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Cr-RICH AND Al-RICH SPINELS IN THE KIBI-KOGEN ALKALI BASALTS, SOUTHWEST JAPAN

by

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(with 10 text-figures and 3 tables)

Abstract

Cr-rich spinel (Cr_2O_3 11~33 wt%, Al_2O_3 18~39 wt%) and Al-rich spinel (Cr_2O_3<2 wt%, Al_2O_3 48~65 wt%) occur in the Kibi-kogen alkali basalts. The latter is extremely high in Al_2O_3 and plots near the Al end of the Fe^{+3}-Al side of Cr-Fe^{+3}-Al diagram. The occurrence and chemistry of the spinels indicate that the Cr-rich spinel crystallized as a near-liquidus phase, whereas the Al-rich one formed subsequently. The calculation in terms of the Rayleigh fractional crystallization model resulted in notable depletion of Cr in the residual liquid with progress of crystallization of the Kibi-kogen alkali basalt magma. Thus, when the Cr content of the magma became less than the solubility limit of Cr, the Cr-rich spinel may have ceased from crystallization and the Al-rich spinel in turn crystallized as an aluminous phase prior to the appearance of plagioclase. The composition of the near-liquidus Cr-rich spinel depends mainly on the Cr content of host magma rather than the Al content and Si/Al ratio.

Introduction

Spinel covers a wide compositional range exchanging Cr, Al, Fe^{+3} and Ti in the octahedral site, and Mg and Fe in the tetrahedral site, and is potentially an important indicator of the physicochemical conditions under which its host rocks were formed (Irvine, 1965). It is known that Fe^{+3} content in the octahedral site is affected by f_{O_2} (Hill and Roeder, 1974; Fiske and Bence, 1980; Batiza and Vanko, 1984) and temperature (Hill and Roeder, 1974; Fiske and Bence, 1980). The Cr/Cr+Al ratio is controlled by pressure (Irvine, 1967; Ridley et al., 1974; Sigurdsson and Schilling, 1976), f_{O_2} (Irvine, 1967), P_{H_2O} (Onuma and Tohara, 1984), and the chemical composition of host magma (Irvine, 1967, 1976; Sigurdsson and Schilling, 1976; Allan et al., 1983; Dick and Bullen, 1984), and is affected by mineral assemblage (Irvine, 1967; Hill and Roeder, 1974; Ridley, 1977).

Alkali basalt is considered to originate in the deeper place than tholeiitic basalt is derived (Kuno, 1960; Kushiro, 1968). Therefore, Shiraki et al. (1979) considered that the low Cr/Al ratio of chromian spinel in Nanzaki basanitoid may be attributd to the crystallization at relatively high pressures. However, Nagao et al. (1980) pointed out that the chromian spinel from alkali basalts~basanites shows a wide variation in Cr/Cr+Al ratio and overlaps the compositional range of the spinel from silica-saturated rocks. They stressed the effect of Al_2O_3 content of host magma on the spinel chemistry.

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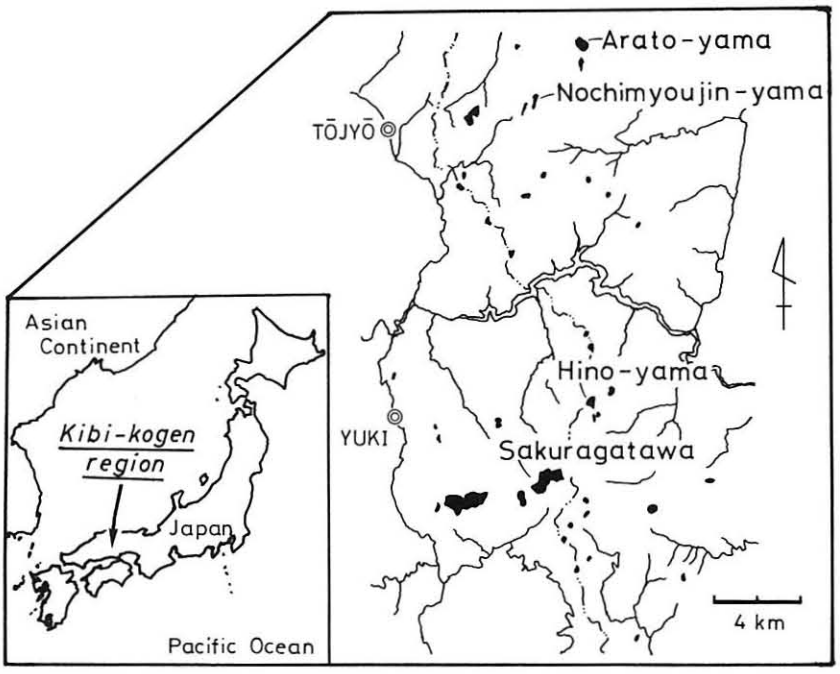
Kibi-kogen alkali basalts (alkali basalt ~ basanitoid) (8 ~ 9 Ma; Uto et al., 1986) in southwest Japan contain spinel grains. The host basalts vary in Al_2O_3 to a small extent, whereas the spinel shows a wide range in Cr/Cr + Al ratio. The occurrence, composition and origin of the spinel in these alkali basalts were studied and the compositional effect of host magma on the spinel chemistry is discussed in this paper.

Host rock

The host basalts were collected from Arato-yama (ART8), Hino-yama (HIN11), Sakuragatawa (SKR6, SKR14), and Nochimyoujin-yama (NCH4) (Text-fig. 1). These alkali basalts occur as domes, dikes or lavas, and include many ultramafic xenoliths (Ogura, 1930; Oji and Oji, 1964; Takamura, 1970, 1973; Tazaki, 1971; Fujiwara and Arai, 1982).

The chemistry of the host rocks is listed in Table 1. ART8 is regarded to be a least differentiated rock among the basalts in the Kibi-kogen region because of its high Cr and Mg' (Mg/Mg + Fe). In the Sakuragatawa basalts, SKR14 is more evolved than SKR6.

All the samples have olivine and clinopyroxene as phenocrysts. These phenocrystic minerals show normal compositional zoning. As shown in Table 1, except for SKR14,



Text-fig. 1 Index map showing the distribution of alkali basalts (black area) in the Kibi-kogen region, southwest Japan.

Table 1 Host rock analyses

No. Sample	1 ART8	2 HIN11	3 SKR6	4 SKR14	5 NCH4
SiO_2 (wt %)	46.61	47.94	45.72	47.51	48.20
TiO_2	2.01	2.12	2.44	2.22	1.80
Al_2O_3	12.73	14.05	14.91	14.29	13.49
FeO-t	10.61	10.55	11.88	10.85	10.13
MnO	0.19	0.17	0.20	0.19	0.18
MgO	12.65	10.23	10.58	10.25	10.81
CaO	11.08	10.48	11.16	10.16	10.75
Na_2O	2.60	2.62	1.57	2.42	2.50
K_2O	1.02	1.33	0.85	1.54	1.62
P_2O_5	0.50	0.51	0.69	0.57	0.53
Cr (ppm)	482	371	448	341	372
100Mg′	70	66	64	65	68
Fo (calc)	89	87	86	86	88
Cpx-mg (calc)	91	89	89	89	90
Range of composition of ol and cpx phenocrysts					
Olivine					
Fo	89 ~ 76	87 ~ 78	89 ~ 78	76 ~ 73	90 ~ 76
Cr_2O_3 (wt %)	0.07 ~ 0.00	0.09 ~ 0.00	0.09 ~ 0.00	0.06 ~ 0.02	0.07 ~ 0.00
Clinopyroxene					
100mg	90 ~ 78	91 ~ 80	91 ~ 76	88 ~ 80	88 ~ 82
Cr_2O_3 (wt %)	0.85 ~ 0.00	0.94 ~ 0.02	1.0 ~ 0.00	0.62 ~ 0.00	0.80 ~ 0.00
Modal composition					
phenocrysts					
Ol	9.6	3.5	4.8	5.8	10.2
Cpx	6.6	5.4	3.6	1.8	3.8
Cr-sp	tr	tr	tr	none	none
Al-sp	none	none	0.1	tr	0.1
groundmass	78.3	9.10	91.4	92.4	85.9

Major element chemistries were determined by using XRF at Yamaguchi Univ., and Cr was analyzed by the method of Tsuchiya *et al.* (in prep.) using XRF at Hokkaido Univ. All analyses are summed to 100 percent on a volatile-free basis. FeO-t is total Fe expressed as FeO. Mg′ and mg are $Mg/(Mg+Fe^{+2})$ for rock and mineral, respectively. Mg′ was calculated after adjusting Fe so that Fe^{+2}/(total Fe as molar FeO) = 0.9, and mg of Cpx was computed after recasting of Fe^{+2} and Fe^{+3}, assuming stoichiometry. The Fo(calc) and Cpx-mg (calc) are compositions calculated from Mg′, assuming $Kd_{(FeO/MgO)}^{ol-liq} = 0.30$ (Roeder and Emslie, 1970) and $Kd_{(FeO/MgO)}^{cpx-liq} = 0.23$ (Grove *et al.*, 1982), respectively.

Fo and mg (Mg/Mg + Fe) values of the most magnesian olivine and clinopyroxene are nearly equal to those calculated from the Mg' values of the host rocks, respectively. Therefore, it is considered that these magnesian phenocrysts were crystallized as liquidus phases.

Spinel occurs as small grains in olivine phenocrysts or as discrete microphenocrysts. In the groundmass titano-magnetite occurs as small grains. Ilmenite is rarely found only in SKR14 as a groundmass phase.

Occurrence of spinel

The spinel occurs as inclusions in the certain compositional zones of olivine phenocrysts, or as discrete microphenocrysts. In SKR6 and NCH4, both types of occurrence are found. The spinel inclusions are mostly euhedral, but the discrete grains are slightly corroded. The host olivine phenocrysts do not show kink-band texture. The occurrence of the spinel is summarized in Table 2.

Chemistry of spinel

The spinel inclusions in olivine in ART8 are mostly picotite ($Al > Cr$, $3 > Fe^{+2}/Mg > 1$; according to Deer et al., 1966), and contain 19 to 33 wt% Cr_2O_3 (Table 3), whereas those in HIN11 and SKR6 are pleonaste ($Al > Cr$, $3 > Mg/Fe^{+2} > 1$), containing 11 ~ 16 wt% and 24 ~ 26 wt% Cr_2O_3, respectively. The

Table 2 Occurrence of spinel

Host rock	ART8	HIN11	SKR6	SKR14	NCH4
Inclusions in olivine					
size	$<70\mu m$	$<30\mu m$	$<80\mu m$	$<50\mu$	$<50\mu m$
color	black	dark brown ~ black	brown	black	black
shape	euhedral	euhedral	euhedral	euhedral	euhedral
host olivine*	Fo_{82} ($Fo_{89} \sim Fo_{76}$)	$Fo_{81} \sim Fo_{83}$ ($Fo_{87} \sim Fo_{78}$)	Fo_{87} ($Fo_{89} \sim Fo_{78}$)	Fo_{76} ($Fo_{76} \sim Fo_{73}$)	Fo_{76} ($Fo_{90} \sim Fo_{76}$)
chemistry	Cr-rich	Cr-rich	Cr-rich	Al-rich	Al-rich
Discrete grains					
size	(none)	(none)	$<350\mu m$	(none)	$<100\mu m$
color			dark green ~ black		dark green
shape			slightly corroded		slightly corroded
chemistry			Al-rich		Al-rich

*: Fo values of the zone including spinel grains. Those in parentheses show the whole compositional ranges of phenocrysts.

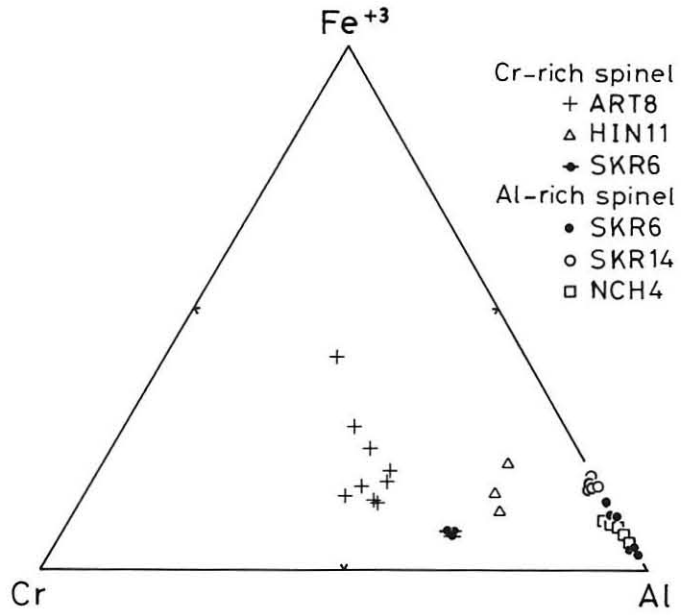
Table 3 Representative spinel analyses

No.	1	2	3	4	5	6	7	8	9
occurrence	i (82)	i	i (81)	i (87)	d	i (76)	i (76)	i (78)	d
Host rock	ART8	ART8	HIN11	SKR6	SKR6	SKR14	SKR14	NCH4	NCH4
SiO_2	0.50	0.25	0.17	0.32	0.00	0.20	0.18	0.15	0.14
TiO_2	0.68	1.42	0.67	0.70	0.05	1.32	1.36	0.79	0.72
Al_2O_3	25.82	24.87	37.72	37.12	64.78	48.68	48.57	55.47	56.99
Cr_2O_3	30.58	25.82	11.01	24.13	0.09	0.84	1.11	1.71	0.04
Fe_2O_3	10.89	15.48	17.95	6.71	3.13	14.77	14.70	8.89	8.01
FeO	20.21	20.55	18.62	14.41	15.00	20.28	22.56	17.64	18.45
MnO	0.39	0.40	0.27	0.22	0.26	0.15	0.10	0.09	0.22
NiO	0.08	0.20	0.00	0.00	0.00	0.13	0.18	0.25	0.08
MgO	10.70	10.46	12.65	15.66	17.91	13.25	12.01	15.54	14.96
CaO	0.17	0.04	0.03	0.05	0.00	0.02	0.00	0.01	0.00
total	100.02	99.49	99.09	99.32	101.22	99.64	100.77	100.54	99.61
	Numbers of ions on the basis of 32 oxygens								
Si	0.123	0.062	0.040	0.074	0.000	0.045	0.040	0.032	0.030
Al	7.511	7.327	10.457	10.061	15.492	12.832	12.794	13.964	14.407
Cr	5.967	5.103	2.048	4.388	0.014	0.149	0.196	0.289	0.007
Fe^{+3}	2.023	2.911	3.178	1.162	0.478	2.486	2.473	1.430	1.293
Ti	0.126	0.267	0.119	0.121	0.008	0.222	0.229	0.127	0.116
Mg	3.936	3.897	4.435	5.638	5.417	4.417	4.001	4.947	4.783
Fe^{+2}	4.171	4.297	3.662	2.771	2.546	3.793	4.217	3.150	3.310
Mn	0.082	0.085	0.054	0.043	0.045	0.028	0.019	0.016	0.040
Ni	0.016	0.040	—	—	0.000	0.023	0.032	0.043	0.014
Ca	0.045	0.011	0.008	0.012	0.000	0.005	0.000	0.002	0.000
mg	0.486	0.476	0.548	0.660	0.680	0.538	0.487	0.611	0.591
Cr/Cr+Al	0.443	0.411	0.164	0.304	0.001	0.011	0.015	0.020	0.000
Cr*	38.50	33.26	13.06	28.11	0.09	0.96	1.27	1.84	0.04
Al*	48.45	47.76	66.68	64.45	96.92	82.97	82.74	89.04	91.72
Fe^{+3*}	13.05	18.98	20.26	7.44	2.99	16.07	15.99	9.12	8.23

Analysis was made by using JCMA-733 electron probe microanalyzer at Hokkaido University, and corrected after Yui and Aoki (1986). The operating conditions were at 15kV and 0.02μA. Occurrence i: included in olivine phenocryst. Fo content of host olivine is shown in ( ). d: discrete grain. Total Fe was partitioned between octahedral and tetrahedral sites as Fe^{+3} and Fe^{+2} in the proportion required for stoichiometry. $Cr^* = 100Cr/(Cr+Al+Fe^{+3})$, $Al^* = 100Al/(Cr+Al+Fe^{+3})$, and $Fe^{+3*}/(Cr+Al+Fe^{+3})$.

spinel inclusions in ART8 are highest in Cr_2O_3 content among these Cr-rich spinel grains, despite of their low mg-values (Text-figs. 2 and 3).

The discrete spinel grains in SKR6 are highly aluminous (Al_2O_3 52～65 wt%) and poor in Cr_2O_3 (<1 wt%). Similarly, the spinel inclusions in SKR14 and both of the spinel inclusions and the discrete grains in NCH4 are aluminous and poor in Cr_2O_3 (Al_2O_3 48～49 wt% and 55～62 wt%; Cr_2O_3<1 wt% and <2 wt%, respectively).



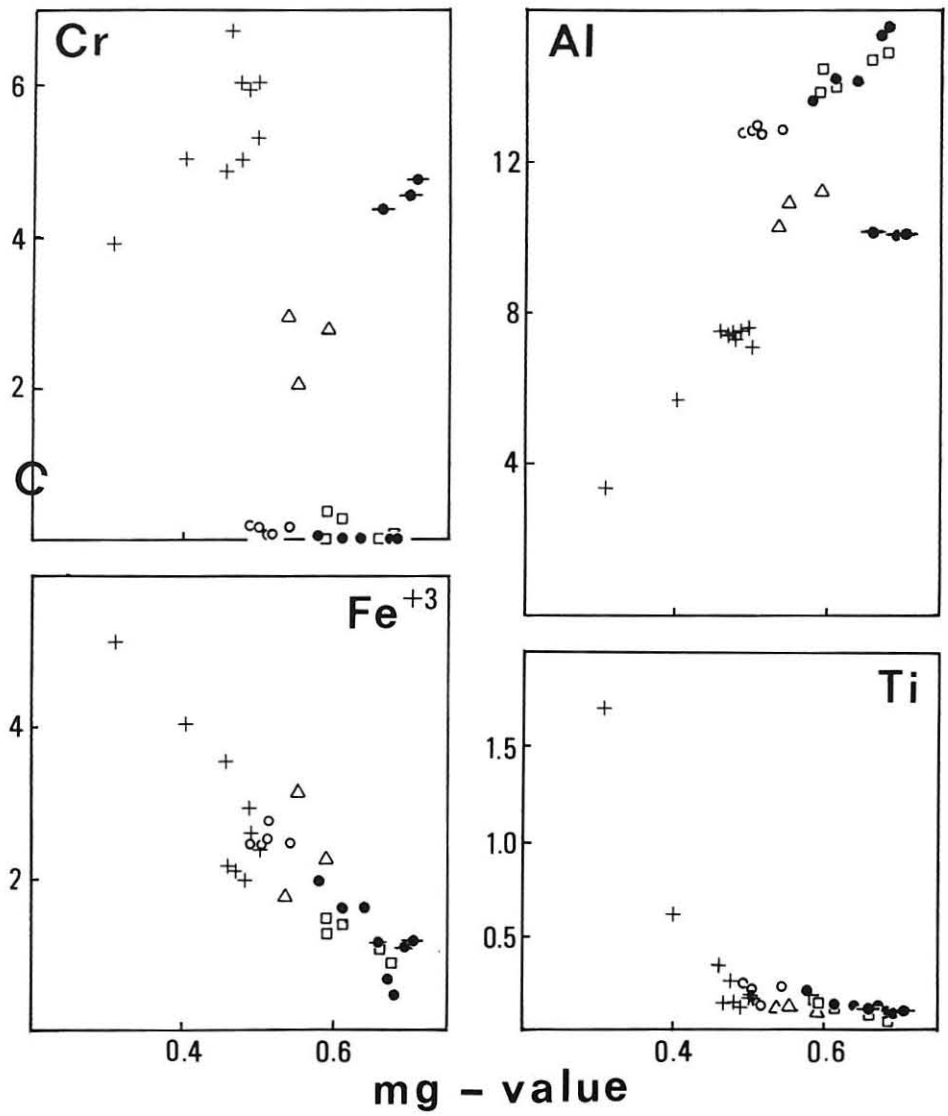
Text-fig. 2 Cr-Fe^{+3}-Al diagram of spinels from the Kibi-kogen alkali basalts.

These Al-rich spinels plot near the Al end of the Fe^{+3}-Al side in the Cr-Fe^{+3}-Al diagram (Text-fig. 2). The Al-rich spinel in SKR6 and SKR14 show a continuous compositional variation, decreasing in Al content with decrease in mg-value (Text-fig. 3). The Al-rich spinel inclusions and discrete grains in NCH4 show a similar compositional trend. This suggests that these Al-rich spinel grains are of magmatic origin. The Cr-rich spinel may be also of magmatic origin, judging from the occurrence as inclusion in the certain zones of olivine phenocrysts, and the chemical correspondence to the host basalts as discussed later.

In the Kibi-kogen alkali basalts, the olivine phenocrysts including Cr-rich spinel (Fo_{81-87}) are more magnesian than those including Al-rich spinel (Fo_{76-78}) (Table 2).

Discussion

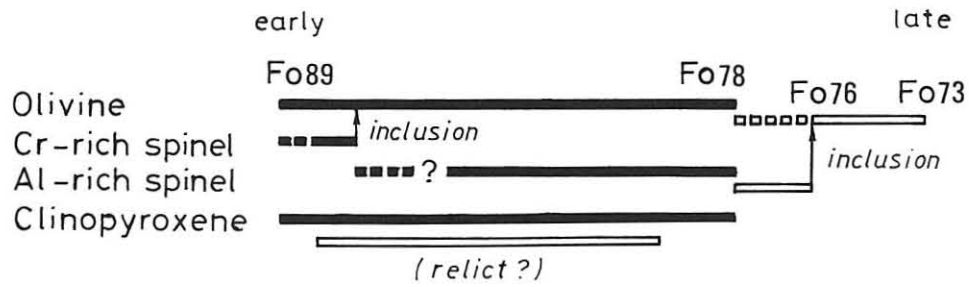
Phenocrysts of nearly Cr-free ($Cr_2O_3 < 2$ wt%), Al-rich spinel have been rarely reported from some volcanic rocks (e.g. Binns, 1969; Binns et al., 1970). Such Al-rich spinel, however, frequently occurs in ultramafic rocks (e.g. Kutolin and Frolova, 1970; Frey and Prinz, 1978; Arai and Hirai, 1985) and pelitic ~ calcareous metamorphic rocks (e.g. Stewart, 1942; Knauer et al., 1974). Therefore, as for the Al-rich spinel in the volcanic rocks containing ultramafic inclusions, it is often doubtful whether this mineral is phenocryst or xenocryst. However, as mentioned previously, both of the Cr- and Al-rich spinels in the Kibi-kogen alkali basalts are regarded as the crystallization products from the host magmas.



Text-fig. 3 Cr, Al, Fe^{+3} and Ti contents (atomic ratio) of spinel. Symbols as in Text-fig. 2.

Crystallization stage of Cr- and Al-rich spinels

In SKR6, the Cr-rich spinel is slightly higher in mg-value than the coexisting Al-rich spinel. In addition, the zone of host olivine (Fo_{87}) including Cr-rich spinel in SKR6 is more magnesian than that (Fo_{76}) including Al-rich spinel in SKR14 (Table 2). Therefore, the Cr-rich spinel is considered to have formed earlier than the Al-rich spinel during the crystallization of the SKR basalt magma (Text-fig. 4). The high Fo value of the spinel inclusion zone of host olivine in SKR6 indicates that the Cr-rich spinel is a near-



Text-fig. 4 Crystallization sequence of phenocrystic minerals in the SKR6 and SKR14 basalts. solid symbol: SKR6, open symbol: SKR14.

liquidus phase.

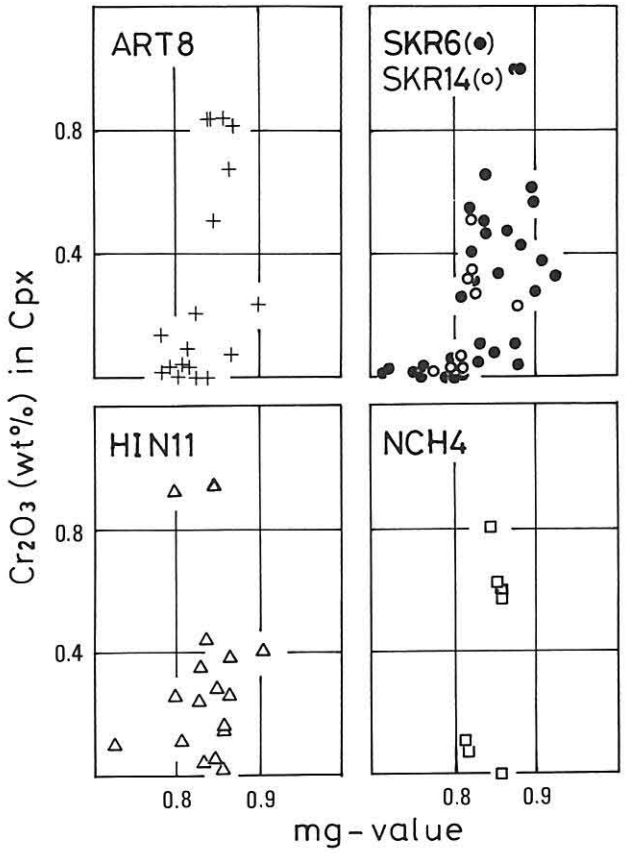
As for the Al-rich variety of spinel, high pressure origin was suggested by Irvine (1967), Green et al. (1971, 1972), Ridley et al. (1974), Sigurdsson and Schilling (1976) and Jaques and Green (1980). However, there is no evidence to indicate that the Al-rich spinel in SKR6 crystallized under higher pressures than the Cr-rich spinel. Hence, the Al-rich spinel in the Kibi-kogen alkali basalts may have formed depending largely on the chemistry of host magma rather than pressure.

Crystallization of Al-rich spinel

Crystallization of olivine + clinopyroxene ± Cr-rich spinel from the Kibi-kogen alkali basalt magma may yeild the residual liquid more aluminous and less chromian. In consistent with this, as shown in Text-figs. 5 and 6, the clinopyroxene phenocrysts in the Kibi-kogen alkali basalts decrease in Cr_2O_3 and increase in Al_2O_3 with decreasing of the mg-value. The Cr-rich spinel also decreases in Cr with decreasing of the mg-value (Text-fig. 3). At the same time, however, the Cr-rich spinel increases not in Al but in Fe^{+3} and Ti. This compositional variation suggests that the defficiency due to the depletion in Cr in the octahedral site of spinel is filled not with Al but with Fe^{+3} and Ti, probably being controlled by the f_{O_2} and/or temperature of magma (Hill and Roeder, 1974; Fiske and Bence, 1980; Batiza and Vanko, 1984). Therefore, the Al content of the Cr-rich spinel may not be controlled directly by the Al content of magma, but indirectly by the Cr content, f_{O_2} and/or temperature of magma.

The amount of depletion in Cr_2O_3 in the residual liquid can be estimated in terms of the Rayleigh fractionation model ($C_L = C_O \times F^{(D-1)}$, C_L: Cr content of the liquid, C_O: that of original magma, D: the bulk distribution coefficient, F: fraction of the liquid remaining ($0 \leqq F \leqq 1$)). The D is calculated for SKR6 which contains both of the Cr- and Al-rich spinels, using the distribution coefficients for olivine/SKR6 (1.4) and for clinopyroxene/SKR6 (15.3) and their modal proportion (0.57: 0.43). The resultant D_{Cr} value is about 7.

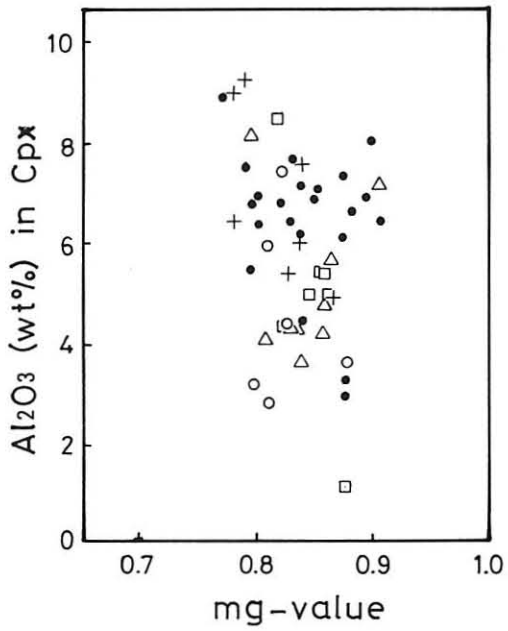
As shown in Text-fig. 7, the Cr content of the residual magma rapidly decreases as crystallization proceeds. It decreases down to 250ppm at $F = 0.91$ which corresponds to



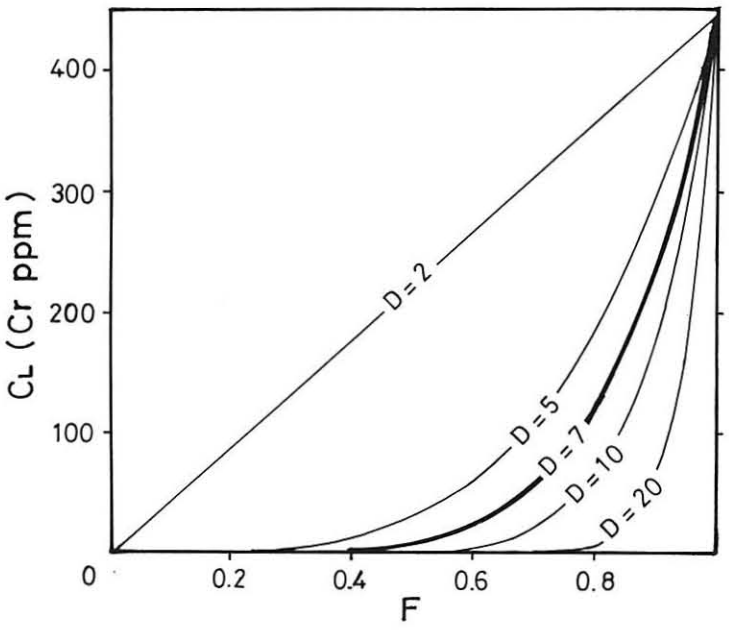
Text-fig. 5 Cr_2O_3 content vs. mg-value of clinopyroxene phenocrysts in the Kibi-kogen alkali basalts. Symbols; 1: ARTS, 2: HIN11, 3: SKR6, 4: SKR14, 5: NCH4.

the modal proportion of the groundmass of SKR6 (Table 1). The crystallization of Cr-rich spinel further promotes the depletion of Cr in the liquid because of the high distribution coefficient (230) for the Cr-rich spinel. Therefore, in case of SKR6, the Cr content of the residual magma probably decreased to around 250ppm at the time of crystallization of the Al-rich spinel.

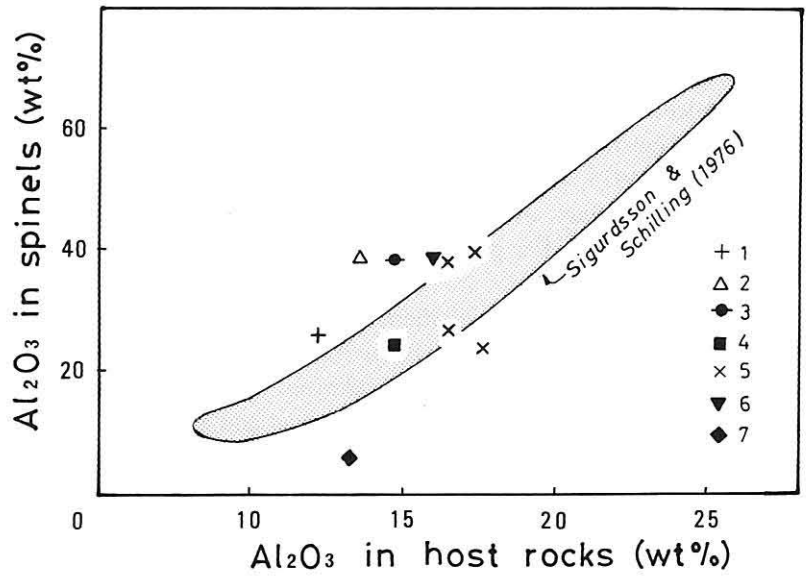
Hill and Roeder (1974) suggested that the solubility limit of chromium in basaltic liquids is approximately 200 to 300ppm at 1200°C and a log f_{O_2} of -8, and that Cr-rich spinel may not crystallize from the liquid under this solubility limit. Accordingly, in case of SKR6, when the Cr content of the residual magma decreased down to the solubility limit, the Cr-rich spinel may have ceased from crystallization, and the almost Cr-free, Al-rich spinel in turn started to crystallize as an aluminous phase. No appearance of plagioclase phenocrysts at this stage suggests that the Al-rich spinel was stable rather than plagioclase, probably under higher pressures and/or temperatures. Other Al-rich spinels in the Kibi-kogen alkali basalts may have formed through the same



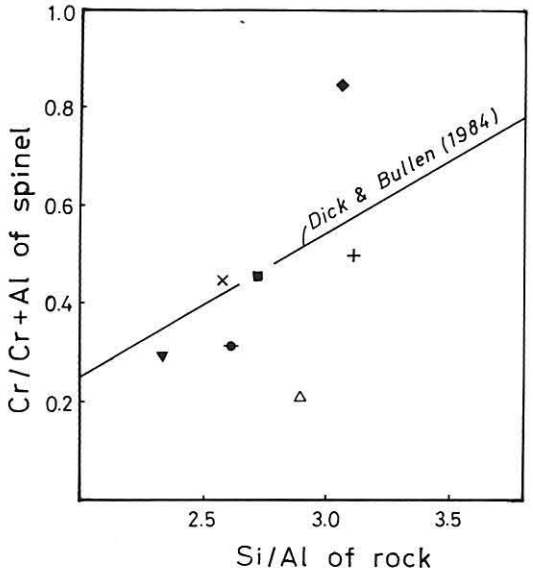
Text-fig. 6 AL_2O_3 content vs. mg-value of clinopyroxene phenocrysts. Symbols as in Text-fig. 5.



Text-fig. 7 Calculated variation of Cr in the residual liquid (C_L) with fraction (F).



Text-fig. 8 Al_2O_3 content of spinel vs. Al_2O_3 content of host rocks. Near-liquidus spinels from alkali basalt ~ basanitoid from Japan are plotted. Symbols; 1-3: Kibi-kogen alkali basalts (1: ART8, 2: HIN11, 3: SKR6), 4: Abu alkali basalt (Koyaguchi, 1986), 5: Misasa alkali basalt (Nagao et al., 1980), 6: Nanzaki basanitoid (Shiraki et al., 1979), 7: Oshima-Oshima alkali basalt (rock; Katsui et al., 1978, spinel; Yamamoto, 1980).



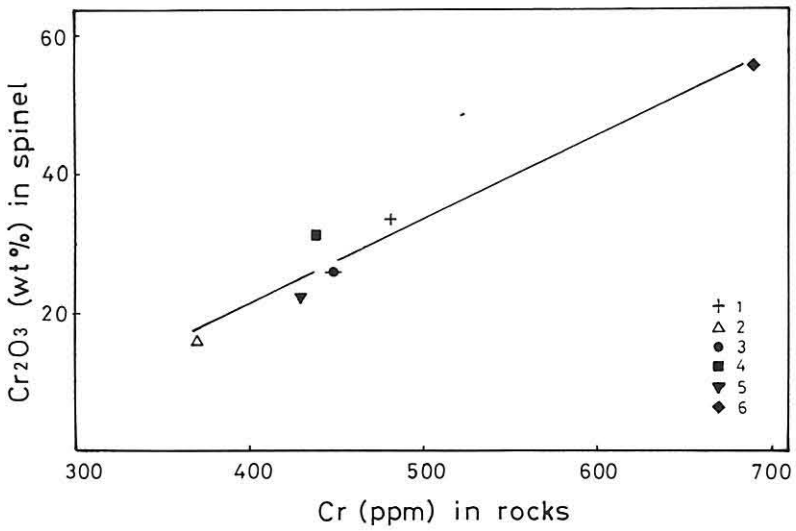
Text-fig. 9 Cr/Cr + Al of spinel vs. Si/Al of rocks. Symbols as in Text-fig. 8.

process as above.

Some comments on the composition of near-liquidus spinel

It has been noticed that liquidus spinel shows a good correlation in Al_2O_3 content with its host rock (Sigurdsson and Schilling, 1976; Nagao et al., 1980; Allan et al., 1988). Besides, Irvine (1975, 1976) suggested the effect of SiO_2 content of magma on the Cr/Cr + Al ratio of spinel, on the basis of his experimental work. Consistently, Dick and Bullen (1984) found a good correlation between rock Si/Al and spinel Cr/Cr + Al.

Text-figs. 8 and 9 show the above chemical relationships for the near-liquidus spinels from Kibi-kogen alkali basalts and those from other alkali basalts in Japan. In both figures, however, there are no good correlations between spinel and host rock Al_2O_3 contents, and between Cr/Cr + Al ratio of spinel and Si/Al ratio of host rock. On the other hand, as shown in Text-fig. 10, these spinels show a good correlation with their host rocks in Cr content. Although Allan et al. (1988) pointed that there is no good correlation between spinel and host glass Cr contents in case of the MORB-type lavas, the composition of near-liquidus spinel in the alkali basalts studied here depends largely on the Cr content of host magma.



Text-fig. 10 Cr_2O_3 in spinel vs. Cr in rocks. Near-liquidus spinels from alkali basalt ~ basanitoid from Japan are plotted. Symbols; 1-4: same as in Text-fig. 8, 5: Nanzaki basanitoid, 6: Oshima-Oshima alkali basalt (4-6: data sources are same as in Text-fig. 8).

Conclusion

1) Both Cr- and Al-rich spinels in the Kibi-kogen alkali basalts are considered to be of magmatic origin. The Cr-rich spinel was possibly crystallized as a near-liquidus phase with olivine and clinopyroxene prior to the crystallization of the Al-rich spinel.
2) The calculation in terms of the Rayleigh fractionation model (olivine + clinopyroxene) resulted in strong depletion of Cr in the residual liquid with progress of crystallization of the Kibi-kogen alkali basalt magma. Thus, when the Cr content of the liquid became less than the solubility limit of Cr, the Cr-rich spinel may have ceased from crystallization and the Al-rich spinel in turn crystallized as an aluminous phase.
3) The composition of the near-liquidus spinel from the alkali basalts in Japan depends mainly on the Cr content of host magma rather than the Al content and Si/Al ratio.

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