Geochemistry of Calcareous–Silicate Rocks of the Kharagol Formation, Southern Baikal Region

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Abstract—Complete geochemical characteristics have been found for calcareous–silicate rocks of the Kharagol Formation that serves as a reference unit within the Khamardaban polymetamorphic composite terrain (southern folded framing of the Siberian Platform, Baikal–Khubsugul region). We studied the high-grade (granulite) domain known as Slyudyanka crystalline complex of the southern Baikal region. The Kharagol Formation is composed of different diopside, scapolite–diopside, and wollastonite–scapolite–diopside gneisses and crystalline schists with a variable calcite content. Petrogeochemical data suggest that their protoliths were tuffoids with different amounts of calcareous material. The volcanoclastic component of tuffoids is closest to the Andean-type calc-alkaline andesites. The Kharagol paleobasin was probably located at the active continental margin. Within the studied metamorphic sequence, the Kharagol Formation marks a sharp change in sedimentation conditions and may be considered a boundary of subterrains with protoliths formed in different paleogeodynamic settings. Geochemical indicators of calcareous–silicate rocks of the Kharagol Formation in different metamorphic zones may be high Cr, Ni, and Ba contents.

INTRODUCTION

The Kharagol Formation is one of the metamorphic sequences in the southern folded framing of the Siberian Platform. It is widespread in the Baikal-Khubsugul region as a member of the Khamardaban composite terrain (Vasilyev et al., 1998). The Kharagol Formation mainly consists of calcareous-aluminosilicate rocks. It is composed of well recognized specific green diopside gneisses and schists in moderate- to high-temperature metamorphic zones. However, the Kharagol formation is difficult to recognize in low-temperature zones, where calcareous-silicate rocks are composed of micaceous-carbonate, carbonate-actinolite, and other schists. It extends over more than 200 km from the southern coast of Lake Baikal along the southern wall of the Tunkin Valley to the northern termination of Lake Khubsugul. Its counterparts are more or less reliably distinguished in the central Khamar Daban and eastern Khubsugul areas, on the left bank of the Irkuta River (upstream from the Settlement of Monda), possibly in the Tunka Ridge, and some other areas of the region (Shafeev, 1970; Evolyutsiya..., 1988; Boos, 1991; Vasil'ev et al., 1999; and others).

The Kharagol Formation occupies a specific position in the most comprehensively studied part of the southern Baikal region (Slyudyanka Complex). Metamorphic sequences in this area are divided into the lower Slyudyanka and upper Khangarul groups by a discordant boundary interpreted as discordance or unconformity (Vasil'ev et al., 1981a). However, tectonic contact along premetamorphic overthrust cannot be ruled out. The rock groups strongly differ in composition and structure. The Slyudyanka Group is primarily composed of mafic crystalline schists developed after low-K tholeiite basalts. The Kharagol Formation is located at the base of the Khangarul Group and its calcareous-silicate rocks are considered terrigenouschemogenic-volcanogenic products. Volcanogenic material of these rocks corresponds to calc-alkaline andesites and basaltic andesites with K specifics. The Kharagol Formation is replaced upsection by the Bezymyannyi Formation representing, the major part of the Khangarul Group, and mainly composed of terrigenous rocks. In the framework of geosynclinal paradigm, the above facts were interpreted as a consequence of the eugeosynclinal evolution from the initial magmatic stage to the mature flyschoid stage. The protolith age was considered Late Archean for the Slyudyanka Group and Early Proterozoic for the Khangarul Group. The age of granulite metamorphism, which affected both groups, was also considered Early Proterozoic (Vasil'ev et al., 1981a).

Thus, the Kharagol Formation has the significance of an indicator in the geology of the region. On the one hand, it serves as a regional marker (stratigraphic link) of a large polymetamorphic terrain. On the other hand, it records a sharp change in sedimentation settings,

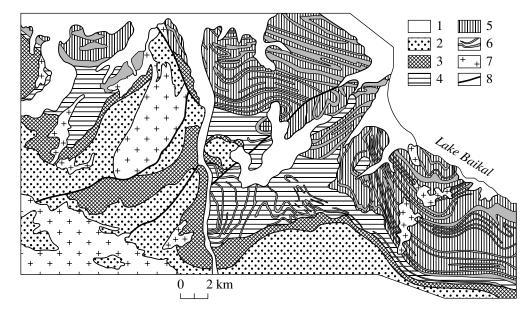


Fig. 1. Schematic geological map of the Slyudyanka Complex (Vasil'ev *et al.*, 1981a). (1) Cenozoic sediments and basalts; (2, 3) Khangarul Group: (2) Bezymyannyi Formation, (3) Kharagol Formation; Slyudyanka Group: (4) Pereval Formation, (5) Kultuk Formation, (6) mafic crystalline schists (metabasalts) in the Slyudyanka Group; (7) granites and syenites; (8) faults.

which was either sequential or nearly coeval (at tectonic boundary) in laterally separated paleobasins. In any case, it may be considered an inferred boundary of subterrains.

Our investigation was aimed at a detailed geochemical study of calcareous-silicate rocks of the Kharagol formation in order to reconstruct the most probable nature and accumulation setting of protolith and to decipher indicator geochemical features for the reliable identification of this formation in different-temperature metamorphic zones and stratigraphic correlations. The test site for our investigations was the high-grade (granulite) area traditionally distinguished as the Slyudyanka metamorphic complex (Fig. 1). Since the Kharagol Formation within this complex hosts stratiform wollastonite deposits, we also attempted to decipher specific features of the protolith that could serve as the most appropriate source for metamorphic wollastonite ores.

BRIEF CHARACTERISTICS OF THE KHARAGOL FORMATION IN THE SLYUDYANKA COMPLEX

The Kharagol Formation occupies a significant portion (1200 m) in the summary stratigraphic succession of the Slyudyanka Complex and consists of the following units: metaterrigenous rocks (biotite gneisses) 10%, carbonates (marbles and calciphyres) 15%, and calcalkaline rocks (diopside gneisses and crystalline schists) 75%. In individual sections, the thickness is less and rock proportions strongly differ owing to facies variability or tectonic boudinage. Some sections contain more than 90% calcareous–silicate rocks. Metaterrigenous rocks are most typical of the lower subformation, whereas carbonate rocks are mainly confined to the upper, more calcareous subformation. The upper subformation also contains horizons and beds of specific manganese rocks (gondites and kodurites) (Vasil'ev *et al.*, 1981a; Koneva, 1998). Gneisses and schists alternating with them are enriched in Mn. All significant wollastonite occurrences are also contained in the upper subformation.

Calcareous-silicate rocks have a polymineral composition. Clinopyroxene is ubiquitous and varies from Fe-rich diopside to salite. Other rock-forming minerals are quartz, plagioclase, microcline, calcic scapolite, calcite, and wollastonite. Ugrandite garnets, biotite, hornblende (more often, diaphthoritic), and epidoteclinozoisite minerals (retrometamorphic) occur in subordinate amounts. Titanite is a common accessory mineral. In terms of parageneses and proportions of the rock-forming minerals, one can recognize numerous major (petrotypes) and transitional rock varieties. They have laminated and banded structures defined by variations in the content of melanocratic laminae and calcite admixture. The lamina thickness ranges from a few millimeters to a few centimeters. No massive rocks were found. Calcareous-silicate and carbonate rocks show both sharp and gradual (schist or gneiss-calcareous schist-calciphyre-marble) transitions.

COMPILATION OF REPRESENTATIVE SAMPLES AND ANALYTICAL METHODS

The Kharagol Formation was systematically sampled during the detailed geological mapping along several sections extending from the Utulik–Solzan River in the east to the Bol'shoi and Malyi Tibel'ti rivers and Elovskii spur (Tunkin Valley) in the west, i.e., over 70 km along the strike. Most part of the wollastonitebearing samples was taken from minings carried out during the exploration of ore deposits and occurrences (Malobystrinsk, Tultui, and others).

When preparing and systematizing the analytical sample set from a large collection (hundreds of samples), we met with methodological problem of the rock type. The mineral assemblages are mainly related to decarbonatization, which leads to loss of CO₂ and proportional increase of other components in the metamorphic rocks relative to protoliths. The effect of decarbonatization can be readily demonstrated for wollastonite formed by the following reaction: calcite + quartz =wollastonite + CO₂. If the content of newly formed wollastonite is 10 wt %, the amount of CO₂ loss will be 3.8 wt %. Let us suppose that the rock contains 30%wollastonite, while the protolith contained 50 wt % SiO₂ and 20 wt % CaO. Then, the metamorphic rock will contain 56.4% SiO₂ and 22.6% CaO due to the loss of 11.4% CO₂. Generally, it is impossible to quantitatively estimate the degree of decarbonatization. Moreover, if calcite is absent, one cannot reliably judge about its presence in the primary sediment. This statement is particularly valid for parageneses with wollastonite and (or) scapolite formed by reaction between calcite and plagioclase. The ubiquitous clinopyroxene forms by different ways, including the optional sequential dehydration and decarbonatization reactions: Chl + $Calc \longrightarrow Amph + Pl + H_2O + CO_2; Bi + Calc +$ $Qtz \longrightarrow Amph + H_2O + CO_2; Amph + Calc + Qtz \longrightarrow$ $Px + H_2O + CO_2$, and so on (*Evolyutsiya*..., 1988).

One can suggest that primary CaCO₃ content in sediments and the degree of their decarbonatization during transition into gneisses must increase in the following paragenetic rock sequence: diopside rock ---- (diopside + scapolite) rock \rightarrow (diopside + wollastonite + scapolite) rock (scapolite occurs in all wollastonitebearing rocks). Based on this assumption, all gneisses and crystalline schists were subdivided into three groups: (1) diopside gneisses and crystalline schists, (2) scapolite-diopside rocks, and (3) wollastonite-scapolite-diopside rocks. In order to increase the homogeneity, we included into the groups only rocks with the calcite content of no more than 10%. However, several rocks transitional to calciphyres (up to 30% calcite) were additionally analyzed in each group to estimate the geochemical variations of protoliths with increasing carbonate constituent.

Manganese rocks described in (Koneva *et al.*, 1998) were omitted from the sampling. The MnO content accepted was less than 1 wt % for groups 1 and 2 and no more than 1.5 wt % for wollastonite rocks, because they are enriched in Mn. We also omitted subordinate rocks that are atypical of the Kharagol Formation, such as the extremely rare amphibole–pyroxene schists. The more detailed metabasites subdivision of each group, for example, into gneisses (presence of quartz and plagioclase as major minerals) and crystalline schists, was

not justified, because the subgroups (from three to seven in each group) turned out to be nonequivalent. They strongly overlapped in chemical composition, although the mineral composition was often different.

As a result, the sampling contained 17–20 samples from each group. The sampling reflects close proportions of groups 1 and 2 in the sequence. The abundance of wollastonite rocks in the sequence is significantly lower. For statistical purposes, the share of wollastonite rocks in the sampling was taken as equal to that of other groups. The geochemical significance of wollastonite rocks as mineral resource was also taken into consideration.

Major elements were determined by the classical wet chemistry and XRF methods. In the latter case, contents of volatiles, FeO, and Fe_2O_3 were analyzed by chemical analysis. Trace elements were measured by the optical spectroscopy, XRF, and atomic adsorption methods. Replicate measurements of some trace elements by different methods showed a good convergence of the results. Rare earth elements were analyzed by ICP-MS on a Plasma Quad PQ2 spectrometer using international standards. All analyses were performed in laboratories of the Vinogradov Institute of Geochemistry and Analytical Chemistry and the Institute of the Earth's Crust.

RESULTS

The average contents and variation ranges of major and trace elements are listed in Tables 1 and 2. Because of different degrees of decarbonatization, the ratios of elements are more informative than their absolute values. The decarbonatization is less prominent in the distribution of trace elements because of their low contents and significantly higher dispersion. However, the general chemical characteristics of rocks may be obtained by the comparison with model continental crust (CC) composition close to andesite. Major elements show a subhorizontal pattern (Fig. 2a), indicating their distribution similar to that in CC. However, the concentrations are significantly lower due to high Ca contents. The groups are distinctly distinguished in terms of Ca anomaly (Table 1), thus confirming the validity of their classification based on the primary CaCO₃ content in protoliths and degree of decarbonatization. The Mn anomaly is distinctly expressed only for wollastonitebearing rocks. The trace element distribution is more complex (Fig. 2b). The groups strongly differ in terms of elements characterized by high concentrations, like Cr (up to 680 ppm), Ni (up to 330 ppm), Ba (up to 4400 ppm) and Sr (up to 1900 ppm). In particular, groups 1 and 2 have positive Cr and Ni anomalies. Group 3 has the highest Ba.

Relative to other subgroups, more calcareous subgroups (1a, 2a, and 3a; Tables 1, 2) are depleted in the major components (except CaO and CO_2) and most trace elements. The silicate constituent of protolith

Components	Rock groups									
and modules	1 (16)	1a (3)	2 (16)	2a (3)	3 (19)	3a (9)				
SiO ₂	60.30	51.82	57.42	50.83	56.10	46.86				
	53.8-68.7	42.9–56.6	48.7–68.4	43.04–58.2	47.9–66.8	38.4-60.48				
TiO ₂	0.54	0.41	0.57	0.43	0.37	0.23				
	0.3–0.72	0.4–0.41	0.2–0.75	0.33-0.55	0.3–0.618	0.08-0.38				
Al_2O_3	12.80	11.62	12.10	10.62	9.1	6.88				
	10.1–15.5	10.2–12.4	9.47–15.4	9.06-13.57	3.21–12.28	3.1–11.4				
Fe ₂ O ₃	1.43	0.93	1.36	0.84	0.91	0.71				
	0.8-4.91	0.72-1.31	0.43–3.2	0.63-1.13	0.35–1.7	0.13–2.09				
FeO	4.03	2.99	3.58	2.76	3.05	1.71				
	1.75–5.45	2.8–3.28	0.22-4.48	2.49-3.00	1.35-5.06	0.75–2.8				
MnO	0.13	0.10	0.20	0.29	0.48	0.46				
	0.08–0.3	0.09-0.15	0.07-0.91	0.10-0.63	0.06-1.36	0.07–1.39				
MgO	3.77	2.58	4.50	3.68	2.95	2.54				
-	1.05-7.9	1.78–3.34	2.35-8.87	2.9-4.4	1.36–4.37	1.63-4.22				
CaO	10.10	16.55	14.63	17.72	21.00	30.27				
	5.2-15.6	12.3-23.0	4.8-20.05	14.33–22.4	13.8–29.8	22.55-30.88				
Na ₂ O	2.23	1.79	1.65	0.82	0.80	0.68				
-	1.3–3.7	1.16-2.52	0.28-4.92	0.24-1.73	0.20-2.13	0.08–1.75				
K ₂ O	2.03	2.94	1.46	2.99	1.40	0.91				
2	0.72–5.59	2.3–4.25	0.09–4.1	2.25-3.77	0.08–3.0	0.22–1.88				
P_2O_5	0.16	0.12	0.13	0.11	0.11	0.07				
2 5	0.09-0.21	0.11-0.13	0.02-0.22	0.09-0.14	0.02-0.19	0.04-0.12				
SO ₃	0.05	0.06	0.10	0.03	0.17	0.06				
5	0.01-0.19	0.02-0.12	0.02–0.7	0.02-0.07	0.05-0.95	0.03–0.25				
H_2O	0.36	0.24	0.34	0.75	0.44	0.56				
2 -	0.05-0.99	0.01-0.4	0.03–1	0.55-0.99	0.02-1.76	0.07-1.58				
CO_2	1.82	7.59	1.38	7.48	2.61	7.39				
2	0.11-4.07	5.5-11.55	0.33-4.18	5.2-11.77	0.88-4.51	4.95-10.23				
Total	99.64	99.74	99.42	99.33	99.48	99.33				
HM	0.31	0.31	0.31	0.29	0.25	0.21				
	0.24–0.37	0.30–0.33	0.23-0.41	0.23-0.34	0.10-0.38	0.10-0.32				
AM	0.21	0.22	0.21	0.21	0.16	0.15				
	0.16-0.26	0.21-0.24	0.15-0.29	0.15-0.26	0.05-0.22	0.06-0.23				
NAM	0.33	0.41	0.26	0.36	0.24	0.23				
	0.21-0.57	0.39–0.44	0.09-0.53	0.23-0.46	0.06-0.47	0.08-0.34				
AM	1.57	0.71	2.13	0.33	0.73	0.88				
	0.3–4.64	0.3–1.1	0.14–7.2	0.08–0.76	0.1–2.88	0.12–2.74				
FM	0.16	0.13	0.17	0.15	0.13	0.12				
	0.07-0.27	0.10-0.16	0.07–0.26	0.12–0.17	0.07–0.18	0.07-0.17				
IM	0.42	0.33	0.41	0.35	0.47	0.40				
	0.30-0.74	0.31-0.34	0.21-0.72	0.26–0.45	0.23–0.97	0.23–2.93				
TM	0.042	0.035	0.047	0.041	0.041	0.23-2.93				
1 1 1 1	0.025-0.06	0.033-0.04	0.015-0.07	0.035-0.05	0.016-0.06	0.034				
				0.053-0.03						

 Table 1. Chemical composition of calcareous-silicate rocks of the Kharagol Formation, wt %

Notes: Rock groups: (1) diopside gneisses and crystalline schists; (2) scapolite–diopside; (3) scapolite–wollastonite–diopside with up to 10% calcite; (1a, 2a, 3a) the same with up to 30% calcite. Number of samples is shown in parentheses. Petrochemical modules (Yudovich and Ketris, 2000): (HM) hydrolizate (TiO₂ + Al₂O₃ + Fe₂O₃ + FeO + MnO/SiO₂), (AM) aluminosiliceous (Al₂O₃/SiO₂), (NAM) normalized alkalinity (Na₂O + K₂O/Al₂O₃), (AM) alkaline (Na₂O/K₂O), (FM) femic (Fe₂O₃ + FeO + MnO + MgO/SiO₂), (IM) iron (Fe₂O₃ + FeO + MnO/TiO₂ + Al₂O₃), (TM) titanium (TiO₂/Al₂O₃). Average values and their ranges are shown for components and modules.

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Table 2. Trace element composition of calcareous-silicate rocks of the Kharagol Formation, ppm

Element	1 (16)	1a (3)	2 (16)	2a (3)	3 (19)	3a (9)
Li	26	16	28	6	12	4
	9–56	14–18	10–54	5-8	2–33	2-8
Rb	60	47	46	65	47	31
	14–160	18–68	2-170	12–100	3–110	3–62
Cs	1.8	2.7	2.1	4.3	1.4	1.2
	1–5	1–5	1-8	1–11	1-4	1-2
Ba	880	1583	1035	3400	1510	880
	380-1400	950-2800	36-4400	1900-4500	170-3400	200-3000
Sr	590	863	606	300	522	624
	160-1900	550-1200	120-1800	200–450	170–1600	170-230
Be	1.7	1.5	1.6	1.3	1.7	1.0
	0.7–2.9	1.3–1.6	0.8–3.1	0.85-1.9	0.6–3.0	0.2–1.55
Pb	20	27	18	57	15	10
10	10-59	19–33	3.4-48	20–110	2.9–38	4.5–18
Zn	115	89	111	131	109	79
211	86–140	89–90	71–160	93–160	14–180	36–130
Sn	5.0	4.6	5.5	5.1	5.7	3.0
511	2.7–9.3	2.8–6.0	2.3–10	4-6.1	1.6–11	1.2–5.9
C		13		13	1.0–11	
Co	16		17			10
NT.	4.6–31	9.2–18	4.2-46	10–15	7.3–33	5.1–18
Ni	133	42	100	39	48	25
a	15–330	19–56	18–220	31–49	20–83	10–59
Cr	250	87	190	72	62	36
	33–560	78–92	35-420	65–79	36–110	11–79
V	94	69	98	53	63	42
	35–160	56–84	18–200	51–57	41-88	17–82
Cu	26	17	29	25	34	21
	11–39	8.4–30	13-82	17–34	2–100	7.7–46
Sc	17	16	18	14	13	9
	6.6–26	12–23	10–30	10–18	6.8–23	3.4–20
La	30	43	26	24	26	19
	14-42	37–46	5–48	21–28	7–49	10–31
Ce	54	61	55	49	47	40
	25-86	57–64	22–87	30–66	20–76	20–59
Nd	31	30	33	30	32	24
	21–43		26–50		30-40	12–30
Yb	2.2	2.3	2.2	2.4	2.6	1.5
	1.5-2.8	1.3-3.65	1.45–3.1	2-2.8	1.3–3.8	0.6–2.55
Y	21	25	22	25	25	18
	17–31	14-41	12.5–34	22–31	11–42	6–33
Zr	164	153	143	152	115	63
	122–210	145–160	97–209	99–245	50-179	49–78
F	470	285	460	537	262	131
	170-1300	225-340	40-1000	300–970	90–550	70–200
В	17	15	26	19	18	10
	2.8-46	9.3–2.6	5.9–91	15–21	2.2–45	2-45
Ag	0.09	0.06	0.08	0.16	0.10	0.12
0	0.07–0.1	0.06-0.07	0.03–0.2	0.03–0.4	0.02–0.3	0.02–0.3
Ge	1.9	1.4	1.9	2.4	1.9	1.3

Note: See Table 1.

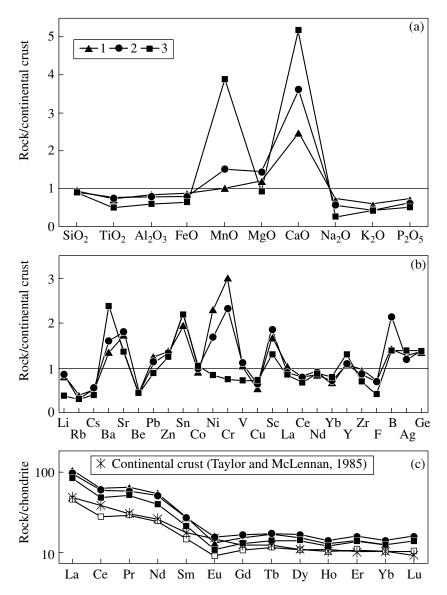


Fig. 2. Distribution patterns of (a) major, (b) trace, and (c) rare earth elements in calcareous–silicate rocks of the Kharagol Formation. (a, b) Normalized to continental crust (Vinogradov, 1962); (c) normalized to chondrite. Numeral designation in Figs. 2–8: (1) Diopside gneisses and crystalline schists (group 1), (2) scapolite–diopside rocks (group 2),

(3) wollastonite–scapolite–diopside rocks (group 3). Open symbols are carbonate subgroups 1a, 2a, and 3a, respectively.

seems to be diluted by the carbonate material. The exceptions are Ba, Sr, Pb, and Y, which can enter calcite as significant isomorphous impurities or possibly coprecipitate with carbonate. Their contents in carbonate subgroups are similar and occasionally higher than those in less calcareous groups. Carbonate subgroups and group 3 more often contain rocks characterized by the predominance of K_2O over Na₂O.

In terms of REE distribution, the groups strongly overlap owing to the presence of garnet-containing varieties and high content of titanite, which are the main REE carriers. Generally, the total REE content regularly decreases from group 1 to group 3 and carbonate subgroups (Table 3). The REE distribution is

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generally similar to that in CC, except for the higher LREE and total REE contents and significant negative Eu anomaly (Fig. 2c).

Data points of the rocks are spread over a wide field in the a-b diagram (Fig. 3) compiled after Neelov (1980). In most cases, parameter a shows a range of 0.16–0.3, which corresponds to psammitic and aleurolitic subgroups of terrigenous rocks. Parameter bshows a wide scatter, mainly reflecting the carbonate content in rock. Generally, the data points form a wide band extending from calc-alkaline andesite–basalt volcanic rocks to carbonatites. Most data points are plotted in the field of mixed compositions. In spite of strong overlapping, the fields of groups (1 + 2) and 3 are

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Element		1			2		3	3	a	CC
Element	BC-537	BC-549	I-22	BC-545	BC-560	BC-332	TB-75	BC-340	BC-194	<i>cc</i>
La	43.82	24.18	33.94	17.8	44.91	25.76	29.30	23.44	6.00	16
Ce	65.58	42.51	53.29	28.04	74.69	36.15	45.89	37.82	9.00	33
Pr	9.61	7.1	8.41	4.12	10.69	4.77	7.36	5.90	1.60	3.9
Nd	37.19	28.87	35.54	18.61	43.35	18.75	30.89	24.44	6.10	16
Sm	5.64	4.91	6.02	3.17	7.71	2.78	5.76	4.43	1.40	3.5
Eu	1.20	0.95	1.46	0.73	1.54	0.59	1.04	1.03	0.32	1.1
Gd	4.34	3.55	4.57	2.64	6.24	1.95	5.00	4.27	1.50	3.3
Tb	0.87	0.69	0.85	0.45	1.18	0.33	1.00	0.87	0.25	0.6
Dy	4.92	4.84	5.74	3.00	8.12	2.32	7.01	5.60	1.50	3.6
Но	0.97	0.85	1.02	0.59	1.49	0.46	1.31	1.17	0.32	0.78
Er	3.21	2.72	3.13	1.79	5.07	1.53	4.45	3.76	0.91	2.2
Yb	2.88	2.22	2.81	1.58	4.32	1.43	3.73	3.33	0.90	2.2
Lu	0.47	0.38	0.46	0.28	0.73	0.26	0.63	0.53	0.13	0.3
Total REE	180.7	123.76	157.24	82.78	210.04	97.07	143.36	166.6	29.93	86.48
Eu/Eu*	0.75	0.7	0.86	0.77	0.68	0.78	0.59	0.73	0.68	0.99
La _n /Yb _n	10.2	7.3	8.1	7.5	7.0	12.1	5.2	4.7	4.5	4.86

Table 3. REE composition of calcareous-silicate rocks of the Kharagol Formation, ppm

Notes: Numbers of rock groups as in Table 1. (CC) average composition of the continental crust (Taylor and McLennan, 1985). (BC-537) diopside gneiss; (BC-549) biotite-diopside gneiss; crystalline schists: (I-22) garnet-diopside, (BC-545) diopside-scapolite, (BC-560) garnet-diopside, (BC-332) low-calcareous wollastonite-diopside-scapolite, (TB-75) garnet-wollastonite-diopside-scapolite, (BC-340) garnet-wollastonite-diopside-scapolite, (BC-194) calcareous wollastonite-diopside-scapolite.

slightly separated and only samples from groups 1 and 2 fall into the volcanic field. If decarbonatization is taken into consideration, compositions of protoliths must shift to the left with respect to metamorphic rocks, but

the general pattern should not change. The same situation is observed in the Predovskii diagram (Vasil'ev *et al.*, 1981a) where the majority of wollastonite-free rocks (groups 1 and 2) falls into the tuffite field (ortho-

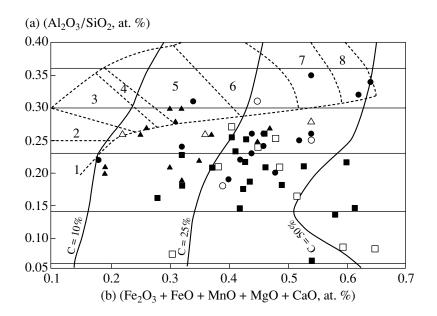


Fig. 3. (a, b) Diagram for calcareous–silicate rocks of the Kharagol Formation. Solid lines discriminate supergroups and groups after A.N. Neelov. Dashed lines outline field of calc-alkaline volcanic rocks with subdivision from (1) ultraacid rhyolites to (8) basanite–basalts.

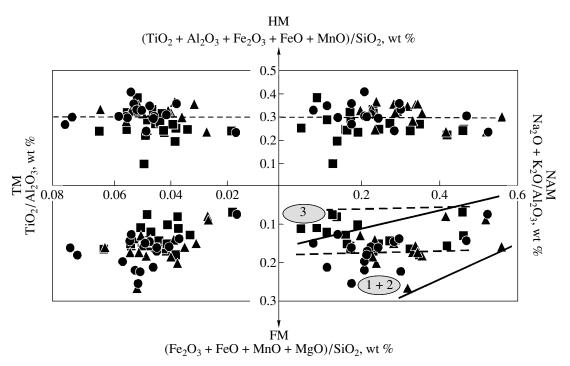


Fig. 4. Module diagram for calcareous–silicate rocks of the Kharagol Formation. The FM–NAM quadrant contains fields of groups 1 + 2 (solid lines) and 3 (dashed lines).

rocks at the boundary of intermediate and mafic compositions with basaltoids), while the remainder rocks are plotted in the field of mixed volcanosedimentary rocks.

The application of petrochemical modules proposed by Yudovich and Ketris (2000) suggests the following conclusions (Fig. 4). Based on hydrolizate module (HM), the main indicator of hypergene differentiation degree, the majority of samples are grouped near the value of HM = 0.3, which is the boundary between silites and siallites, and the groups are significantly discriminated. Based on the MgO content, rocks of group 1 are mainly attributed to pseudosiallites (MgO > 3%); group 3, to silites (partly, pseudosilites); and group 2, to both fields. Negative HM-titanium module (TM) and HM-normalized alkalinity module (NAM) correlations, which are typical of normal sedimentary (lithogenic) rocks, are absent. In contrast, positive TM-femic module (FM) correlation typical of petro- and pyrogenic sediments is observed.

In terms of FM and NAM values, the sampling set is divided into two overlapping fields. Groups 1 and 2 show negative FM–NAM correlation that is typical of volcanogenic rocks. This correlation is absent in wollastonite rocks (group 3). Comparison of rocks based on two hydrolizate elements (Ti and Al) reveals an inverse correlation (Fig. 5). Positive correlation between these elements for group 3 and subgroup 3a indicates a possible contribution of intensely weathered products in protoliths. Some samples from groups 1 and 2 fall near this trend, but the total set of data points for these two groups show no correlation between TiO_2 and Al_2O_3 . It should be noted that group 1 is closest to the andesite standard in terms of petrochemical modules (Yudovich and Ketris, 2000).

Let us consider trace elements Ba, Cr, and Ni characterized by the maximal discrimination between the groups (Table 2). The Ba distribution suggests that it is accumulated in sediments as an isomorphous impurity in carbonate. The initial $CaCO_3$ content in protoliths can roughly be estimated by the normalization of CaO

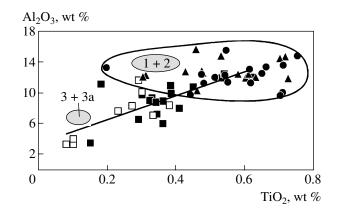


Fig. 5. Al_2O_3 vs. TiO₂ relationship in calcareous–silicate rocks of the Kharagol Formation. The calculated trend is shown for groups 3 and 3a and field is outlined for groups 1 and 2.

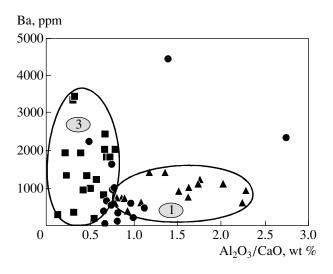


Fig. 6. Ba vs. Al₂O₃/CaO in calcareous–silicate rocks of the Kharagol Formation. Fields of groups 1 and 3 are outlined.

to Al_2O_3 or SiO_2 . In the Ba– Al_2O_3/CaO (Fig. 6) or Ba–SiO₂/CaO diagrams, groups 1 and 3 distinctly differ but lack any correlation. Like in many other cases, group 2 occupies a transitional position Ba presumably precipitated together with carbonate as an individual mineral phase (barite or witherite?) and accumulated in biotite and feldspars of the clastic aluminosilicate constituent.

Cr and Ni are more informative. In rocks of groups 1 and 2, Cr and Ni show positive correlation (Fig. 7a), which is typical of magmatic rocks. The positive Ni–MgO correlation, which is typical of ultramafic–mafic igneous rocks, is observed only for group 1 (Fig. 7b). Group 3 shows no such correlations; however, lower Cr contents positively correlate with Al and Fe (Figs. 7c, 7d). Such correlations are typical of lithogenic sediments (especially, pelites), where Cr is adsorbed by clay minerals and iron hydroxides.

Thus, the Kharagol rocks reveal distinct signatures of volcanogenic (magmatic) sources. They are most prominent in low-calcareous rocks (group 1) and less developed in other groups with high CaCO₃ contents, particularly in group 3 marked by significant signs of hypergene differentiation.

Chemically, the volcanogenic material is close to andesites. Based on the generally accepted subdivision of orogenic volcanic rocks into island-arc and continental types, the Kharagol andesite-type rocks may be considered continental rocks with respect to high K, Rb, Ba, Sr, Ni, and Cr concentrations and the presence of negative Eu anomaly. More precise interpretation needs the comparison with counterparts, since tectonic discriminant diagrams are only available for basaltic rocks. Therefore, we chose compositions best corresponding to pure volcanic rocks (hereinafter, metavolcanic rocks) based on the following criteria: absence of calcite and minimal CaO contents suggesting the minimal CaCO₃ content in protoliths; and esite range of SiO₂ contents (59–60 wt %); confinement to volcanic fields in the petrochemical diagrams; and correspondence of petrochemical modules and coefficients to the average and esites.

Geochemical criteria for the identification of orogenic andesites in different tectonic settings have been scrutinized in (Bailey, 1981) based on the following three-member (instead two-member) classification of volcanic rocks: (a) oceanic island arcs, (b) continental island arc and active continental margins with thin and normal crust, and (c) active margins with thick continental crust (Andean type). A similar classification of volcanic rocks has been proposed in (Miyashiro, 1974; Ewart, 1976).

The Kharagol metavolcanic rocks are close to average andesites but enriched in Cr, Ni, and Ba, which are typical of Andean-type andesites (Fig. 8a). Based on the majority of element ratios distinguished by Bailey as the most sensitive indicators of tectonic setting, the Kharagol rocks also correspond to Andean type (Table 4). The REE patterns of all andesites are similar (insignificant fractionation and weak Eu minimum), but the Kharagol metavolcanic rocks approximate the Andeantype andesites in terms of some LREE enrichment (Fig. 8b).

DISCUSSION

The Kharagol Formation shows a distinct rhythmiccyclic stratification of rocks (Vasil'ev *et al.*, 1981a) and complex banded and laminated structures. Structural and compositional features indicate an almost complete absence of metamagmatic rocks (resulted from lavas and sills). The distinct clastic nature of volcanogenic material suggests that the majority of rocks were tuffs. Geochemically, the volcanoclastic rocks are closest to calc-alkaline andesites of the Andean type. The amount of volcanoclastics in volcanic rocks of the Andean and Central American continental chains is estimated at 97– 98% (Carey and Sigurdson, 1987). Our interpretation of protoliths of the Kharagol rocks is well consistent with these data.

The second constituent of protoliths was carbonate material. Its proportion with volcanoclastic material was mainly responsible for the compositional variation. It is difficult to calculate the terrigenous component, but its contribution is indicated by the presence of metaterrigenous rocks in the Kharagol Formation. One more source may be synsedimentary exhalation–hydro-thermal processes, which are most distinctly indicated by specific manganese rocks, including high-Si rocks (up to spessartite quartzites) that are atypical of the Kharagol Formation. High contents of S (up to 1.5 wt % SO₃), Zn, Cu, and Ba are often observed. It is possible that Mn, Si, S, and some trace elements were supplied into the basin together with exhalations.

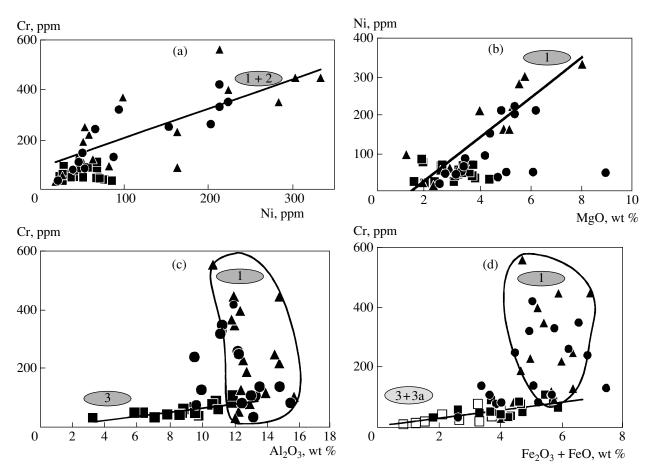


Fig. 7. (a) Cr–Ni, (b) Ni–MgO, (c) Cr–Al₂O₃, and (d) Cr–iron oxides diagrams for calcareous–silicate rocks of the Kharagol Formation. Calculated trends are shown for groups (a) 1 + 2, (b) 1, (c) 3, and (d) 3 + 3a. Fields of group 1 are outlined in Figs. c and d.

Exact identification of the tectonic type of the "Kharagol" (or "Khangarul") paleobasin is complicated owing to not only the uncertainty of such reconstructions for strongly metamorphosed sequences, but also the imperfectness and ambiguity of classifications of their recent and young counterparts (Oberg et al., 1987; Litogeodinamika..., 1988; and others). The twomember structure of the Khangarul Group (volcanogenic Kharagol Formation at the base and terrigenous Bezymyannyi Formation at the top) corresponds to the succession typical of the subduction-related back-arc basins (Litogeodinamika..., 1998). However, it is considered that typical marginal basins do not form in the rear part of continental volcanic arcs. Basins formed on the continental crust at the destructive plate margins are occasionally distinguished as epicontinental or marginal troughs, immature marginal basins, rear basins of foreland, and so on (Kuz'min, 1985; Zonenshain and Kuz'min, 1993; Oberg et al., 1987; Litogeodinamika..., 1998; and others). The paleobasin of the Kharagol time can be determined with certain confidence as a epicontinental, relatively shallow-water structure with mainly subaerial island-type explosive volcanism and predominance of telepyroclastic (aqueous and eolic transportation) sedimentation.

Unlike the Khangarul Group, mafic rocks of the Slyudyanka Group (16% of the section) are metamagmatic rocks (lava flows and sills) (Vasil'ev *et al.*, 1981a). Petrogeochemically, they approximate mid-oceanic ridge low-Ktholeites (Petrova and Levitsky, 1986). Rocks with volcanoclastic features are relatively scarce and mainly associated with metabasalts probably owing to subaqueous (below pressure compensation level) fissure eruptions with a limited production of hyaloclastic material (due to "thermal shock" of lavas) and its proximal transport. Geochemical parameters indicate that metaterrigenous rocks (15% of the section) can be interpreted as recycled products ("second cycle rocks") of an ancient granulite–gneiss complex (Petrova *et al.*, 2002).

Subaqueous hydrothermal gases presumably supplied P, Si, and, possibly, Cr and V fixed in specific siliceous–carbonate metaphosphorites (Vasil'ev *et al.*, 1981a; Reznitsky, 1995).

Diverse carbonate rocks occupy more than one-half of the group volume. The Slyudyanka and Khangarul paleobasins are sharply different. Although the Slyudy-

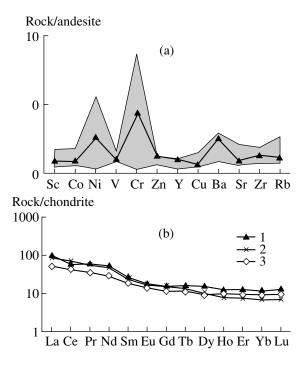


Fig. 8. Spidergrams for andesites. (a) Normalized to average andesite. Shaded field is Andean-type andesites. Solid line denotes the Kharagol metavolcanic rocks; (b) normalized to chondrite. (1) Kharagol metavolcanic rocks, (2) andesites of the Andean-type active continental margins, (3) island-arc andesites.

anka paleobasin also has no adequate recent counterpart, it can be ascribed to active (with back-arc spreading) marginal basins initiated on a relatively thin continental crust. Significant supply from island arc was absent (proximity to continent?), and the rock sequence formation is mainly related to intrabasin volcanism and continental input associated with intense carbonate accumulation.

The age of granulite metamorphism of the Slyudyanka Complex was estimated as Early Paleozoic. The Nd data suggest the lower age limit of protoliths of both groups at 1.8–1.9 Ga (Kotov *et al.*, 1997, 2000). If tectonic boundary between the groups is not ruled out, their age relations become uncertain and one can suppose two versions: accumulation of protoliths in subsequently formed paleobasins or simultaneous accumulation in laterally separated basins with the subsequent tectonic juxtaposition of their sequences. The second version is applicable for the western Baikal region where the Olkhon and Anga groups correlate with the Slyudyanka and Khangarul groups, respectively (Petrova *et al.*, 1995; Makrygina and Petrova, 1998).

The protoliths are assumed to accumulate in a conjugate system of marginal sea (Olkhon Group) and mature island arc (Anga Group). However, this model has met with some contradictions in the Southern Baikal region, since the Slyudyanka paleobasin is closer to a typical back-arc basin formed in the rear part of an island arc, while the Khangarul basin was initiated on a thick Andean-type continental crust (evidently, in the rear part of the continental arc). Detailed mapping showed that the Kharagol Formation is always present, but it rests on different stratigraphic levels of the Slyudyanka Group. This is inconsistent with the inferred tectonic contact. According to paleostructural reconstructions, the Slyudyanka Group underwent at least one deformation event prior to the deposition of the Khangarul Group (Vasil'ev *et al.*, 1981b).

Thus, available data support the following scenario of subsequent accumulation or cyclic development of rock groups: closure of the Slyudyanka Basin during ongoing subduction, subsequent deformation and metamorphism, growth and thickening of the crust, and initiation of the Khangarul Basin on the Slyudyanka basement. It should be noted that the Andean orogenic belt evolved through several cycles with certain temporal changes in types of basin and volcanism (Oberg *et al.*, 1987).

Let us consider two issues mentioned in the introduction: specifics of the protolith of wollastonite rocks and geochemical indicators of the Kharagol Formation.

The wollastonite rocks are marked by not only high CaO contents but also increased SiO₂ contents in the aluminosilicate portion (lower Al/Si ratio and hydrolizate module). These rocks reveal a more significant evidence of hypergene differentiation. They approach specific manganese rocks as a result of high contents of MnO and episodic sulfur, as well as highest contents of Cu (up to 100 ppm) and Zn (up to 180 ppm) (Tables 1, 2), indicating similar sedimentation conditions. Their protoliths presumably accumulated during the attenuation of volcanism and clastogenic sedimentation accompanied by increasing contribution of aquagenic sedimentation and exhalation sources. Allochemical alteration of pyroclastics (pelitization, carbonatization, and silicification) could also be intensified during these periods of relatively slow sedimentation.

In any case, simultaneous excess of calcite and silica is the main feature of protoliths considered above. Such a composition of sediments is explained by specifics of the metamorphogenic wollastonite formation. In the granulite complex, wollastonite is confined to areas of relatively short-term decrease in partial CO₂ pressure. In such a situation, mineral dissolution and component diffusion rates become the major kinetic factors. The optimal environment is provided by the presence of direct contacts between quartz and calcite. The scale of such contacts and wollastonite formation is naturally governed by the contents of both minerals. Wollastonite is the highest-temperature mineral among decarbonatization products of calcareous-silicate rocks in the Kharagol Formation. Free silica and calcite are gradually consumed with increase in metamorphic grade. At the high-grade stage, their excess is preserved only

Element ratios	1	2	3	4
Rb/Sr	0.039	0.094	0.113	0.13
	(0.018–0.047)	(0.029–0.4)	(0.048–0.45)	(0.11–0.15)
ΣΡЗЭ	35.2	94.4	146	157.24
	(31–51.6)	(64–135)	(106–213)	
La/Yb	1.2	8.9	16.5	13.55
	(0.92–1.26)	(3.3–12.6)	(12.2–21.4)	(8.1–19)
La/Y	0.11	0.93	1.46	1.5
	(0.076–1.25)	(0.6–1.1)	(1.28–1.63)	(1.4–1.6)
Th/U	0.72	5.36	6.0	5.1
	(0.27–2.57)	(3.64–7.8)	(4.0-8.2)	
Zr/Y	2.2	5.4	14.6	7.0
	(1.5–3.8)	(4.3–10)	(6.6–30)	(5.6–8.3)
Zn/Cu	1.9	2.3	2.8	5.0
	(0.77–3.1)	(0.95–3.9)	(1.4–5.8)	(3.3–6.7)
Ni/Co	0.29	0.95	1.4	4.0
	(0.2–0.76)	(0.45–2.2)	(0.33–3.3)	(2.4–5.6)
Sc/Cr	3.8	0.61	0.36	0.13
	(0.7–15.5)	(0.13–2.1)	(0.06–0.75)	(0.08–0.17)
Sc/Ni	3.4	1.1	0.55	0.31
	(2.1–6.2)	(0.44–4.2)	(0.18–1.0)	(0.20-0.42)

Table 4. Indicator ratios of elements in calc-alkaline andesites of different geodynamic settings (Bailey, 1981)

Notes: Andesites: (1) oceanic island arcs, (2) continental island arcs, (3) Andean-type active continental margins; (4) metavolcanic rocks of the Kharagol Formation. Average values and variation ranges are shown.

in rocks with a high content of both components in the protolith.

The most distinct geochemical feature of calcareous-silicate rocks of the Baikal region is high Cr and Ni contents in low-calcareous (andesite-type) rocks and high Ba contents in high-calcareous rocks. As was mentioned above, Kharagol rock outcrops continuously extend along the southern wall of the Tunkin Valley, varying in metamorphic grade from the granulite to greenschist facies. In the moderate-temperature interval, the rocks are close to high-grade epidote-hornblende-diopside (with occasional scapolite) calcareous gneisses and schists. In the low-grade facies, they are observed as mica-carbonate-(quartz, plagioclase) and biotite-(epidote, chlorite)-actinolite calcareous schists. Available analytical data (several tens of samples) indicate that, regardless of metamorphic grade and mineral assemblage, low-calcareous rocks have anomalously high contents of Cr (up to 1000 ppm) and Ni (up to 500 ppm), while high-calcareous rocks have a high Ba content (up to 1700 ppm). Contents of these elements in 50–70% of samples are close to or higher than average values for corresponding three groups distinguished in the Slyudyanka Complex. Hence, preliminary data indicate that geochemical features of the rocks are fairly stable. Therefore, in combination with geological and petrographic data, they can serve as a guide for identification of the Kharagol Formation in problematic situations.

CONCLUSIONS

(1) Protoliths of calcareous–silicate rocks of the Kharagol Formation were tuffoids with different contents of calcareous material. In terms of petrogeochemistry, volcanoclastic material of tuffoids is close to Andean-type calc-alkaline andesites.

(2) The Kharagol (Khangarul) paleobasin is an ensialic-type structure developed on a thick continental crust in an active margin setting.

(3) In the summary section of metasedimentary sequences of the Khamar Daban composite terrain, the Kharagol Formation can be considered a boundary between subterrains with protoliths accumulated in different paleogeodynamic settings.

(4) Geochemical indicators of the Kharagol Formation in different metamorphic zones may be increased contents of Cr, Ni, and Ba in calcareous–silicate rocks.

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