

SIGNIFICANCE OF THE SYSTEM  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ -  
 $\text{CaTiAl}_2\text{O}_6$  TO Ca-AL-RICH INCLUSIONS IN  
CARBONACEOUS CHONDRITES

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**Abstract:** The system  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$  was found to be significant for elucidating the crystallization of Ca-Al-rich inclusions in carbonaceous chondrites. Two clinopyroxenes(ss) (ss: solid solution) were found in the present system: One is Ti-bearing diopside(ss) and the other is Ti-pyroxene(ss) containing  $\text{TiO}_2$  up to 8.7 wt.%. The chemical composition of this Ti-pyroxene(ss) is very close to that of clinopyroxene found in Ca-Al-rich inclusions of the Allende meteorite. An invariant point was confirmed at 1230°C where the Ti-pyroxene coexists with spinel, perovskite, anorthite, melilite, and liquid. The assemblage of the crystalline phases is the same as that of Ca-Al-rich inclusions of the Allende meteorite. The bulk chemical compositions of the Type B inclusions of the Allende meteorite (GROSSMAN, 1975) are calculated in terms of  $\text{CaMgSi}_2\text{O}_6$ ,  $\text{CaAl}_2\text{SiO}_6$ ,  $\text{CaTiAl}_2\text{O}_6$  and  $\text{MgAl}_2\text{O}_4$  (spinel), and projected to the present system. The possible formation mechanism of the inclusions is discussed with reference to the crystallization of the system.

## 1. Introduction

Ti-Al-rich pyroxenes were found in Ca-Al-rich inclusions of the Allende meteorite and the  $\text{TiO}_2$  content attains as much as 17 wt.% (MASON, 1974; GROSSMAN, 1975). The constituent molecules of these Ti-Al-rich pyroxenes are  $\text{CaMgSi}_2\text{O}_6$  (diopside),  $\text{CaAl}_2\text{SiO}_6$ , and  $\text{CaTiAl}_2\text{O}_6$ , and therefore the system  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$  is significant to the discussion on the formation of Ca-Al-rich inclusions in the carbonaceous chondrites.

## 2. Phase Diagram

Fig. 1 shows the liquidus diagram of the system  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$  at 1 atm. The detailed study on this system is presented elsewhere (ONUMA and KIMURA, 1978). Spinel ( $\text{MgAl}_2\text{O}_4$ ), perovskite ( $\text{CaTiO}_3$ ), forsterite ( $\text{Mg}_2\text{SiO}_4$ ), and anorthite ( $\text{CaAl}_2\text{Si}_2\text{O}_8$ ) appear as primary phases and it is noticed that the primary field of clinopyroxene(ss) (ss: solid solution) is divided into two fields by the forsterite primary field, indicating that the two types of clinopyroxenes(ss)

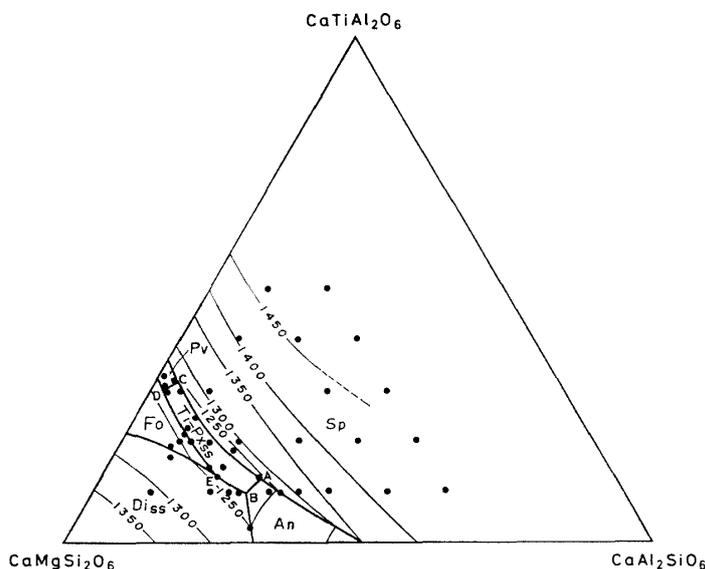


Fig. 1. Liquidus diagram of the system  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$ . *Di<sub>ss</sub>*, diopside<sub>ss</sub>; *Fo*, forsterite; *An*, anorthite; *Ti-Px<sub>ss</sub>*, Ti-pyroxene<sub>ss</sub>; *Pv*, perovskite; *Sp*, spinel.

crystallize as the primary phases. One is Ti-bearing diopside(ss) ( $\text{TiO}_2 < 4$  wt.%) and the other is Ti-rich pyroxene(ss) ( $\text{TiO}_2 > 4$  wt.%).

The system  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$  is a part of the quinary system  $\text{CaO}$ - $\text{MgO}$ - $\text{Al}_2\text{O}_3$ - $\text{TiO}_2$ - $\text{SiO}_2$ , so that the points A, B, C, D and E in Fig. 1 where three crystalline phases coexist with liquid are neither piercing points nor invariant points. The temperatures of these points show the beginning of the divariant assemblage in the system. The temperatures and compositions of these points are:

- A: 1235°C, [ $\text{CaMgSi}_2\text{O}_6(60)\text{CaAl}_2\text{SiO}_6(27)\text{CaTiAl}_2\text{O}_6(13)$ ]
- B: 1324°C, [ $\text{CaMgSi}_2\text{O}_6(64)\text{CaAl}_2\text{SiO}_6(26)\text{CaTiAl}_2\text{O}_6(10)$ ]
- C: 1260°C, [ $\text{CaMgSi}_2\text{O}_6(65)\text{CaAl}_2\text{SiO}_6(3)\text{CaTiAl}_2\text{O}_6(32)$ ]
- D: 1235°C, [ $\text{CaMgSi}_2\text{O}_6(68)\text{CaAl}_2\text{SiO}_6(2)\text{CaTiAl}_2\text{O}_6(30)$ ]
- E: 1240°C, [ $\text{CaMgSi}_2\text{O}_6(68)\text{CaAl}_2\text{SiO}_6(17)\text{CaTiAl}_2\text{O}_6(15)$ ].

To understand the phase relations below the liquidus temperature, two sections are shown in Figs. 2 and 3. Forsterite is present as a primary phase, but is consumed within a narrow temperature range by the reaction with liquid. Perovskite has a very small primary field, but crystallizes in the area rich in  $\text{CaTiAl}_2\text{O}_6$  component (more than 10 wt.%) at the subliquidus and subsolidus temperatures. Although melilite does not appear as a primary phase, it crystallizes below 1300°C. Spinel has a wide crystallization range from the liquidus temperatures 1260°-1500°C to the solidus temperatures at about 1210°-1230°C. In the  $\text{CaAl}_2\text{SiO}_6$  and  $\text{CaTiAl}_2\text{O}_6$  component rich regions it is followed by perovskite, anorthite or melilite depending on the starting compositions, and finally Ti-pyroxene(ss) joins



60 wt.%. In the fields between these compositions, there are regions where the two pyroxenes(ss) are coexisting.

Since the present system belongs to the quinary system  $\text{CaO-MgO-Al}_2\text{O}_3\text{-TiO}_2\text{-SiO}_2$ , a six-phase assemblage  $\text{Ti-pyroxene(ss)+anorthite+spinel+perovskite+melilite+liquid}$  at  $1210^\circ\text{C}$  is an isobaric invariant assemblage. Three univariant assemblages around this invariant point are confirmed: (1)  $\text{spinel+melilite+anorthite+perovskite+liquid}$ , (2)  $\text{Ti-pyroxene(ss)+anorthite+melilite+spinel+liquid}$  and (3)  $\text{Ti-pyroxene(ss)+anorthite+melilite+perovskite+liquid}$ . Due to the solid solutions of the  $\text{Ti-pyroxene(ss)}$  and the melilite, crystallization sometimes ceases on the univariant lines as shown in the sections in Figs. 2 and 3, and in the section of 10 wt.%  $\text{CaTiAl}_2\text{O}_6$  the phase assemblage  $\text{Ti-pyroxene(ss)+anorthite+melilite+spinel}$  is encountered at subsolidus temperatures. This assemblage is relevant to the  $\text{Ca-Al}$ -rich inclusions of the carbonaceous chondrite as well as that of crystalline phases at invariant point as will be mentioned later.

### 3. Ti-pyroxene Solid Solutions

The diopside(ss) in the present system contains  $\text{CaTiAl}_2\text{O}_6$ , at least up to 15 wt.% ( $\text{TiO}_2$  5 wt.% and  $\text{Al}_2\text{O}_3$  7 wt.%). The chemical analyses of the  $\text{Ti-pyroxenes(ss)}$  by EPMA are given in Table 1. It is remarkable that the  $\text{Ti-pyroxene(ss)}$  contains 8 wt.%  $\text{TiO}_2$  and 14 wt.%  $\text{Al}_2\text{O}_3$ . YANG (1973) also synthesized  $\text{TiO}_2$ -rich pyroxenes in the same system, which contain 8–13 wt.%  $\text{TiO}_2$ . Recently ONUMA *et al.* (1978) confirmed that similar  $\text{Ti-pyroxenes(ss)}$ , containing 13 wt.%  $\text{TiO}_2$ , crystallize in  $\text{CaTiAl}_2\text{O}_6$ -rich part of the system  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaTiAl}_2\text{O}_6$  and coexist with anorthite, melilite, spinel, and perovskite at subsolidus temperatures.

Clinopyroxene containing as much  $\text{TiO}_2$  without  $\text{Fe}_2\text{O}_3$  has never been found in the terrestrial rocks, but has been found in  $\text{Ca-Al}$ -rich inclusions in the carbonaceous chondrite (MASON, 1974; GROSSMAN, 1975).

### 4. Petrography and Chemistry of Type B Inclusions

The mineral assemblage of coarse-grained  $\text{Ca-Al}$ -rich inclusions in the Allende meteorite (C-3 type meteorite) is similar to that of the phase assemblages in subsolidus region confirmed in the system  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$ :  $\text{Ti-pyroxene(ss)+anorthite+melilite+spinel}$  and  $\text{Ti-pyroxene(ss)+anorthite+melilite+spinel+perovskite}$ . GROSSMAN (1975) classified the coarse-grained inclusions into two types: Type A consisting mainly of spinel, melilite, and perovskite and Type B consisting mainly of  $\text{Ti-pyroxene}$ , spinel, anorthite, melilite, and rarely perovskite. The chemical compositions of the  $\text{Ti-pyroxenes}$  from the Type B inclusions are given in Table 1 in comparison with the  $\text{Ti-pyroxenes}$  in the system  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$ . The chemical compositions of both the  $\text{Ti-}$

Table 1. Compositions of Ti-pyroxenes of Type B inclusions (GROSSMAN, 1975) and of the system  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$ .

	DCT System				Type B inclusion					
	$\text{D}_{07}\text{C}_{10}\text{T}_{23}$	$\text{D}_{09}\text{C}_8\text{T}_{23}$	$\text{D}_{05}\text{C}_{12}\text{T}_{20}$	$\text{D}_{60}\text{C}_{27}\text{T}_{13}$	TS8F3-3C	TS23F1-5B	TS12F3-20	TS4F1-4C	TS4F1-1C	TS12F3-21
SiO <sub>2</sub>	40.31	39.39	39.50	40.24	41.41	41.97	39.60	39.31	37.08	37.17
TiO <sub>2</sub>	7.58	8.71	7.48	4.71	4.28	5.01	6.08	7.02	8.07	9.35
Al <sub>2</sub> O <sub>3</sub>	14.67	14.44	14.24	16.21	18.38	18.21	18.16	18.20	19.58	19.46
MgO	12.71	11.44	12.24	10.34	10.35	10.52	9.44	9.90	8.62	8.07
CaO	24.80	26.60	25.90	28.53	25.72	25.93	24.94	25.03	25.38	24.77
FeO	—	—	—	—	0.13	<0.03	<0.03	0.49	<0.03	<0.03
Total	100.07	100.67	99.36	100.03	100.27	101.64	98.22	99.95	98.73	98.82
Si	1.46	1.46	1.47	1.51	1.51	1.51	1.47	1.44	1.38	1.38
Al	0.54	0.54	0.53	0.49	0.49	0.49	0.53	0.56	0.62	0.62
Total	2.00	2.00	2.00	2.00	2.00	2.00	2.00	2.00	2.00	2.00
Al	0.11	0.09	0.09	0.20	0.30	0.28	0.27	0.23	0.24	0.23
Ti	0.21	0.24	0.21	0.13	0.12	0.14	0.17	0.19	0.23	0.26
Mg	0.71	0.63	0.68	0.61	0.56	0.56	0.52	0.54	0.48	0.45
Fe	—	—	—	—	0.00	0.00	0.00	0.02	0.00	0.00
Ca	0.98	1.04	1.03	1.08	1.00	1.00	0.99	0.99	1.01	0.99
Total	2.01	2.00	2.01	2.02	1.98	1.98	1.95	1.97	1.96	1.93

D =  $\text{CaMgSi}_2\text{O}_6$ , C =  $\text{CaAl}_2\text{SiO}_6$ , T =  $\text{CaTiAl}_2\text{O}_6$

pyroxenes are very similar to each other. Spinel is always included in Ti-pyroxene, anorthite, and melilite. MARVIN *et al.* (1970) reported Ca-Al-rich and Si-poor inclusions consisting of spinel and glass, and it seems that these inclusions belong to Type B. BLANDER and FUCHS (1975) also described Ca-Al-rich inclusions. In their 6/1 inclusion spinel and melilite are surrounded by clinopyroxene and eutectic intergrowth is observed between melilite and clinopyroxene or anorthite and clinopyroxene.

The estimated bulk chemical compositions of Type B inclusions in the Allende meteorite (GROSSMAN, 1975) are given in Table 2. When these compositions are calculated in terms of  $\text{CaMgSi}_2\text{O}_6$ ,  $\text{CaAl}_2\text{SiO}_6$ ,  $\text{CaTiAl}_2\text{O}_6$ , and  $\text{MgAl}_2\text{O}_4$  (spinel), the sums of the values make almost 100%, indicating that the Type B inclusions belong to the composition tetrahedron  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$ - $\text{MgAl}_2\text{O}_4$  in the quinary system  $\text{CaO}$ - $\text{MgO}$ - $\text{Al}_2\text{O}_3$ - $\text{TiO}_2$ - $\text{SiO}_2$ . Fig. 4 shows plots of the Type B inclusions in the tetrahedron. It is noticed that although the composition of normative spinel ranges from 16% to 32%, the projections to the basal system  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$  from the  $\text{MgAl}_2\text{O}_4$  apex fall in a narrow area as indicated in Fig. 5, reflecting that the Type B inclusions have similar values in terms of the  $\text{CaMgSi}_2\text{O}_6$  :  $\text{CaAl}_2\text{SiO}_6$  :  $\text{CaTiAl}_2\text{O}_6$  ratio.

Table 2. Calculated compositions of Type B inclusions (GROSSMAN, 1975) and norms.

	Chemical composition			
	iii	iv	v	vi
CaO	17.69	18.09	21.45	20.75
$\text{Al}_2\text{O}_3$	40.27	37.30	32.81	31.72
$\text{TiO}_2$	2.99	3.68	3.73	4.39
MgO	15.37	14.55	12.03	12.59
$\text{SiO}_2$	23.68	26.36	30.01	30.55
Total	100.00	99.98	100.03	100.00
Norm				
Di	31.0(44.8)	31.8(44.8)	37.7(44.9)	37.5(46.0)
Tp	8.8(12.7)	10.9(15.3)	11.2(13.3)	13.1(16.0)
CaTs	29.4(42.5)	28.4(39.9)	35.1(41.8)	31.0(38.0)
Sp	31.7	27.0	16.2	16.2
Total	100.9	98.1	100.2	97.8

The figures in parentheses show the projected composition in the system  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$ .

Di =  $\text{CaMgSi}_2\text{O}_6$ , Tp =  $\text{CaTiAl}_2\text{O}_6$ , CaTs =  $\text{CaAl}_2\text{SiO}_6$ , Sp =  $\text{MgAl}_2\text{O}_4$

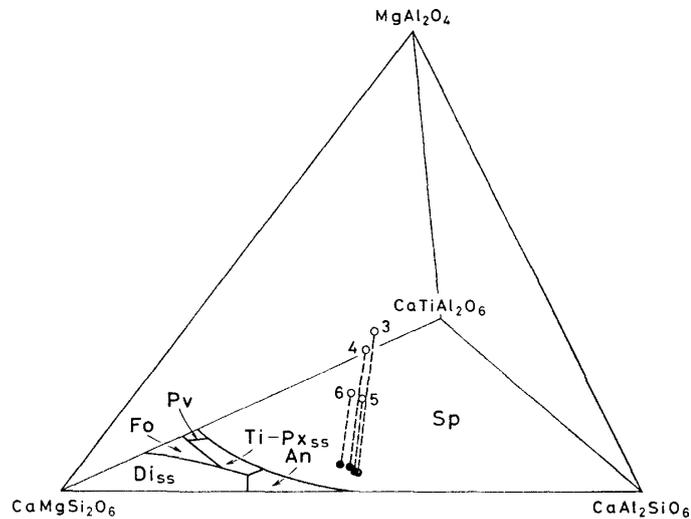


Fig. 4. Plots of Type B inclusions in the Allende meteorite in the  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$ - $\text{MgAl}_2\text{O}_4$  tetrahedron.

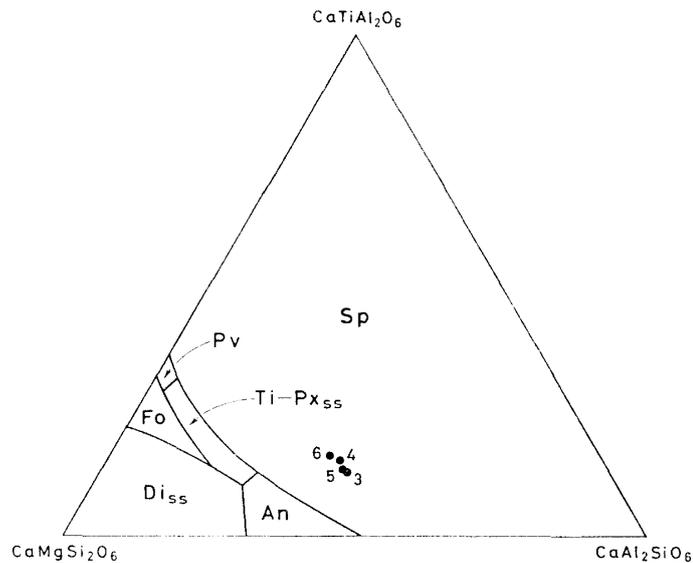


Fig. 5. Projection of Type B inclusions in the Allende meteorite on the  $\text{CaMgSi}_2\text{O}_6$ - $\text{CaAl}_2\text{SiO}_6$ - $\text{CaTiAl}_2\text{O}_6$  basal system from Sp (spinel) apex.

### 5. Crystallization of the Compositions of Type B Inclusions

The petrography of the Type B inclusions indicates that the inclusions have passed a liquid state, and that spinel is the first phase to appear or at least there has been a state at which only spinel coexists with liquid.

Judging from the data of the bounded systems, the compositions of the Type B inclusions are plotted in the spinel primary field of the system  $\text{CaMgSi}_2\text{O}_6$ – $\text{CaAl}_2\text{SiO}_6$ – $\text{CaTiAl}_2\text{O}_6$ – $\text{MgAl}_2\text{O}_4$ . Therefore, when the liquids of these compositions are cooled, the liquids reach spinel liquidus above about  $1500^\circ\text{C}$ . With the falling temperature spinel continues to precipitate and the liquids move towards the basal system  $\text{CaMgSi}_2\text{O}_6$ – $\text{CaAl}_2\text{SiO}_6$ – $\text{CaTiAl}_2\text{O}_6$ . It is noticed that the residual liquids coexisting with spinel in equilibrium have similar composition in terms of the  $\text{CaMgSi}_2\text{O}_6$  :  $\text{CaAl}_2\text{SiO}_6$  :  $\text{CaTiAl}_2\text{O}_6$ , although the starting compositions are different from each other. Consequently, the residual liquids of the Type B inclusions reach the basal system at almost the same temperature and the crystallization trends in this system are the same. At  $1300^\circ\text{C}$  the second phase anorthite begins to precipitate and within a temperature range of  $70^\circ\text{C}$  crystallization of other phases begins and solidus is attained. Thus, the crystallization of the Type B inclusion composition in the system  $\text{CaMgSi}_2\text{O}_6$ – $\text{CaAl}_2\text{SiO}_6$ – $\text{CaTiAl}_2\text{O}_6$  can explain the petrography of the inclusions. According to GROSSMAN (1975) the crystallization sequence of the Type B inclusion is anorthite→Ti-pyroxene→melilite. KURAT *et al.* (1975) considered that melilite and Ti-pyroxene crystallize at almost the same time and then followed by anorthite. Judging from the texture and mineral paragenesis, however, the minerals in question would be crystallized within a small temperature range as in the case of the compositions of Type B inclusions in the present system and the small difference in compositions in a multi-component system probably gives the different order of crystallization, but this is not so serious a problem to the mechanism of the formation of the inclusions. The significance is that spinel is a first phase in crystallization trend and has a long crystallization range, and coexists with the liquids which have the similar composition in terms of  $\text{CaMgSi}_2\text{O}_6$ ,  $\text{CaAl}_2\text{SiO}_6$ , and  $\text{CaTiAl}_2\text{O}_6$  components, resulting in the same phase assemblage of final products. Finally they are followed by anorthite, melilite, Ti-pyroxene with or without perovskite, depending on the starting composition. If the crystallization ceases on the univariant line Ti-pyroxene(ss) + anorthite + melilite + spinel + liquid before the invariant point is reached, perovskite does not join the final assemblage as mentioned before.

BLANDER and FUCHS (1975) suggest that at first the Type B inclusions were formed as liquid condensed from the nebula. On the other hand, some workers (GROSSMAN, 1975; GROSSMAN and CLARK, 1973; CLAYTON *et al.*, 1977) considered that aggregates consisting of spinel, anorthite, melilite, perovskite, and clinopyroxene were formed by condensation from vapor and then melted to liquid. The two hypotheses indicate, either way, that the inclusions once passed a liquid state. The present investigation indicates that once a liquid, or at least an assemblage of spinel+liquid, is formed, then the formation of the Type B inclusions is explained by the crystallization of the system  $\text{CaMgSi}_2\text{O}_6$ – $\text{CaAl}_2\text{SiO}_6$ – $\text{CaTiAl}_2\text{O}_6$ .

CLAYTON *et al.* (1977) proposed the following mechanism: All the solid phases

were condensed from vapor, and then reacted with vapor to form liquid with cooling. Finally the assemblage spinel+liquid is obtained at about 1520°K (1350°C), and from this liquid clinopyroxene, anorthite and melilite crystallize below 1500°K (1230°C) at  $10^{-2}$  atm. In the present investigation, the temperature (1230°C) of an invariant point at which Ti-pyroxene(ss), anorthite, melilite, spinel, and perovskite crystallize agrees with the temperature estimated by CLAYTON *et al.* The assemblage spinel+liquid is obtained above 1300°C from the aggregate of Ti-pyroxene+anorthite+melilite+spinel+perovskite. This also supports the temperature of CLAYTON *et al.*

## 6. Conclusion

The Type B inclusions in carbonaceous chondrite are plotted in the system  $\text{CaMgSi}_2\text{O}_6\text{-CaAl}_2\text{SiO}_6\text{-CaTiAl}_2\text{O}_6\text{-MgAl}_2\text{O}_4$ . The compositions of the liquids in the spinel+liquid assemblage are similar in terms of  $\text{CaMgSi}_2\text{O}_6$ ,  $\text{CaAl}_2\text{SiO}_6$ , and  $\text{CaTiAl}_2\text{O}_6$  components. Therefore, once the liquid or the assemblage spinel+liquid is formed, as suggested by petrography, then the formation of the inclusion is explained by the crystallization in the system  $\text{CaMgSi}_2\text{O}_6\text{-CaAl}_2\text{SiO}_6\text{-CaTiAl}_2\text{O}_6$ . Based on the phase diagram of this system, the following thermal history for the formation of the Type B inclusion is obtained: Spinel+liquid assemblage is formed at least above 1300°C, and then Ti-pyroxene, anorthite, melilite, spinel and sometime perovskite crystallize from the liquid in the temperatures range from 1300°C to 1230°C.

A few carbonaceous chondrites were collected from Antarctica. We expect that Ca-Al-rich inclusions will be found in these chondrites and detailed petrological and mineralogical studies of the inclusions will offer the more information to elucidate the formation mechanism of carbonaceous chondrite.

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