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PHASE RELATIONS IN THE CHROMIUM-POOR PART OF THE SYSTEM DIOPSIDE-ANORTHITE-FORSTERITE-$MgCr_2O_4$ IN AIR AT 1 ATM

by

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(with 5 tables and 8 text-figures)

Abstract

The effect of chromium on the silicate system was studied at 1 atm by adding $MgCr_2O_4$ to the join diopside-anorthite-forsterite which is a thermal divide in the system $CaO\text{-}MgO\text{-}Al_2O_3\text{-}SiO_2$. When a small amount of $MgCr_2O_4$ (0.2 – 0.5 wt%) is added to the join, the liquidus surface for spinel expands at the expense of anorthite. Thus, the assemblage diopside + forsterite + anorthite + liquid at a ternary invariant point becomes unstable and the assemblages of diopside + anorthite + spinel + liquid and diopside + forsterite + spinel + liquids appear on the liquidus surface. The points at which these assemblages are present are not "invariant" point but "piercing" point, indicating that the join diopside-anorthite-forsterite with a small amount of chromium is not thermal divide, and that the residual liquid in the crystallization can move through the join diopside-anorthite-forsterite.

Introduction

Chromium-bearing silicate systems have a wide liquidus surface for spinel (Keith, 1954; Muan, 1975), indicating that the presence of very small amount of Cr gives considerable influence on the phase relation in the silicate system. For example, spinel is a liquidus phase for the composition containing 3 wt% of $CaCrAlSiO_6$ (1 wt%' Cr_2O_3) in the join diopside-$CaCrAlSiO_6$ (Dickey et al., 1971). Irvine (1975) has pointed out the significance of Cr in the formulation of petrologic models. Thus, Cr should be taken into account in the discussion of petrogenetic models.

For this reason, the experimental study of the effect of Cr on the phase relation in the system $CaO\text{-}MgO\text{-}Al_2O_3\text{-}SiO_2$, which is an important model system in igneous petrology, has been performed. In this paper preliminary data of the join diopside-anorthite-forsterite at 1 atm, which is a thermal divide in the system $CaO\text{-}MgO\text{-}Al_2O_3\text{-}SiO_2$, are presented and their petrologic significance is briefly discussed.

Oxidation state of Cr (Cr^{2+}, Cr^{3+} and C^{6+}) is always problematical in experimental petrology as discussed by Schreiber (1978) and Ikeda and Yagi (1978), although several experimental studies were carried out in air (Keith, 1954; Dickey et al., 1971; Ikeda and Yagi, 1972, 1977; Onuma and Tohara, 1981) as well as present study. Cr redox state is affected by oxygen fugacity, temperature, and composition of melt (Schreiber and Haskin, 1976). However, fortunately the data of Cr redox state in the join diopside-anorthite-forsterite with 0.5 wt% Cr_2O_3 are available. Schreiber and Haskin (1976) demonstrated that at about $10^{-0.7}$ atm fO_2 (prevailing atmosphere of air) 90% of Cr are in Cr^{3+} state at

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1500°C, and that the ratio of Cr^{3+} to total Cr increases with falling temperature. Since the present experiment was undertaken below 1500°C, more than 90% of Cr^{3+} may be expected. Of course it is needed to examine the effect of minor concentration of other redox state of Cr in establishing precice phase boundary in the silicate system. Nevertheless, the results obtained in the experiments in air provide some important information on the petrology, because terrestrial basalts have formed under the conditions that fabor the presence of Cr^{3+} (Burns and Burns, 1975).

Phase Relations

Starting materials were prepared by crystallizing glasses at 900°C, which were made by melting the mixtures composed of $CaCO_3$, MgO, Cr_2O_3, Al_2O_3, and SiO_2 at 1400–1450°C in a Pt90-Rh10 crucible in air. The color of glasses change from light green to dark green as Cr content increases, while the crystalline mixtures show slightly bluish gray, except those in diopside-forsterite join which are deep blue. The experiments were performed at 1 atm in air by the quenching method. Since Cr is mainly fixed in spinel, it was added to the system as a form of $MgCr_2O_4$.

In the system clinopyroxene, anorthite, forsterite, and spinel are encountered. Although the compositions of these minerals have not analysed yet, judging from the X-ray diffraction patterns, the clinopyroxene is chromian diopside, the spinel is a solid solution between $MgAl_2O_4$ and $MgCr_2O_4$, and anorthite and forsterite have nearly pure composition.

Chromian pyroxene components would be formed as follows:

$$\underset{\text{diopside}}{CaMgSi_2O_6} + MgCr_2O_4 = \underset{\text{pyroxene}}{CaCr_2SiO_6} + Mg_2SiO_4$$

$$\underset{\text{anorthite}}{CaAl_2Si_2O_8} + MgCr_2O_4 = \underbrace{CaCrAlSiO_6 + MgCrAlSiO_6}_{\text{pyroxene}}$$

The clinopyroxenes crystallized from the Al-bearing compositions are pale green in the temperature range 1350-1250°C, while those crystallized from the compositions in the diopside-forsterite join blue even at higher temperature, say around 1300°C. An interpretation of this sort of phenomena is given by Ikeda and Yagi (1977) and Ikeda (1981).

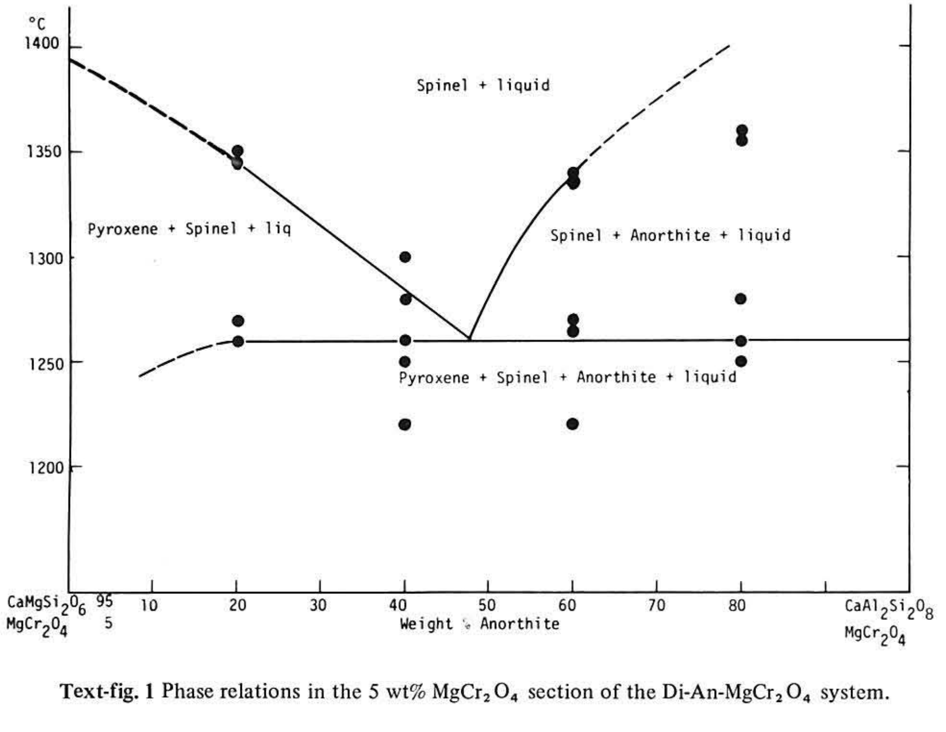
The join diopside-anorthite-$MgCr_2O_4$

The results of experiment are given in Tables 1, 2, and 3. Text-figs. 1, 2, and 3 show the phase relation in the sections with 5, 1, and 0.5 wt% $MgCr_2O_4$. In the 5% $MgCr_2O_4$ section spinel is the first crystalline phase to appear throughout all the compositions studied. Clinopyroxene crystallizes next to spinel in the Di ($CaMgSi_2O_6$)-rich region of this section, while anorthite in the An($CaAl_2Si_2O_8$)-rich region. The assemblage clinopyroxene + anorthite + spinel + liquid is present at 1260 ±5°C in all the compositions.

In the 1% $MgCr_2O_4$ section, the liquidus for clinopyroxene appears and crosses with that of spinel at 1360 ±5°C and the clinopyroxene + spinel + liquid region is considerably decreased compared to that in the 5% $MgCr_2O_4$ section. The clinopyroxene + anorthite + spinel + liquid region is also decreased into the temperature interval of 5°C. The phase diagram of the 0.5% $MgCr_2O_4$ section was tentatively constructed from preliminary data. The liquidii for spinel and clinopyroxene cross with each other at 1300°C ±5°C. The

Table 1 Runs in the 5 wt% $MgCr_2O_4$ section of the system diopside-anorthite-$MgCr_2O_4$

Composition (wt%)		Temp. (°C)	Time (h)	Phases Present
$CaMgSi_2O_6$	$CaAl_2Si_2O_8$			
75	30	1350	72	*sp + gl*
		1345	48	*cpx + sp + gl*
		1270	168	*cpx + sp + gl*
		1260	168	*cpx + sp + an + gl*
55	40	1300	72	*sp + gl*
		1280	168	*sp + gl*
		1260	168	*cpx + sp + gl*
		1250	168	*cpx + sp + an + gl*
		1220	336	*cpx + an + sp + gl*
35	60	1350	18	*sp + gl*
		1345	45	*an + sp + gl*
		1270	168	*an + sp + gl*
		1260	168	*cpx + an + sp + gl*
		1220	336	*cpx + an + sp + gl*
15	80	1360	16	*an + sp + gl*
		1355	45	*an + sp + gl*
		1280	168	*an + sp + gl*
		1260	168	*an + sp + gl*
		1250	168	*cpx + an + sp + gl*



Text-fig. 1 Phase relations in the 5 wt% $MgCr_2O_4$ section of the Di-An-$MgCr_2O_4$ system.

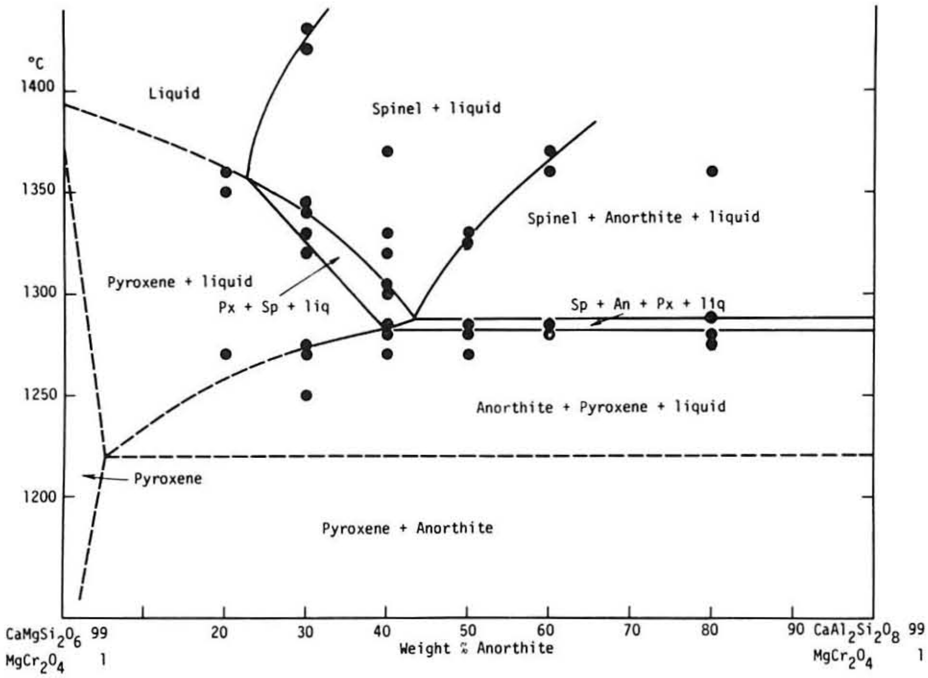
Table 2 Runs in the 1 wt% $MgCr_2O_4$ section of the system diopside-anorthite-$MgCr_2O_4$

Composition (wt%)		Temp. (°C)	Time (h)	Phases Present
$CaMgSi_2O_6$	$CaAl_2Si_2O_8$			
79	20	1360	16	*gl*
		1350	18	*cpx + gl*
		1270	168	*cpx + gl*
69	30	1430	16	*gl*
		1420	16	*sp + gl*
		1345	18	*sp + gl*
		1340	18	*cpx + sp + gl*
		1330	18	*cpx + sp + gl*
		1320	18	*cpx + gl*
		1270	168	*cpx + gl*
		1270	258	*cpx + an + gl*
		1250	258	*cpx + an + gl*
59	40	1370	18	*sp + gl*
		1305	20	*sp + gl*
		1300	24	*cpx + sp + gl*
		1285	45	*cpx + sp + gl*
		1280	70	*cpx + gl*
		1270	168	*cpx + (an) + gl*
49	50	1325	24	*sp + gl*
		1320	20	*an + sp + gl*
		1285	72	*an + sp + gl*
		1280	258	*cpx + an + gl*
		1250	258	*cpx + an + gl*
39	60	1270	3	*sp + gl*
		1360	18	*an + sp + gl*
		1285	48	*an + sp + gl*
		1280	72	*cpx + an + gl*
19	80	1350	72	*sp + an + gl*
		1285	72	*an + sp + gl*
		1280	48	*cpx + an + sp + gl*
		1275	72	*cpx + an + gl*

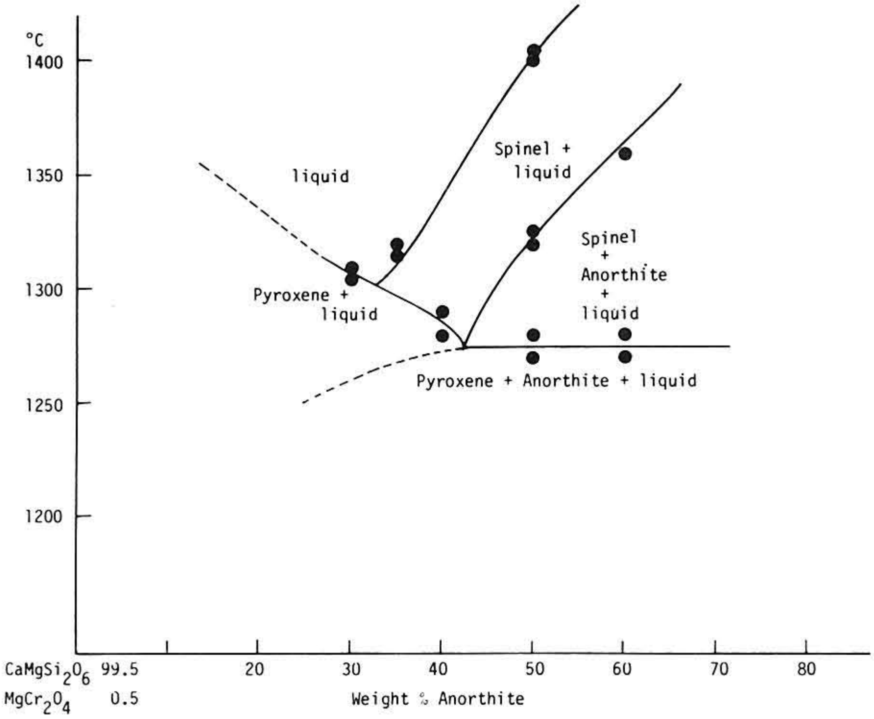
assemblages clinopyroxene + spinel + liquid and clinopyroxene + anorthite + spinel + liquid do not have temperature interval within the experimental error, indicating that the spinel is consumed by the reaction with liquid to form clinopyroxene. The temperature at which clinopyroxene, anorthite, and spinel coexist with liquid decreases with increasing Cr content of the system throughout the three sections. It is also noticed that clinopyroxene + anorthite + spinel + liquid region becomes narrow with a decrease in $MgCr_2O_4$ component, and finally may be converged to a line. The temperatures at which the spinel disappears and clinopyroxene appears are almost constant throughout all the compositions in the 0.5% $MgCr_2O_4$ section. This means that, although the system diopside-anorthite-$MgCr_2O_4$ is pseudoternary, the assemblage clinopyroxene + anorthite + spinel + liquid can be treated like an invariant point (peritectic point) in the portion extremely poor in Cr. Therefore, when some other component (Mg_2SiO_4) is added to this system, this assemblage may behave like an "univariant" curve in quaternary system.

Table 3 Runs in the 0.5 wt% $MgCr_2O_4$ section of the system diopside-anorthite-$MgCr_2O_4$

Composition (wt%)		Temp. (°C)	Time (h)	Phases Present
$CaMgSi_2O_6$	$CaAl_2Si_2O_8$			
69.5	30.0	1310	18	*gl*
		1305	18	*cpx + gl*
64.5	35.0	1320	18	*gl*
		1315	18	*tr. sp + gl*
59.5	40.0	1330	18	*sp + gl*
		1315	5	*sp + gl*
		1290	48	*sp + gl*
		1280	48	*cpx + gl*
49.5	50.0	1405	45	*gl*
		1400	18	*sp + gl*
		1325	20	*sp + gl*
		1320	18	*an + sp + gl*
		1280	72	*an + sp + gl*
		1270	72	*cpx + an + gl*
39.5	60.0	1360	5	*an + sp + gl*
		1280	48	*an + sp + gl*
		1270	72	*cpx + an + gl*



Text-fig. 2 Phase relations in the 1 wt% $MgCr_2O_4$ section of the Di-An-$MgCr_2O_4$ system.



Text-fig. 3 Phase relations in the 0.5 wt% $MgCr_2O_4$ section of the Di-An-$MgCr_2O_4$ system.

Liquidus phase relations in the system diopside-anorthite-forsterite-$MgCr_2O_4$

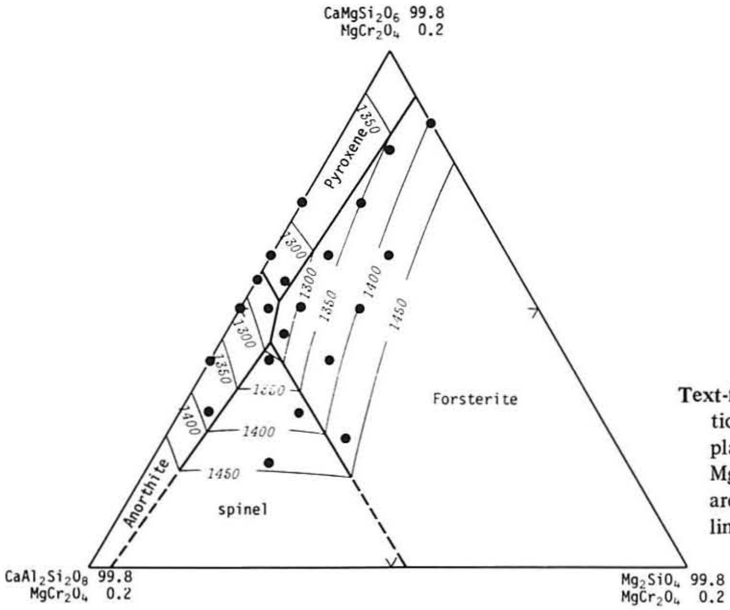
The liquidus diagrams of the 0.2 and 0.5 wt% $MgCr_2O_4$ (0.16 and 0.40 wt% Cr_2O_3) planes are shown in Text-fugs. 4 and 5, respectively. The results of quenching experiments are given in Tables 4 and 5.

On the 0.2% plane (Text-fig. 4), clinopyroxene, anorthite, forsterite, and spinel are encountered as primary phase. There are two points showing four phase assemblage: one, at 1265 ±5°C, shows the liquid coexisting with clinopyroxene, anorthite and forsterite, and the other, at 1270 ±5°C, the liquid coexisting with anorthite, forsterite, and spinel. The former point is treated as an "eutectic point" in the ternary system, becuase the compositions around this point crystallize into the assemblage clinopyroxene + forsterite + anorthite at a same temperature 1265° ±5°C within the experimental error. While the latter point behaves like "piercing point", through which an "univariant curve" anorthite + forestrite + spinel + liquid of the quaternary system passes. Along this "univariant curve" the spinel reacts with liquid to form clinopyroxene.

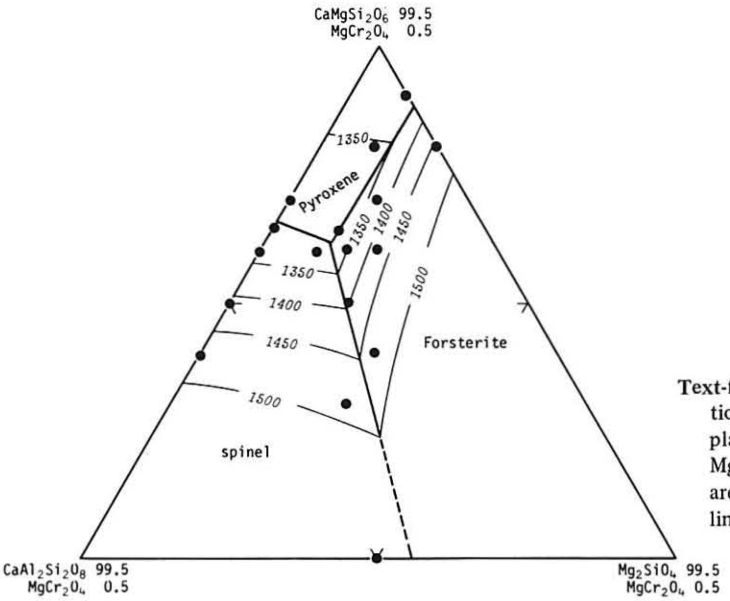
Text-fig. 5 shows the liquidus diagram of 0.5 wt% $MgCr_2O_4$ plane. On the liquidus diagram clinopyroxene, forsterite and spinel are present, but anorthite absent. The primary fields for clinopyroxene, spinel and forsterite cross with each other at 1305° ±5°C. This point, however, behaves like piercing point, and an "univariant curve" clinopyroxene +

Table 4 Runs on the 0.2 wt% $MgCr_2O_4$ plane of the system diopside-anorthite-forsterite-$MgCr_2O_4$

Composition (wt%)			Temp. (°C)	Time (h)	Phases Present
$CaMgSi_2O_6$	$CaAl_2Si_2O_8$	Mg_2SiO_4			
84.8	0	15.0	1390	18	*fo + gl*
79.8	10.0	10.0	1355	18	*gl*
			1350	18	*fo + gl*
69.8	20.0	10.0	1340	22	*gl*
			1335	18	*fo + gl*
			1330	22	*cpx + fo + gl*
69.8	30.0	0	1330	22	*gl*
			1325	22	*cpx + gl*
59.8	20.0	20.0	1425	40	*gl*
			1420	18	*fo + gl*
59.8	30.0	10.0	1325	22	*gl*
			1320	18	*fo + gl*
59.8	40.0	0	1275	18	*gl*
			1270	18	*cpx + gl*
			1260	72	*an + cpx + gl*
54.8	40.0	5.0	1280	48	*gl*
			1275	48	*cpx + gl*
			1260	72	*an + cpx + gl*
54.8	45.0	0	1280	48	*gl*
			1275	48	*an + gl*
			1260	72	
49.8	30.0	20.0	1395	18	*gl*
			1390	18	*fo + gl*
49.8	40.0	10.0	1295	18	*gl*
			1290	40	*fo + gl*
			1270	72	*fo + gl*
			1265	72	*cpx + an + fo + gl*
49.8	45.0	5.0	1275	18	*gl*
			1270	18	*an + gl*
49.8	50.0	0	1320	22	*gl*
			1315	22	*an + gl*
			1270	72	*an + gl*
			1260	72	*cpx + an + gl*
44.8	45.0	10.0	1275	18	*gl*
			1270	18	*fo + gl*
39.8	40.0	20.0	1375	18	*gl*
			1370	18	*fo + gl*
39.8	50.0	10.0	1315	22	*gl*
			1310	22	*sp + gl*
			1270	72	*sp + gl*
			1265	72	*cpx + an + fo + gl*
39.8	60.0	0	1380	18	*gl*
			1375	18	*an + gl*
			1270	72	*an + gl*
			1260	72	*cpx + an + gl*
29.8	50.0	20.0	1365	22	*gl*
			1360	22	*sp + gl*
			1325	18	*sp + gl*
			1320	72	*fo + sp + gl*
29.8	65.0	5.0	1390	18	*gl*
			1380	18	*an + gl*
24.8	45.0	30.0	1420	3	*gl*
			1410	4	*fo + gl*
			1400	18	*fo + gl*
			1380	18	*sp + fo + gl*
19.8	60.0	20.0	1420	3	*sp + gl*
			1310	72	*sp + gl*
			1305	72	*an + sp + gl*



Text-fig. 4 Liquidus phase relations on the 0.2 wt% $MgCr_2O_4$ plane of the Di-An-Fo-$MgCr_2O_4$ system. Heavy lines are boundary curve and light lines isotherm.



Text-fig. 5 Liquidus phase relations on the 0.5 wt% $MgCr_2O_4$ plane of the Di-An-Fo-$MgCr_2O_4$ system. Heavy lines are boundary curve and light lines isotherm.

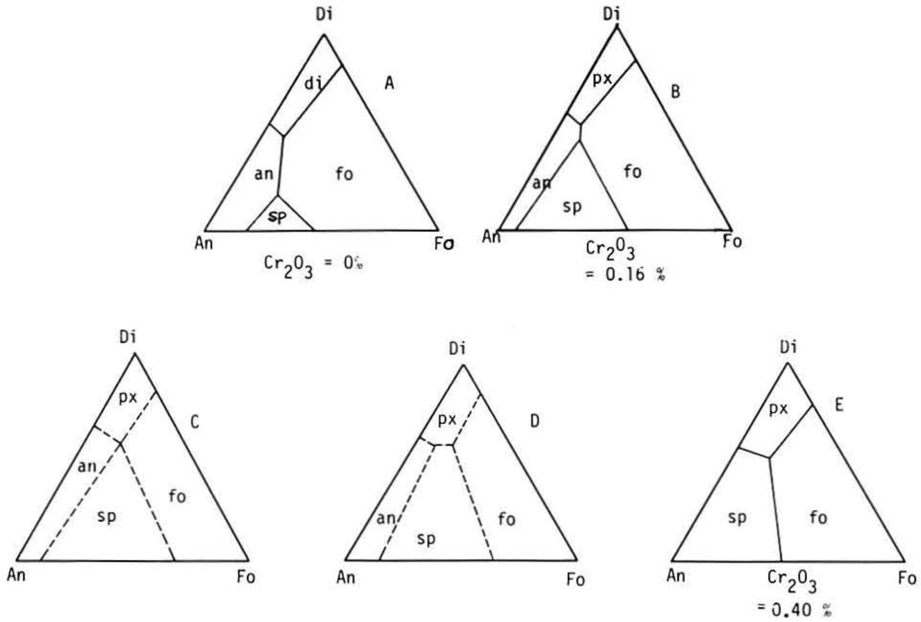
spinel + forsterite + liquid passes through this point. Clinopyroxene and spinel are in recation relation along this curve, therefore the spinel present at liquidus disappears and clinopyroxene appears at subliquidus temperatures.

Table 5 Runs on the 0.5 wt% $MgCr_2O_4$ plane of the system diopside-anorthite-forsterite-$MgCr_2O_4$

Composition (wt%)			Temp. (°C)	Time (h)	Phases Present
$CaMgSi_2O_6$	$CaAl_2Si_2O_8$	Mg_2SiO_4			
89.5	0	10.0	1390	18	*gl*
			1385	20	*cpx* + *gl*
84.5	0	15.0	1425	18	*gl*
			1420	5	*fo* + *gl*
			1350	48	*cpx* + *fo* + *gl*
79.5	0	20.0	1460	20	*gl*
			1455	20	*fo* + *gl*
79.5	10.0	10.0	1340	20	*gl*
			1335	45	*cpx* + *gl*
69.5	15.0	15.0	1355	45	*gl*
			1350	24	*fo* + *gl*
			1340	24	*fo* + *gl*
			1330	24	*cpx* + *fo* + *gl*
64.5	25.0	10.0	1330	22	*gl*
			1325	18	*fo* + *cpx* + *gl*
59.5	20.0	20.0	1420	20	*gl*
			1415	20	*fo* + *gl*
59.5	25.0	15.0	1360	18	*gl*
			1355	18	*fo* + gl
			1330	24	*fo* + *gl*
			1310	48	*cpx* + *fo* + *gl*
59.5	30.0	10.0	1320	18	*gl*
			1315	20	*sp* + *gl*
			1300	48	*sp* + *gl*
			1295	20	*cpx* + *gl*
49.5	30.0	20.0	1405	18	*gl*
			1400	24	*sp* + *fo* + *gl*
			1330	5	*sp* + *fo* + *gl*
			1325	24	*cpx* + *fo* + *gl*
39.5	30.0	30.0	1470	5	*gl*
			1460	20	*fo* + *gl*
			1450	18	*fo* + *gl*
			1430	18	*sp* + *fo* + *gl*
39.5	40.0	30.0	1470	5	*gl*
			1460	3	*sp* + *gl*
			1430	5	*fo* + *sp* + *gl*
			1310	24	*fo* + *sp* + *gl*
			1305	24	*an* + *fo* + *sp* + *gl*

Effect of Chromium on the Liquidus Phase Relations on the Join Diopside-Anorthite-Forsterite

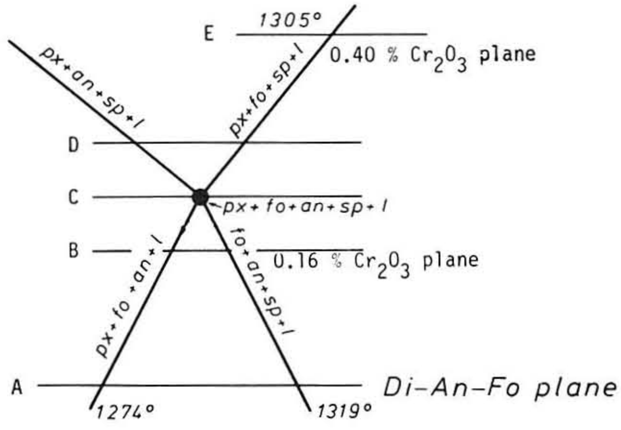
The change of liquidus phase relations on the join diopside-forsterite-anorthite with increasing $MgCr_2O_4$ content from 0 to 0.5 wt% (0.4 wt% Cr_2O_3) is demonstrated in Text-fig. 6. The join diopside-forsterite-anorthite forms a thermal divide in the quaternary system forsterite-diopside-CaTs-SiO_2 (Presnall et al., 1978). On the 0.5 wt% $MgCr_2O_4$ plane, however, the point having the "invariant" assemblage clinopyroxene + anorthite +



Text-fig. 6 Change of the liquidus phase relations on the planes at various Cr content of the Di-An-Fo-$MgCr_2O_4$ system. Cr increases in the order of A, B, C, D, and E. A is quoted from Osborn and Tait (1957). B and E are present work. Di, $CaMgSi_2O_6$; An, $CaAl_2Si_2O_8$; Fo, Mg_2SiO_4; px, clinopyroxene; an, anorthite; sp, spinel; fo, forsterite.

forsterite + liquid is not observed and the assemblage clinopyroxene + forsterite + spinel + liquid appears. The latter assemblage does not represent the "invariant" assemblage of ternary system, but "univariant" assemblage of quaternary system, suggesting that the join is no longer thermal divide.

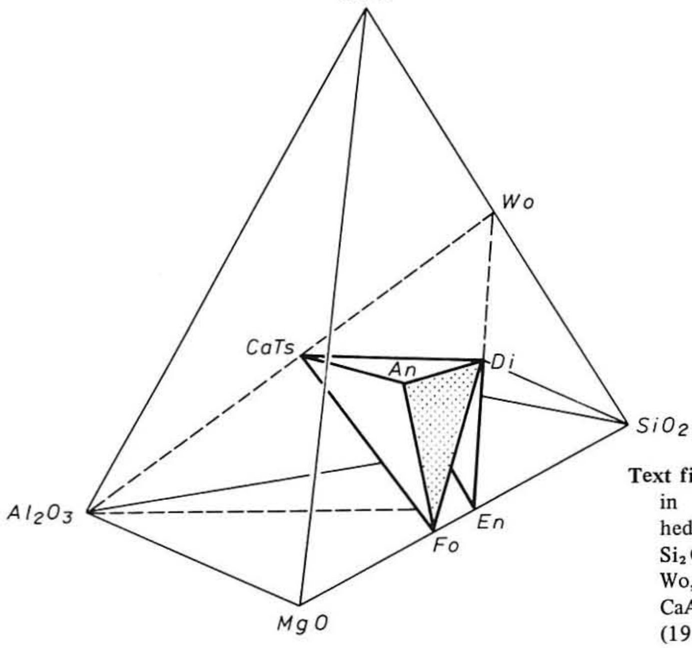
The results obtained in this study show, although the present system belongs to the quinary system CaO-MaO-Al_2O_3-Cr_2O_3-SiO_2, that as Cr_2O_3 content is very low the system can be treated as the quaternary system CaO-MgO-Al_2O_3-SiO_2, and that the spinel field increases and the anorthite field decreases considerably with increasing Cr content. Arculus et al. (1974) demonstrated in the study of the system MgO-iron-oxide-Cr_2O_3-SiO_2 that the pyroxene-silica boundary surface is bulged toward silica compared to the Cr-free system. Whereas in the present system bulging of phase boundary with the addition of Cr is not observed. The phase relation is schematically shown in Text-fig. 7. Judging from this relation, when Cr_2O_3 content is increased, the point showing the liquid coexisting with clinopyroxene, anorthite, spinel and forsterite is expected between 0.16 and 0.40 wt% as demonstrated in Text-fig. 6C, which is an "quaternary invariant point". Increasing chromium content, the plane cutting two "univariant curves", clinopyroxene + forsterite + spinel + liquid and clinopyroxene + anorthite + spinel, is expected to be as shown in Text-fig. 6D. This plane is no longer thermal divide, and therefore the Cr content of the plane of Text-fig. 6C, a little higher than 0.16 wt% in Cr_2O_3, is critical for the join diopside-anorthite-forsterite to behave as thermal divide.



Text-fig. 7 Schematic diagram showing the relationship between invariant point and univariant curves, and the planes shown in Text-fig. 6 as A, B, C, D, and E correspond to those in Text-fig. 7. px, clinopyroxene; an, anorthite; sp, spinel; fo, forsterite; l, liquid.

Petrologic Significance

The system CaO-MgO-Al_2O_3-SiO_2 is an important model to elucidate genesis and differentiation of basaltic magma (Schairer and Yoder, 1970; O'Hara and Bigger, 1969). Presnall et al. (1978, 1979) paid special attention on the sub-system Di-Fo-SiO_2-CaTs and called simplified basalt tetrahedron (Text-fig. 8). The join diopside-anorthite-forsterite is a thermal barier, dividing the system diopside-anorthite-forsterite into the SiO_2-saturated part and the SiO_2-undersaturated part and Yoder and Tilley (1962) proposed



Text fig. 8 Simplified basalt tetrahedron in the CaO-MgO-Al_2O_3-SiO_2 tetrahedron. Di, $CaMgSi_2O_6$; An, $CaAl_2Si_2O_8$; Fo, Mg_2SiO_4; En, $MgSiO_3$; Wo, $CaSiO_3$; Sp, $MgAl_2O_4$; CaTs, $CaAl_2SiO_6$. After Presnall et al. (1978)

the expanded basalt tetrahedron, Qz-Ol-Di-Ne, as a fundamental model for basaltic magma, and showed that this tetrahedron is divided into two parts by the join clinopyroxene-plagioclase-olivine at 1 atm; one represents tholeiite and the other alkali basalt. They discussed on this basis that tholeiite can not be derived from alkali basalt magma by fractional crystallization at shallower depth and vice versa. On the other hand, Presnall et al. (1978) made a high pressure experiment on the join diopside-anorthite-forsterite and clarified that this join can not be thermal divide at the pressure higher than 4 kbar, and discussed that tholeiite magma can be derived from alkali olivine basalt magma above the pressure of 4 kbar.

The experimental results in the present study clearly show that, when a small amount of Cr, about 0.2 wt% in Cr_2O_3, is present, the join diopside-anorthite-forsterite is no longer thermal divide even at 1 atm. This value of Cr_2O_3 content is expected in natural rocks (Cox and Jamieson, 1974; Dietrich, 1981). Therefore, Cr is effective to lower the critical pressure, 4 kbar, shown by Presnall et al. (1978), and it is possible to derive tholeiite magma from alkali basalt magma by fractional crystallization below 4 kbar, even at 1 bar.

The effect of Cr on the system diopside-forsterite-anorthite-SiO_2 is very important in the discussion on partial melting of the mantle. As stated before spinel field expands by the addition of Cr, while anorthite field deminishes, resulting in disappearance of the assemblage clinopyroxene + anorthite + forsterite + liquid. This gives considerable influences on the relationship among the "invariant" points and "univariant" curves; in other words, the chemical composition of liquid generated by partial melting of the mantle would be influenced by Cr content.

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