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Experimental measurement coefficient distribution TR, Ni, Mn melilite-melt

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The paper discusses the distribution coefficients of trace elements obtained in the experimental melting of melilitic melts containing between pyroxene-melt and melilite-melt. The data on the distribution of Zr, Hf, TR, Ni, Co, Mn compared with the estimates of the distribution coefficients in natural melilite-bearing porphyritic volcanic rocks. Comparison of the data and analysis of literary sources suggest the possibility of changing ratios Zr/Hf, Th/U, Ni/Co in the fractionation of melilite and pyroxene in the high-Ca alkaline melts.

Key word: distribution coefficients of trace elements, melilite, alkaline magma differentiation

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One of fundamental problems of geochemistry is genesis and formation conditions larnite-normative magmas. Interest to this type of magmas is caused by that of large-scale deposits is closely connected with them (Kovdor, the Gulinsky intrusion, Afrikanda). In paragenesis with them carbonatite complexes are observed, also. Subvolcanic rocks of such type quite rare. The most known regions where they are widespread it the African province central rift [*Gerasimovsky & Polikov, 1974; Bailey et al., 2005*] and Maymecha Kotuiy province [*Juk Pochekutov et. al., 1975; Butakova & Egorov, 1962; Egorov, 1969; Rass, 2008]*, the Kola (Turiy Peninsula) province [*Ivanikov et al 1998, Beard et al., 1998, Dunworth & Bell 2003, Arzamastsev, et al 2009*], the Rhine graben [*Hegner et al., 1995; Wilson et al., 1995; Dunworth & Wilson, 1998, Ulianov et al., 2007*]. The same type found from the oceanic islands. They are presented on Trindade islands, Cape Verde, Fernando de Noronha, Hawaii, Gran Canary. The assessment of coefficient distribution (Kd) of trace elements for these liquids represents considerable interest as for understanding of processes of a fractionation of trace elements, and for an assessment of influence of a high alkaline melts on the variation Kd value.
We investigated several natural samples porhyritic melilitites (Ol, Mell, ±Cpx as porphyric-crystal) (African and Maimecha Kotuyi province), and spent several isothermal experiments, with the melt close to turyaite and melilitite (sample 11073).

Table 1. Composition of the rock (val) and groundmass (om) and glass (Liq) in micro inclusion in porphyric mineral

Sample	phase	SiO_2	TiO_2	Al_2O_3	FeO	MnO	MgO	CaO	Na_2O	K_2O	P_2O_5	Sum
87_50	val	36.52	1.77	12.07	10.14	0.143	8.65	20.57	5.24	2.09	0.44	98.96
87_51	val	34.97	2.47	8.78	10.31	0.234	25.86	11.17	3.83	0.349	0.416	99.08
87_51	om	36.24	2.46	9.75	10.77	0.27	24.68	10.68	2.485	0.92	0.045	98.58
87_51	om	35.85	2.92	9.06	10.43	0.265	26.78	11.78	1.25	0.87	0.028	99.51
87_49	val	39	1.94	11.94	8.696	0.11	9.19	16.61	5.58	1.526	0.178	98.43
87_49	om	38.07	2.18	11.49	9.42	0.114	9.4	21.16	4.52	1.37	0.084	99.26
87_49	Liq	36.2	3.5	2.62	23.41	0.085	0.64	33.5	0.011	0.018		99.83
87_49	Liq	33.5	8.76	2.71	20.18	0.069	1.39	33.3	0.0237	0.007		101.42
87_48	val	37.25	2.24	10.88	10.29	0.13	8.65	20.37	4.18	1.55	0.13	101.42
87_48	om	44.11	0.015	8.716	2.564	0.1	7.66	33.32	3.426	0.081		98.98
87_48	om	43.93	0.018	8.283	2.46	0.099	8.13	33.89	3.04	0.089		100.39
87_48	Liq	40.62	5.237	16.05	6.31	0.009	16.74	9.72	1.928	2.87		100.21
87_48	Liq	40.73	5.202	15.58	6.69	0.04	15.44	12.32	1.914	1.66		101.49
87_48	Liq	40.89	5.015	15.33	6.51	0.044	15.93	12.48	1.94	1.56		99.43
87_48	Liq	51.96	0.415	7.75	4.86	0.11	17.23	16.3	0.986	0.077		100
0014	om	41.18	3.16	6.94	12.7	0.167	12.15	10.94	3.4	2.17	0.56	98.07

0014	val	39.08	1.896	6.82	12.49	0.146	15.6	15.2	3.48	2.05	0.67	99.93
0014	val	38.88	1.857	6.69	12.46	0.156	15.75	15.12	3.47	2.02	0.656	99.56
turiyte	val	41.84	3.18	7.4	13.03	0.21	14.64	15.34	2.25	2.07		99.96
11073	val	41.14	9.697	3.25	10.93	0.132	9.88	11.44	1.93	4.901	0.96	97.93
11073	om	39.22	8.54	1.79	13.58	0.18	16.11	14.54	2.24	1.82		98.02
11512	val	34.86	7.05	4.034	12.3	0.191	15.07	16.12	1.44	4.07	1.028	98.78
11512	om	40.53	9.21	3.41	11.32	0.141	9.89	11.81	2.24	5.345	1.147	97.83
11511	val	35.8	6.87	4.079	12.13	0.187	14.32	16.5	1.5	3.5	1.014	99.31
11515	val	36.31	7.25	3.827	11.84	0.183	15.53	15.59	1.27	3.76	0.986	100.07
M6538	val	40.58	9.72	2.59	0.057	11.6	0.126	10.39	14.61	3.74	1.186	98.66

The complexity of the fractionation melilitic magma due to the presence of a phase system describing the crystallization of high-calcium silica undersaturated magmas peretektic reactions [*Schairer et al., 1962, Yoder, 1967, Gee & Sack 1988*]:

$$Mell(Ca_2MgSi_2O_7) + Ol(Mg_2SiO_4) + Liq(NaAlSi_3O_8) \leftrightarrow Cpx(CaMgSi_2O_6) + Ne(NaAlSiO_4) \quad (1)$$

$$Ne(KAlSiO4) + Liq(SiO2) \leftrightarrow Lc(KAlSi_2O_6) \quad (2)$$

$$Mell(Ca_2MgSi_2O_7) + Ol(Mg_2SiO_4) \leftrightarrow Cpx(CaMgSi_2O_6) + Mo(CaMgSiO_4) \quad (3)$$

Table 2. Composition (wt. %) porphyric-minerals from melilitic volcanic rock.(Melilite calculate on the 5 cations and 7 oxygen xAk – akermanite, xGe – gelenite, xNaGe – Na-melilite, xFeAk –ferric-akermanite)

Mineral	Clinopyroxene (Cpx)				Melilite (Mell)				
Sample	11073	87_48							
SiO2	51.5.	39.04	38.49	37.87	38.96	44.97	45.06	44.23	44.59
TiO2	1.5	0.06	0.09	0.04	0.10	0.00	0.03	0.01	0.00
Al2O3	3.5	14.85	11.71	16.98	16.59	8.34	8.35	8.59	8.08
FeO	4.51	5.70	13.71	3.91	4.37	1.66	2.60	2.69	2.37
MnO	0.1	0.01	0.12	0.07	0.07	0.06	0.04	0.05	0.04
MgO	15.5	3.74	0.21	3.84	3.47	8.27	7.61	7.70	7.98
CaO	23.5	36.48	35.56	37.00	36.38	33.25	32.91	33.31	33.61
Na2O	0.5	0.04	0.04	0.08	0.06	3.39	3.33	3.34	3.27
K2O		0.03	0.03	0.18	0.00	0.04	0.08	0.09	0.07
NiO		0.026	0.028	0.041	0	0.03	0	0	0
Total	100.6	100.57	101.35	100.33	100.41	100.09	100.25	100.24	100.22
xAk						59.9	55.5	54.9	58.5
xGe						8.5	8.9	9.6	8.3
xNaGe						34.3	34.1	33.8	32.9
xFeAk						7.3	11.5	11.8	10.3
Summ cation						5.2	5.2	5.2	5.2

Besides the main phase of the crystallizing – melilite (Mell), pyroxene(Cpx), have a variable composition, broad miscibility of components, which increases the uncertainty of estimates of the residual melts during fractionation. The compositions of the investigated natural volcanic rocks in the diagram Ln–Fo–An are located close to the lines of these peretectic reactions line and cotectic Ol–Mell and Cpx–Ol (Fig.1). At C point the melt is saturated with melilite and olivine begins to react with the formation of pyroxene and olivine in the process of reaction removes in the direction to point D (content of Ol decrease). In the field of point C is the thermal barrier (Ab–Di peretectic) separates the melilite-pyroxene melts from nepheline-pyroxene. The position of this point is not very defined (the approximate temperature in model systems 1140C°). It is essential important that, depending on the conditions and of the primary melt composition the differentiation trend can go in different directions toward a more high-Ca and potassic melts with melilite and vollastonite (point D), or to less alkali feldspar phonolitic volcanic rock to the end. It is interesting to note that the investigated samples are African right of the point and therefore fractionated by phonolitic trend, whereas samples Maimecha Kotuiy province provide a more alkaline and undersaturated trend.

We conducted experiments with molten melilite-nephelinites (sample 11073) and turyiate. For isothermal experiments used a tube furnace design L. D. Krigman. Powders of the samples put in a platinum capsule with a diameter of 0.05 mm. The full weight of the mixture in the capsule was approximately 200 mg. Duration of exposure at a given temperature the equilibrium of experience ranged from 24 to 57 hours depending on temperature. Since the experiments were conducted at uncontrolled oxygen fugacity - the air, we evaluate it on up to buffer NNO+1 (buffer $Ni+O_2 = NiO$). In rock substance added Zr and Hf and other trace element in the form of aqueous solutions of a given concentration. The concentrations of trace elements have changed from 0.002 to 5 wt.%.

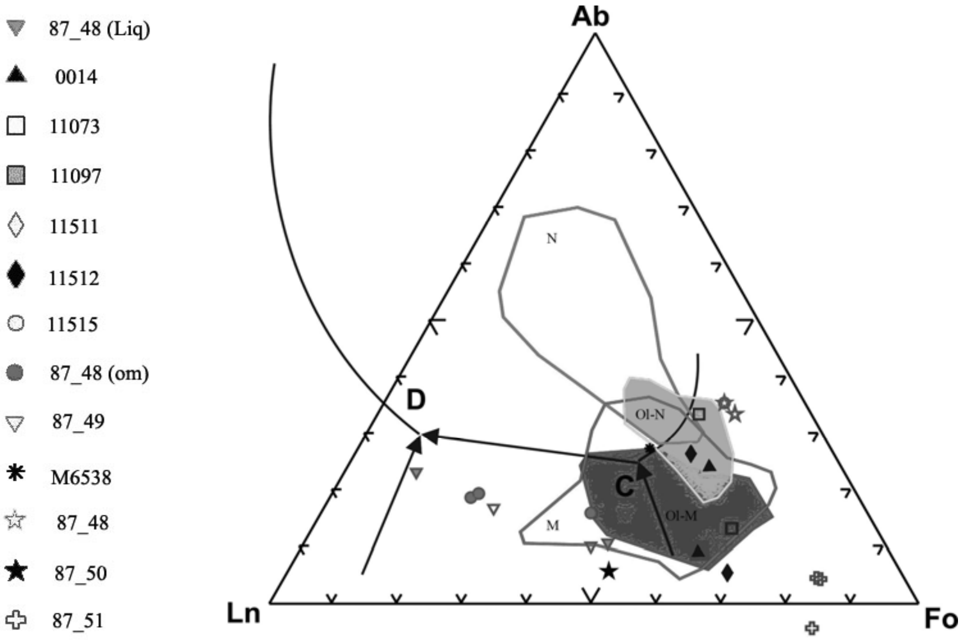

Fig. 1. Our data of melilitites in the diagram La–Ab–Fo [*Peterson, 1989*] Grey lines mark the fields: M – melilitites, Ol-N – olivine nephelinites (melanephelinite), Ol-M – melilitites olivine; N – nephelinites. Our formulations see the legend. Dark blue lines are pseudo-cotectic line and peretektic reaction: D-C (view in text).

These mixtures were melted at temperatures above the liquidus to the complete melting of the charge (incubated for one hour) and then decrease to the temperature of the isothermal sections and incubated for an equilibrium mineral-melt. Exposure time was sufficient to achieve an equilibrium concentration between minerals and liquid. Our measurement of concentration profiles of Zr, Hf, Ca, Si, Al, Mg, Fe in the glasses showed a homogeneous distribution of crystals around the phenocrysts. In the experience of the products were available for analysis of melilite (Mell), olivine (Ol) clinopyroxene (Cpx), magnetite (Mgt) and glass (Liq). Result of the runs show in tabl.2.

Table 3. Product of the run with and condition

№ run	Time duration (hour)	T°C	Phase composition	% vol. of Glass in product of run
Melilite nephelinites -11073				
06	88	1260	Liq–Ol–Mgt	
05	92	1230	Liq–Ol–Mgt	
02	48	1212	Liq–Ol–Cpx–Mgt+Bd	
04	94	1180	Liq–Ol–Cpx–Mgt+Bd	
08	111	1165	Liq–Ol–Cpx–Mgt+Bd	
03	157	1150	Liq–Ol–Cpx–Mgt+Bd	
Turiyte				
7,8	24	1250	Liq–Ol–Mgt	90
13,16	48-72	1220	Liq–Ol–Mell–Mgt	40
5,6	24	1212	Liq–Ol– Cpx–Mell–Mgt	30
12,17,18	48-72	1204	Liq–Ol–Cpx–Mell–Mgt	40
20	48	1180	Liq–Ol–Cpx–Mell–Mgt	10

3	120	1150	Liq–Ol–Cpx–Mgt–?Ne	0

(Mgt- magnetite; Bd – badelleite; Ne - nepheline)

The results of experiments were analyzed by microprobe, including those used in our specially designed technique for the analysis of low contents of Zr and Hf [*Senin et al., 1989*].

Analysis of the contents of TR in melilite from experiments with turiyte was carried out with a laser and spark mass spectrometry (SMS) [*Ramendik et al., 1989*].

Experiments have shown that in dry oxidizing conditions in the melilitic-nephelenite melts melilite does not crystallize. Only in the more undersaturated melts such as turiyte melilite crystallize. As the temperature falls the melt gets in the area of peretektic reaction - melilite is disappears, remains pyroxene. How will the change of Kd value in these conditions?

Clinopyroxene–melt equilibria

We found very high values Kd for Zr and Hf for the pyroxene-melt equilibria in the experiments with the nephelinites (fig.2).

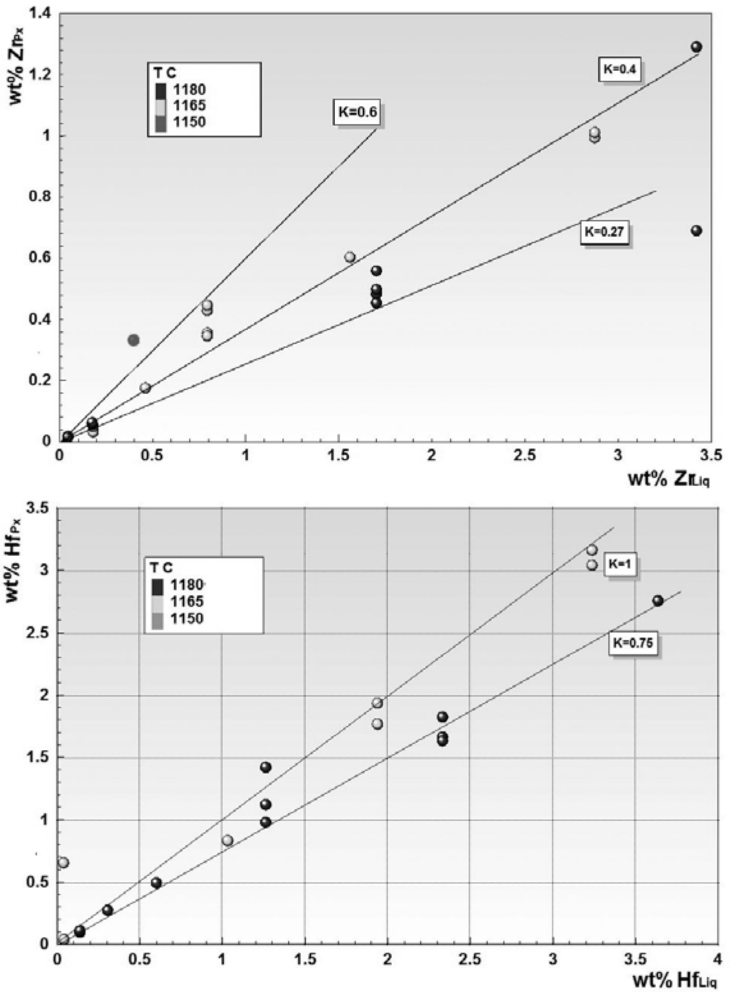

Fig. 2. Coefficient distribution pyroxene-melt of Zr and Hf in the different equilibrium temperatures

Experimentally, Kd pyroxene- melilitite-rich melts virtually not been investigated. From the experimental data, we can mention the work [*Gaetani & Grove, 1995; Hill et al., 2000*] that show Kd increases with increasing TR content in the pyroxene end-member Chermak in high calcium melts. The values Kd of Zr and Hf obtained in [*Hill et al., 2000*] is also very high, and remains a big difference in the values of (almost an order of magnitude) between them. Also published several papers on the study of equilibria in the silica-undersaturated melts. For example, in [*Green et al., 1992; Sweeney et al., 1995; Klemme et al., 1995; Blundy & Dalton, 2000; Adam & Green, 2001*] surveyed the distribution of trace elements in pyroxene carbonatite melts, which can also be viewed as an analogue of high-calcium alkaline magmas. In [*Hill et al., 2000*] studied a model kimberlitic melt.

With the exception of [*Klemme et al., 1995*] in these equilibria also seen a big difference between Kd of Zr and Hf values and also Kd value is higher than in the melts of normal alkalinity.

It is also very limited data on the Kd pyroxene of the in natural melilitic melts. We identified Kp for pyroxene in natural melilite-nepheline for sample 11073. At Fig.3 presents data on Kd in similar high-calcium melts with melilite, and our data. It is seen that the data on Kd vary widely.

In addition, there are a number of data for Kd in the natural lamproitic melts [*Foley, 1996; Foley & Jenner, 2004*]. Although the thatr melts usually have a specific composition - relatively high content of SiO2 and alkalis at low CaO.

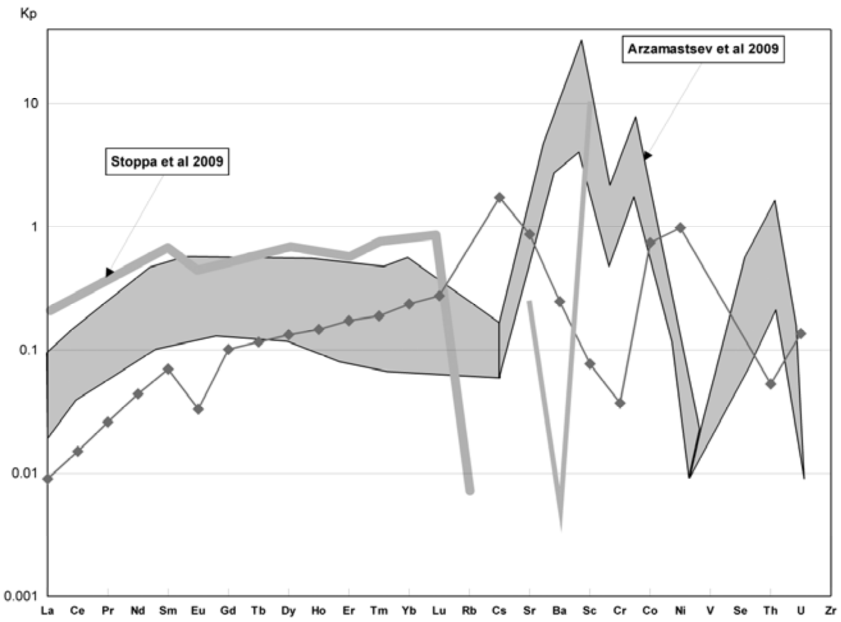

Fig.3. Kd pyroxene measured by us for sample 11073 (line with diamonds) and literature data: [*Stoppa et al., 2009*] for a range of Kd values for melilitic nephelinites – the wide gray line; [*Arzamastsev et al., 2009*] of the Turiy cape and melilite from the Kaiserstuhl - gray area

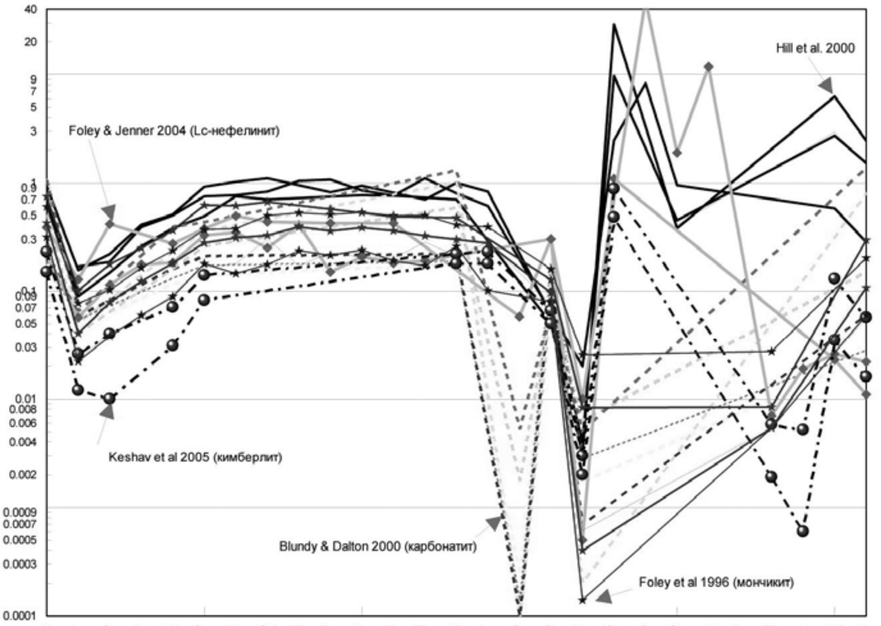

Fig. 4. Kd pyroxene from the literature: thin dashed lines – carbonatites [*Blundy & Dalton, 2000*], the black solid line – with a high content in pyroxene end-Chermak minal [*Hill et al., 2000*], black dash dotted line with a ball marker – an experiment with an analog kimberlite melt at a pressure of 6 GPa [*Keshav et al., 2005*]; line with asterisk markers, natural leucite nephelinite [*Foley & Jenner, 2004*]

The data on the natural volcanic rocks are quite different from those obtained in experimental conditions. First the estimated Kd TR have the lowest values available in literature experimental Kd, second Kd heavy rare earths relatively light monotonically increases (the slope of the curve on the graph Kd is constant), in contrast to published data where from La to Sm Kd value increases the slope and then disappears, and for the Er–Lu may decrease the value of Kd.

In the third for our sample is characterized by a very minimum for Kd europium. Most similar to the results obtained for Kd kimberlite obtained at high pressure (6 GPa) [*Keshav et al., 2005*]. However, these experiments were conducted at high temperatures (decrease values of Kd), and in our case, we would expect higher value of Kd TR. Very big difference in the values Kd of Cr, Co, Ni. Our values are less than 1, and the literature contains a similar and much higher than 1 value.

Lithophile elements are also characterized by a very wide interval of Kd values. And often reveals a large difference between the paired trace elements. Basically Kd Hf > Zr >> Nb, Th > U. All this creates a great opportunity to change the relationship pair of trace elements during the fractionation process. Interestingly, in experiments [*Gaetani & Grove, 1995, Hill et al., 2000*] with pyroxenes that contain a high proportion of Chermak-component, obtained very high values Kd Zr and Hf greater than 1. Under such circumstances, perhaps even melt depletion of lithophile trace elements the last portion of the melt.

Melilite–melt equilibria

Of great interest are the data we obtained for Kd melilite as the information on the quantities of this mineral Kd in natural melts is extremely small. Fig. 5 shows the measured by us in the turiyte Kd and a number of experimental data in the literature.

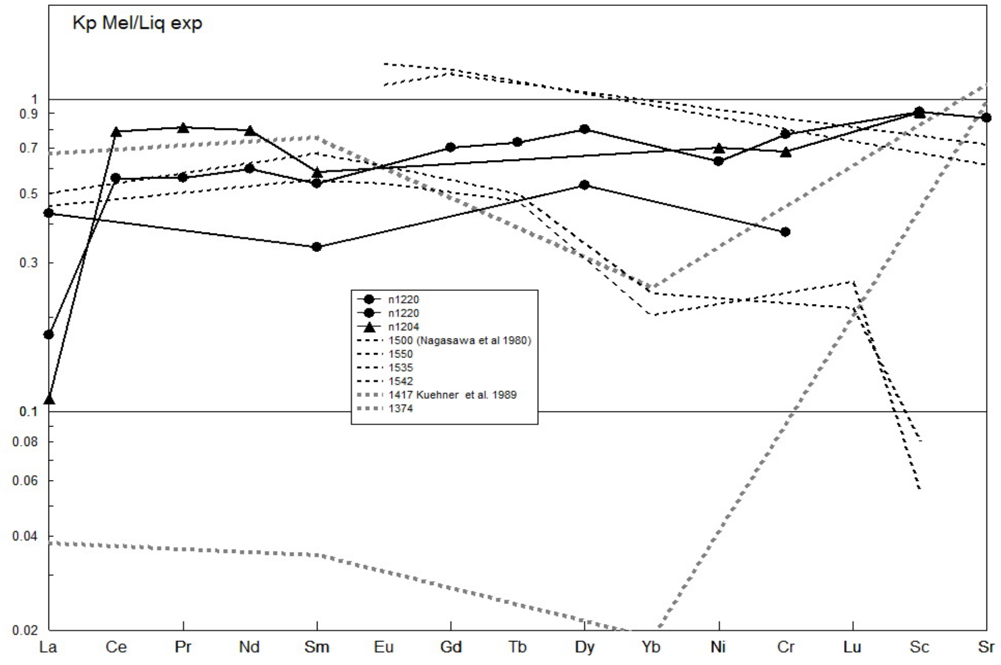

Fig. 5. Kd melilite in experiments on melting turiyte (black lines) measured by the SMS analysis. Addition experimental data [*Nagasawa et al., 1980, Kuehner et al., 1989*].

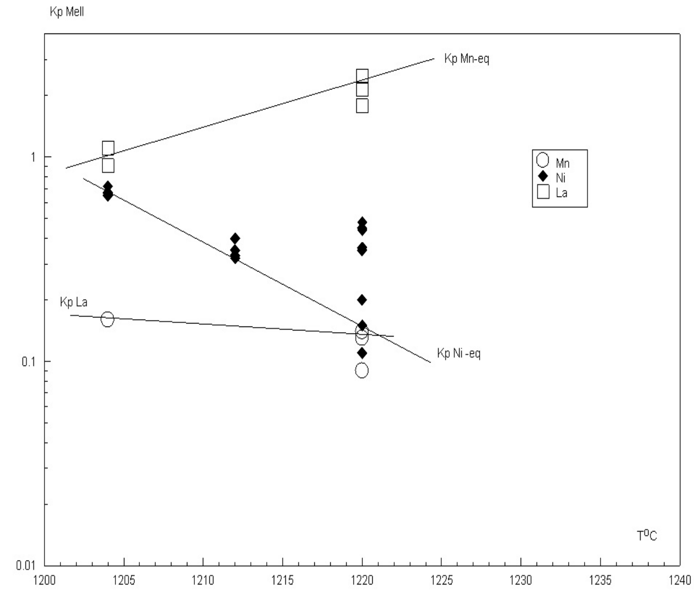

Fig. 6. Kd melilite in experiments with turiyte (lines indicate trends of change of Kd) measured by the microprobe analysis

Our data are consistent with the highest values of Kd. They are close to the Kd rare earth elements from ugandite [*Onuma et al., 1981*] and they are approaching the magnitude of the Kd melilitite lappily tuff [*Stoppa et al., 2009*]. It is also interesting negative Eu anomalies and positive Rb. The high level of Kd values of the sample studied is shown in the considerably greater than 1 Kd Sr and high Kd Pb.

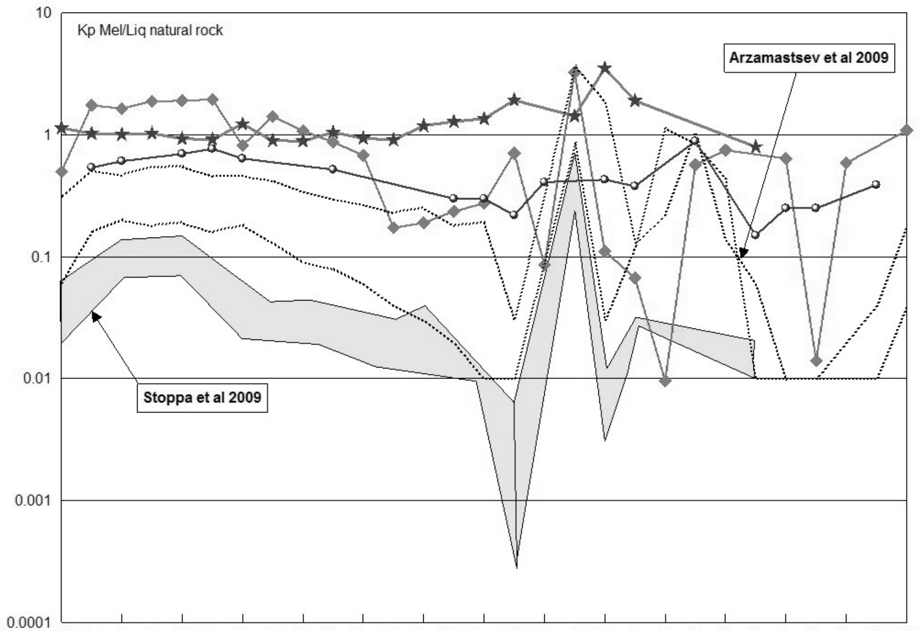

Fig. 7. Kd melilite measured us for sample 87_48 (black line with diamonds) and literature data: [*Stoppa et al., 2009*] gray box – a range of values for Kd melilitic nephelinites; and the line with the Stars – Kd melilite for melilitite–carbonatite lappilly; [*Arzamastsev et al., 2009*] point black lines – for turyaita Turego of the Cape and from the Kaiserstuhl melilitites;[*Onuma et al., 1981*] line with circular dots - Kd in Uganda from Zaire

Our results are similar to the Kd from turiyte studied by [*Arzamastsev et al., 2009*]. It is interesting to note the relatively high (but low 1) the value Kd of Co, Ni, Zr. Significant difference between the Kd Th and U. An indirect confirmation of the availability of high Kd melilite for Fe-group elements and some lithophile elements are the data [*Petibon, 1999*]. In this paper for the equilibrium melilite - carbonatite melt obtained high Kd Th 0.52, Hf 0.11 Y 0.37.

Conclusion

Our investigation of Kd pyroxene and melilite in the high-calcium melts and analysis of data in the literature, above all, shows the poor knowledge of the distribution of trace elements in the crystal-melt equilibria in these magmas.

For quite certain established patterns can be attributed – a slight difference between the LTR and HTR Kd for clinopyroxene and an inverse relationship to melilite.

High Kd melilite TR and probably high Kd clinopyroxene (due to the increase in the concentration Chermak- minal) in general reduce the rate of accumulation of incompatible elements in the evolution of these magmas.

A very important feature is the significant difference in Kd lithophile elements. For melilite and pyroxene set a big difference between Th and U, Zr and Hf.

The peculiarity of the evolution of these melts is the presence of complex peretektic reactions (especially with the addition volatile component) between the crystallizing phases, kinetic factors are likely to lead to the prevalence of non-equilibrium in natural magmas associations. In this relatively low temperature equilibrium and a small difference between the temperatures and eutectic points peretektic reaction process make insignificant variations for Kd by the temperature factor. The greatest importance is the effect of melt composition, and changes in the composition of crystallizing phases (melilite, pyroxene).

It should be emphasized that the evolution of melilitic and nephelinite melts leads to the formation of alkaline agpaitic melts, which a strong change in the Kd values.

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