonatites (Fig. 4) serve as additional evidence for the magmatic origin of the studied carbonate–chloride nodules. Available data testify to the magmatic origin of natrocarbonatites from Oldoinyo Lengai [28], but the origin of alkali carbonate and haloid components in them (and, by analogy, in kimberlites) remains controversial. For example, a possible source of these minerals at Oldoinyo Lengai could be the surrounding water-bearing sedimentary rocks completely assimilated during magma ascent [29]. By analogy, the parental kimberlite melt of the Udachnaya pipe could also assimilate evaporite rocks when passing through the sedimentary cover of the Siberian platform.

This assimilation model has not been confirmed as of yet, because no evaporites were found in the platformal cover during its deep drilling (to 700–1700 m) in the vicinity of the Udachnaya pipe. At the same time, chloride minerals were found in the Monahans chondrite (1998), which was studied immediately after its fall [30], as well as, together with carbonates, in microinclusions of a fluid and a melt in kimberlite diamonds [31–34]. This gives grounds to suggest the mantle origin of the carbonate–chloride component in the Udachnaya–Vostochnaya kimberlites. The studied kimberlites are strongly enriched in alkalis as compared to other kimberlites (Fig. 4). This is caused not only by their alteration but primarily by the derivation of kimberlite melt by means of low-degree melting. Thus, the primary kimberlite magma initially had high contents of alkalis, chlorine, and carbon. An increase in contents of these elements during olivine crystallization resulted

in a carbonate- and chloride-rich composition of the residual kimberlite melt during the final stages.

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