COMPOSITION AND STRUCTURAL SUBSTITUTION OF METEORITIC PLAGIOCLASES (I)

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Abstract: Significant amounts of iron, magnesium and Na (and/or Ca) vacancy are found in meteoritic plagioclases of the Holbrook (L6), Willard (L6), Plainview (H5), ALH-77307,85 (C3) and Allende chondrites and the Juvinas eucrite. Seven components are used in this study, six of which were used already for lunar plagioclases; that is, $KAlSi_3O_8$, $NaAlSi_3O_8$, $CaAl_2Si_2O_8$, $(Fe,Mg)Al_2Si_2O_8$, $Ca(Fe,Mg)Al_3O_8$, $[\ ]Si_4O_8$ and $M(MT)O_8$, where [ ] means Na and/or Ca vacancy, M=K,Na,Ca, and T=Al,Si.

The amounts of components $Ca(Fe,Mg)Si_3O_8$ and $(Fe,Mg)Al_2Si_2O_8$ significantly decrease with ordinary An-content. The highest contents of components $Ca(Fe,Mg)Si_3O_8$ and $(Fe,Mg)Al_2Si_2O_8$ in this study are obtained as 29(mol %) in the ALH-77307,85(C3) and 11(mol %) in the Allende(CV3) chondrites, respectively. The largest values of components $[\ ]Si_4O_8$ and $M(MT)O_8$ are found in this study as 8(mol %) in the Juvinas eucrite and 12(mol %) in the Allende(CV3) chondrite, respectively. Lower Fe/(Fe+Mg) ratio is characteristic of chondrule in meteorite.

The Plainview chondrite showing unique behavior of a considerable variation of the components may indicate a mixture of various parent materials. Small amounts of excess M cations (*i.e.* $M(MT)O_8$) are observed mainly in the exsolved regions of plagioclase crystal in meteorite (esp. the Juvinas eucrite and the Holbrook chondrite). A plot of each end-member abundance *vs.* $CaAl_2Si_2O_8$ may be considered a useful indicator of the formation process of the meteorite.

1. Introduction

A plot of An content *vs.* Fe/(Fe+Mg) which draw a distinction among maskelynite, crystalline and intermediate state in meteoritic plagioclases, was recognized as a potentially useful indicator of the formation process of meteorite (*e.g.* MIÚRA, 1984). The detailed mechanisms of Fe and Mg substitution in meteoritic plagioclases are debatable because of the structural uncertainties of small-sized plagioclase crystals in chondrite. Structurally co-ordinated Fe and/or Mg may change the optical properties of plagioclase crystals (*cf.* BRYAN, 1974). Chemical studies of meteoritic plagioclase with the strict compositional restriction indicate that nearly all deviation from Or-Ab-An chemistry can be explained, same as in the lunar plagioclase, by the components $Ca(Fe, Mg)Si_3O_8$, $(Fe, Mg)Al_2Si_2O_8$ and $[\ ]\ Si_4O_8$, where [ ] signifies M(Na and/or Ca)-site vacancy (*cf.* LONGHI *et al.*, 1976). Calculation method of the above end-member abundances in all lunar plagioclase analyses by BEATY and ALBEE (1980) is based on the assumption that plagioclase is a homogeneous crystal. MIÚRA and TOMISAKA (1984) pointed out that the exsolved plagioclase crystal has excess M atoms, desig-

nated as charge-balanced component $M(MT)O_8$ in this study. If there is no consideration is given to the component $M(MT)O_8$ in the exsolved region, the amount of Ca (Fe, Mg) $AlSi_3O_8$ will increase significantly, and that of (Fe, Mg)$Al_2Si_2O_8$ will decrease. From a large number of meteoritic plagioclase analyses, maskelynite, metamict or microcrystalline phases show the anomalous excess of [ ] Si_4O_8, and increase or decrease of the total cations (*i.e.* more than one percent deviation from 5.000 per 8 oxygens). In this study, the new component $M(MT)O_8$ and analytical data with total cations within one percent deviation are used in meteoritic plagioclase analysis. The purpose of this study is to elucidate the end-member abundances of meteoritic plagioclases, and then to apply conclusions drawn from it to consideration of the formation and crystallization processes of the meteorites which were offered by the National Institute of Polar Research of Japan and were stored at Yamaguchi University, and those of the Y-75 chondritic meteorites for which a meteoritic shower was suggested by MIÚRA and MATSUMOTO (1981, 1982).

The samples used in this study include Y-75135,93 chondrite classified as L5–4 (*i.e.* L5 (67%)) by the statistical method of MIÚRA and MATSUMOTO (1982), ALH-769, 75 (L6), Holbrook (L6), Willard (L6), Plainview (H5), Y-74640,81 (H6), Y-75258, 97 (LL6), ALH-77307,85 (C3) and Allende (CV3) chondrites, and Juvinas (CV3) eucrite. Reported compositional data used for this calculation are those of the Y-75 Antarctic meteorites, namely Y-75102,74 (L4 (67%)), Y-75119,91 (L4 (67%)), Y-75124,91 (L5 (67%)), Y-75128,92 (L4 (83%)) and Y-75133,93 (LL4 (67%)) (MIÚRA and MATSUMOTO, 1982).

2. Experimental

2.1. *Calculation of components*

In order to calculate each component of plagioclase, the EPMA analyzed data of plagioclases in ten meteorites (Y-75135,93, Y-75258,97, ALH-769,75, Y-74640,81, ALH-77307,85, Holbrook, Willard, Plainview and Allende chondrites and Juvinas eucrite) were used from those reported by MIÚRA (1984). Thus, the compositional data

Table 1. Chemical uncertainties (1σ) for the end-member abundances of An-poor(An_{14}) and An-rich(An_{77}) meteoritic plagioclases.

End-member (mol %)	An_{14}(mol %)	An_{77}(mol %)
$KAlSi_3O_8$	2.2(1)	0.5(1)
$NaAlSi_3O_8$	57.7(3)	22.4(5)
$CaAl_2Si_2O_8$	9.5(1)	75.1(11)
$(Fe,Mg)Al_2Si_2O_8$	4.2(1)	0.7(6)
$Ca(Fe,Mg)Si_3O_8$	0.0	0.8(6)
[ ]Si_4O_8	26.4(2)	0.0(0)
$M(MT)O_8$	0.0	0.5(5)
An	13.7(2)	77.2(6)
An(Al,Si)*	9.0(2)	75.9(6)
Fe/(Fe+Mg)	0.49(1)	0.94(5)

*See MIÚRA (1984).

used in this study are satisfied with the total numbers of ions on the basis of 8(O) within the range between 4.950 to 5.050 (*i.e.* 1% deviation) and with the conditions for electrostatic neutrality. The total numbers of the EPMA analytical data fulfilling the above condition are one hundred and five analyzed points (see Table 1 of MIÚRA, 1984).

All plagioclase analyses were normalized to 16 positive charges (assuming ferrous ion), and then the subroutine of the computer program with a PC-9801E (379K) computer was taken for the following manner of calculation: First, the tetrahedral site was filled by making $Ca(Fe, Mg)Si_3O_8=4-(Si+Al)-(K+Na+Ca+Ba-1)$. Next, we set $M(MT)O_8=K+Na+Ca+Ba-1$, $KAlSi_3O_8=K$, $NaAlSi_3O_8=Na$, $BaAlSi_3O_8=Ba$, $CaAl_2Si_2O_8=Ca-Ca(Fe, Mg)Si_3O_8$, $(Fe, Mg)Al_2Si_2O_8=Fe+Mg-Ca(Fe, Mg)Si_3O_8$, and finally $[\ ]Si_4O_8=1-(K+Na+Ca+Ba+dFM)$, where $dFM=Fe+Mg-(4-Si-Al)$. Same grains contain very minor amounts of Ti, Mn and Cr but these were ignored for this calculation because they do not affect the end-member abundances. Although the component $M(MT)O_8$ is not supported structurally so far, it was used as one of the end-member of plagioclase in this study.

2.2. *Analytical error*

The calculated uncertainties (1σ) for the average amounts of any of the major cations are $\pm1.5\%$ for An-content, $\pm1.1\%$ for $CaAl_2Si_2O_8$, $\pm2.3\%$ for $(Fe, Mg)Al_2Si_2O_8$, and $\pm0.8\%$ for $[\ ]Si_4O_8$ in An_{14} meteoritic plagioclase, as shown in Table 1. The accuracy of the measurements listed in Table 1 is similar to that for the lunar plagioclases reported by BEATY and ALBEE (1980), and is enough to discuss the end-member abundances in the meteoritic plagioclases. The above calculated uncertainties are shown by the error bar in Figs. 3 to 5.

Table 2. Data used in Fig. 1. Cations per 8 oxygens for thirty-one analyzed data of seven meteorites used in this study.*

Elements Samples	Ca	Al	Al+2x (Fe+Mg)	Elements Samples	Ca	Al	Al+2x (Fe+Mg)
Holbrook-1	0.106	0.997	1.035	Holbrook-2	0.105	0.968	1.034
ALH-769,75-1	0.086	1.026	1.036	ALH-769,75-2	0.084	1.044	1.050
-3	0.083	1.013	1.035				
Plainview-1	0.150	0.996	1.062	Plainview-2	0.124	1.031	1.083
-3	0.110	1.013	1.097				
Y-75258,97-1	0.086	1.039	1.063	ALH-77307,85-1	0.652	1.157	1.661
Allende-1	0.813	1.648	1.772				
Juvinas- 1	0.821	1.747	1.761	Juvinas- 2	0.927	1.856	1.916
- 3	0.933	1.810	1.826	- 4	0.908	1.828	1.908
- 5	0.846	1.794	1.814	- 6	0.845	1.782	1.796
- 7	0.829	1.718	1.742	- 8	0.789	1.677	1.695
- 9	0.791	1.707	1.723	-10	0.689	1.666	1.694
-11	0.914	1.862	1.868	-12	0.844	1.695	1.819
-13	0.739	1.692	1.710	-14	0.707	1.655	1.671
-15	0.893	1.855	1.869	-16	0.879	1.807	1.821
-17	0.890	1.789	1.801	-18	0.856	1.782	1.824
-19	0.847	1.793	1.817	-20	0.898	1.764	1.974

*Thirty-one analyzed data with probable end-member $Ca(Fe,Mg)AlSi_3O_8$ are used from one hundred and six analyzed data by MIÚRA(1984).

2.3. *Compositional relationship*

For the albite end-member, a variety of structural substituions are shown by all of the charge-balanced type R^{+} (R^{3+}) Si_3O_8 or $R^{2+}R^{2+}Si_3O_8$. In meteoritic plagioclase, the important cations are R^{+}=Na; R^{2+}=Ca, Mg Fe^{2+}; R^{3+}=Al. Because two Al ions balance one Ca ion in the ideal anorthite formula unit and only one divalent ion is required to balance Ca in the albite type, the divalent ions, Mg and Fe, should be doubled and added to Al in the ideal relation for a pure albite-anorthite mixture of plagioclase (BRYAN, 1974). For albite-type substitutions of Ca, Mg and Fe, the relation becomes as follows:

$$Ca=(Al+2Mg+2Fe)-1.$$

For meteoritic plagioclases, thirty-one analyzed data (in Table 2) showing a probable substitution between Al and Fe+Mg are used for the regression analysis, as shown in

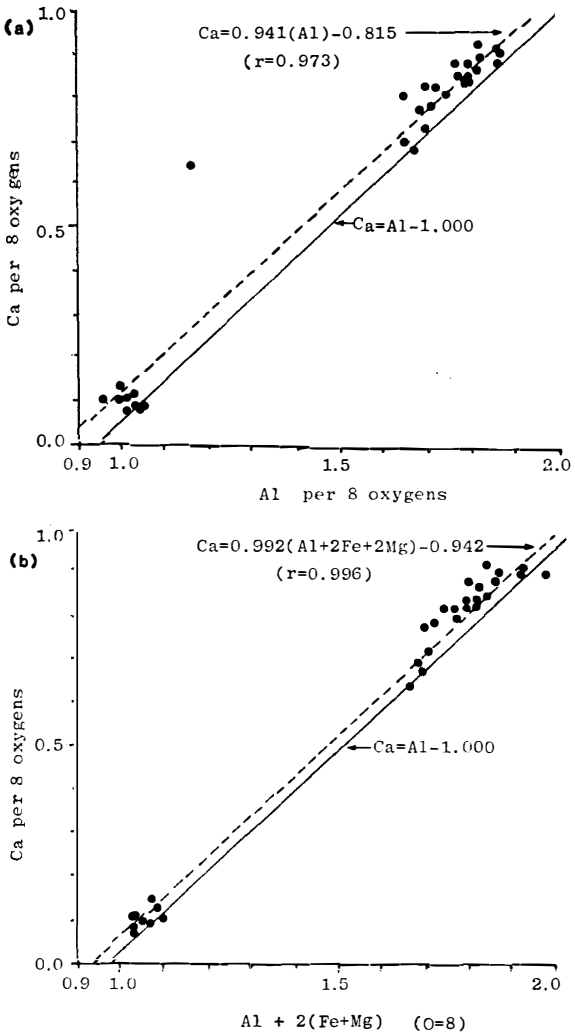

Fig. 1. (a) Regression analysis of Ca=f(Al). Solid line, predicted relation for pure albite-anorthite mixture of plagioclase; dashed line, linear regression line for data set in Table 2. (b) Regression analysis of Ca=f(Al+2Fe+2Mg).

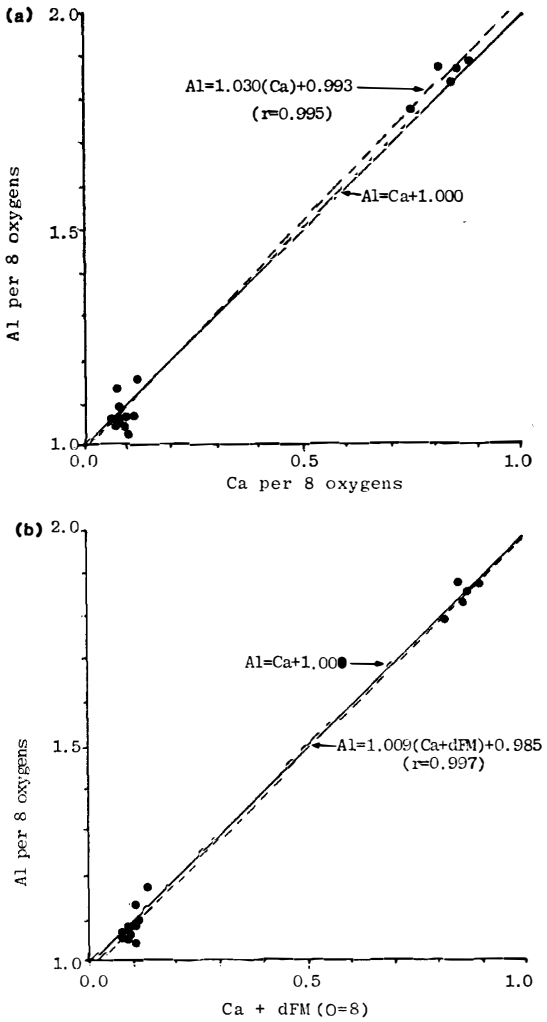

Fig. 2. (a) Regression analysis of Al=f(Ca). Solid line and dashed lines, the same as in Fig. 1 except for Table 4. (b) Al=f(Ca+dFM). dFM=Fe+Mg−(4−Si−Al).

Table 3. Regression equations for the albite-type substitution of Ca, Mg and Fe, by using the data in Table 2.

Regression equation	Standard deviation (σ)
Ideal equation:	
Ca=Al−1.00	1.00
Ca−Al equation:	
Ca=0.941(Al)−0.815	0.973
Ca−(Fe+Mg) equation:	
Ca=0.992(Al+2Fe+2Mg)−0.942	0.996

Fig. 1. A plot of Ca as a function of Al (Fig. 1a) clearly shows an excess of Ca over alumina (*i.e.* standard deviation r=0.973), whereas a plot of Ca as a function of Al+2Mg+2Fe (Fig. 1b) shows that the data more closely approach the expected trend (*i.e.* r=0.996), as shown in Table 3. The calculated regression equation is as follows:

$$Ca=0.992(Al+2Fe+2Mg)-0.942 \qquad (r=0.996).$$

For the anorthite end-member, a variety of structural substitutions are shown by all of the similar charge-balanced type $R^{2+}(R^{3+})_2Si_2O_8$. For anorthite-type substitutions of Al, Mg and Fe, the relation becomes as follows:

Al=(Ca+dFM)+1 . . . (dFM=Fe+Mg−(4−Si−Al)); if Si+Al≥4, then dFM=0.

For meteoritic plagioclases, sixteen analyzed data (in Table 4) showing a probable substitution between Ca and Fe+Mg are used for the regression analysis, as shown in Fig. 2. A plot of Al as a function of Ca (Fig. 2) shows an excess of Al over calcium (*i.e.* r=0.995), and further shows that this excess becomes more pronounced in the more

Table 4. Data used for regression analyses of (Fe,Mg)$Al_2Si_2O_8$ in Fig. 2. Cations per 8 oxygens for sixteen analyzed data of six meteorites used in this study.*

Elements Samples	Ca	Al	Ca+(Fe+Mg)	Ca+dFM**
ALH-769,75	0.080	1.131	0.098	0.098
	0.084	1.077	0.096	0.096
	0.068	1.054	0.083	0.083
	0.093	1.047	0.096	0.093
Plainview	0.095	1.042	0.109	0.101
Y-74640,81	0.095	1.101	0.107	0.107
	0.093	1.087	0.105	0.105
	0.110	1.174	0.126	0.126
Y-75258,97	0.084	1.050	0.089	0.087
	0.079	1.073	0.083	0.079
	0.084	1.073	0.090	0.085
Allende	0.757	1.787	0.823	0.823
Juvinas	0.887	1.881	0.895	0.895
	0.856	1.863	0.861	0.861
	0.813	1.873	0.846	0.846
	0.843	1.848	0.864	0.864

*Sixteen analyzed data with probable end-member (Fe,Mg)$Al_2Si_2O_8$ are used from one hundred and six analyzed data by MIÚRA (1984).

**dFM=(Fe+Mg)−(4.000−Si−Al). If (4.000−Si−Al)≤0, then dFM=0.

Table 5. Regression equations for the anorthite-type substitution of Ca, Mg and Fe, by using the data in Table 4.

Regression equation	Standard deviation (σ)
Ideal equation:	
Al=Ca+1.000	1.000
Al−Ca equation:	
Al=1.030(Ca)+0.993	0.995
Al−(Ca+Fe+Mg) equation:	
Al=1.012(Ca+Fe+Mg)+0.983	0.996
Al−(Ca+dFM)* equation:	
Al=1.009(Ca+dFM)+0.985	0.997

*dFM=(Fe+Mg)−(4.000−Si−Al). If (4.000−Si−Al)≤0, then dFM=0.

calcic plagioclases. This indicates that Fe and Mg substitute for Ca as iron or magnesium anorthite formula unit. Thus a plot of Al as a function of Ca+dFM shows that the data more closely approach the expected trend (*i.e.* $r=0.997$), as shown in Fig. 2 and Table 5. The calculated regression equation is as follows:

$$\mathrm{Al}=1.009\,(\mathrm{Ca}+\mathrm{dFM})+0.985 \quad (r=0.997).$$

The main reason why the regression lines in Figs. 1 and 2 do not completely agree with the trends of $r=1.00$ is that the original data already contain various components of plagioclase.

3. Results and Discussion

Table 6 shows the average values of the end-member (or component) abundances of the plagioclases in nine meteorites. Table 7 shows the compositional data of the highest contents of the components obtained in this study. Figures 3 and 4 show the variation of components Ca(Fe, Mg) Si_3O_8 and (Fe, Mg) $Al_2Si_2O_8$, respectively, as a function of $CaAl_2Si_2O_8$. Table 8 shows the end-member abundances and the Fe/(Fe+Mg) ratios of grains with plagioclase compositions in seven Y-75 chondrites. Table 9 shows average values of the end-member abundances and the Fe/(Fe+Mg) ratios of chondrule and matrix in the Y-74640,81 and Y-75258,97 chondrites. Table 10 shows average values of end-member abundances in the An-content and Fe/(Fe+Mg) diagram of the Plainview chondrite. Table 11 and Fig. 5 show the end-member abundances in the exsolved and homogeneous regions of the Juvinas eucrite (Fig. 6) and the Holbrook chondrite.

From the study of lunar plagioclase (BRYAN, 1974; LONGHI *et al.*, 1976; BEATY and ALBEE, 1980), the component Ca (Fe, Mg) Si_3O_8 is favored by falling temperature, the component (Fe, Mg) $Al_2Si_2O_8$ is experimentally synthesized at a higher pressure of oxygen, and the component [ ] Si_4O_8 is considered an indicator of cooling rate. In meteorite, the existence of the components, Ca(Fe, Mg) Si_3O_8 and [ ] Si_4O_8, is considered as the crystallization at a higher temperature and the quenching process from a higher temperature, respectively. The component of (Fe, Mg) $Al_2Si_2O_8$ indicates the impact event at a relatively lower temperature. Although the further data, as well as the data in this study, are required for the application of the end-member abundances

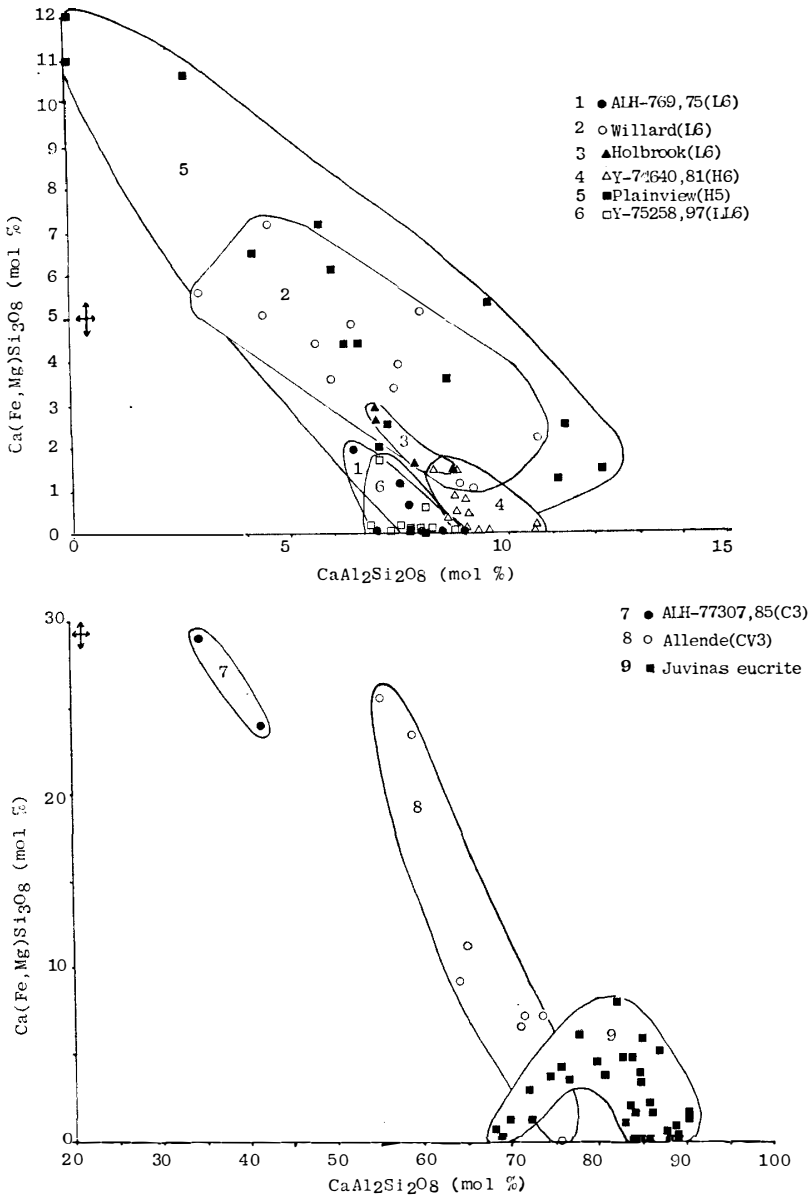

Fig. 3. $Ca(Fe,Mg)Si_3O_8$ as a function of $CaAl_2Si_2O_8$ in nine meteoritic plagioclases. The error bar is calculated chemical uncertainties (1σ). 1. ALH-769,75(L6), 2. Willard(L6), 3. Holbrook(L6), 4. Y-74640,81(H6), 5. Plainview(H5), 6. Y-75258,97(LL6), 7. ALH-77307,85(C3), 8. Allende(CV3), and 9. Juvinas eucrite.

to the formation process of meteorite, a regular relationship of the end-member abundances between the exsolved and homogeneous regions of meteoritic plagioclase is obtained in this study.

3.1. End-member abundances in nine meteorites

Table 6 shows average values of the seven components of plagioclase crystals in the nine meteorites. The difference of the An-content obtained by M atom and the An (Al, Si)-content obtained by T atoms (*cf.* MIÚRA, 1984) is surely explained by the

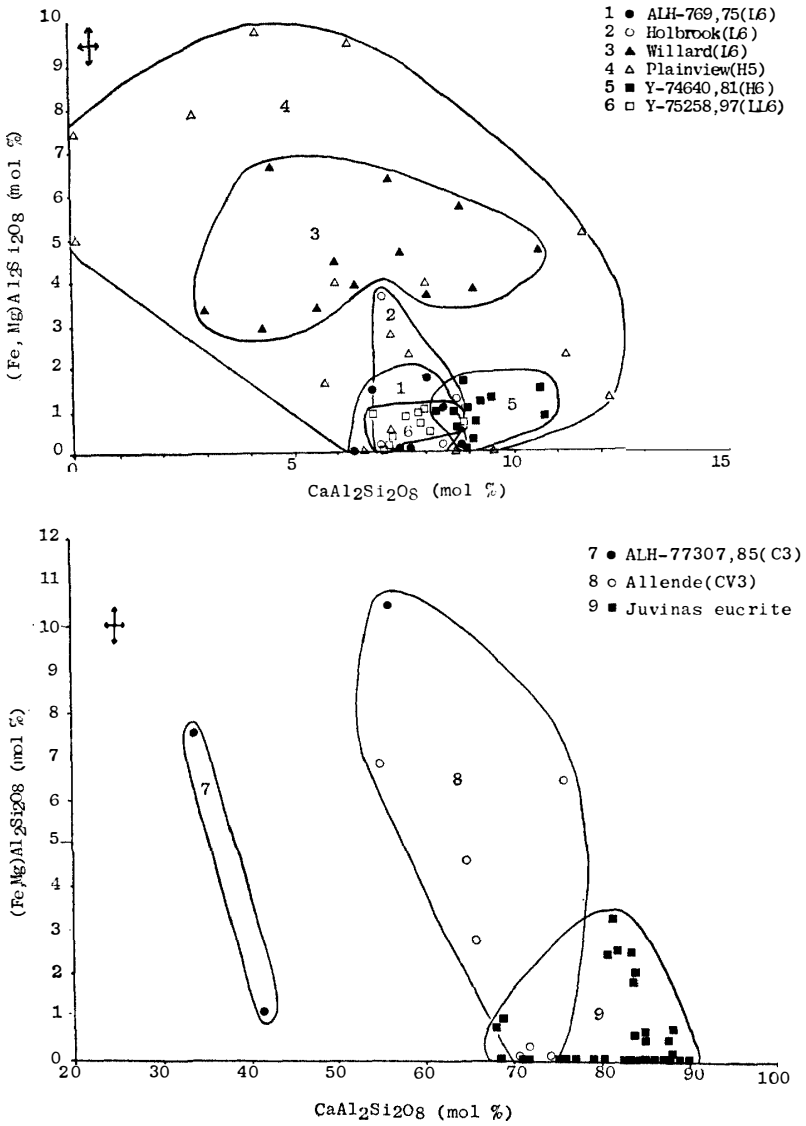

Fig. 4. (Fe,Mg)$Al_2Si_2O_8$ as a function of $CaAl_2Si_2O_8$ for the meteoritic plagioclases. 1. ALH-769,75(L6), 2. Holbrook(L6), 3. Willard(L6), 4. Plainview(H5), 5. Y-74640,81(H6), 6. Y-75258,97(LL6), 7. ALH-77307,85 (C3), 8. Allende(CV3), and 9. Juvinas eucrite.

existence of the four components, (Fe, Mg)$Al_2Si_2O_8$, Ca(Fe, Mg)Si_3O_8, [ ]Si_4O_8 and M(MT)O_8. The larger mean values of An—An(Al, Si) are found in ALH-77307, 85 (C3), Allende eucrite, Holbrook (L6) and Plainview (H5). The ALH-77307,85 (C3) chondrite has the largest average values of An—An(Al, Si)=46.1, Ca(Fe, Mg)Si_3O_8= 26.7 and (Fe, Mg) $Al_2Si_2O_8$=4.4 (mol%) of all the nine meteorites (see Table 6). The amounts of components Ca(Fe, Mg)Si_3O_8 and (Fe, Mg) $Al_2Si_2O_8$ significantly decreased with an ordinary An-content. The Plainview and Allende chondrites showing considerable variation of the components (esp. Ca (Fe, Mg) Si_3O_8 and (Fe, Mg) $Al_2Si_2O_8$)) indicate a mixture of various parent materials (Figs. 3 and 4). Table 7 shows the highest contents of the components obtained in the analyzed meteorites, though

Table 6. Average values of the end-member abundances of the analyzed plagioclases in nine meteorites.

Sample Component (mol %)	Holbrook [4]*	ALH-769,75 [7]	Willard [12]	Plainview [17]	Y-74640,81 [12]
$KAlSi_3O_8$	4.7(7)**	4.0(20)	4.0(16)	5.1(13)	1.0(6)
$NaAlSi_3O_8$	80.5(16)	83.6(24)	79.5(23)	77.4(31)	86.7(14)
$CaAl_2Si_2O_8$	7.8(8)	7.7(8)	6.8(22)	6.7(34)	9.2(7)
$(Fe,Mg)Al_2Si_2O_8$	1.3(14)	0.7(7)	4.4(12)	3.7(31)	1.0(4)
$Ca(Fe,Mg)Si_3O_8$	2.2(6)	0.5(7)	4.0(17)	4.8(36)	0.5(6)
$[\ ]Si_4O_8$	0.0	3.2(23)	0.2(4)	1.0(18)	1.4(13)
$M(MT)O_8$	3.5(9)	0.3(6)	1.1(15)	1.3(18)	0.2(4)
An	10.5(2)	8.6(6)	11.4(14)	12.3(22)	10.0(6)
An(Al,Si)	0.5(5)	5.7(32)	7.6(32)	4.3(24)	9.0(27)
	Y-75258,97 [10]	ALH-77307,85 [2]	Allende [8]	Juvinas [33]	
$KAlSi_3O_8$	3.6(8)	0.5(5)	0.0	0.6(6)	
$NaAlSi_3O_8$	84.6(28)	29.4(11)	14.8(55)	12.6(60)	
$CaAl_2Si_2O_8$	7.7(6)	37.5(36)	66.8(71)	81.8(62)	
$(Fe,Mg)Al_2Si_2O_8$	0.6(3)	4.4(33)	4.1(37)	0.6(9)	
$Ca(Fe,Mg)Si_3O_8$	0.3(5)	26.7(26)	11.6(82)	2.6(22)	
$[\ ]Si_4O_8$	1.0(23)	1.5(13)	1.5(41)	0.4(7)	
$M(MT)O_8$	2.2(22)	0.0	1.2(19)	1.4(19)	
An	8.4(6)	68.3(4)	84.3(51)	87.1(62)	
An(Al,Si)	6.9(19)	22.2(9)	62.0(121)	81.1(63)	

*Numbers of analyzed data used for discussion are shown in square brackets.
**Numbers in parentheses are standard deviation referring to the last decimal place of the analyzed data used for discussion.

the grains of maskelynite, metamict or mixture of more than two phases (*i.e.* microcrystalline phases) show larger values than those of all the crystals with total cations within one percent deviation.

The largest value of $Ca(Fe, Mg)Si_3O_8$ is 29.3 (mol%) found in An-rich (An_{69}) plagioclase of the ALH-77307,85 (C3) chondrite, whereas that in An-poor plagioclase is 10.5 (mol%) in An_{14} plagioclase of the Plainview (H5) chondrite. The most Mg-rich analyzed data in the ALH-77307,85 (C3) chondrite indicate the existence of component $CaMgSi_3O_8$ in An-rich plagioclase. The analysis calculated to formula in An_{69} plagioclase is as follows (Table 7):

$$(Ca_{0.63}Na_{0.28}K_{0.01}Fe_{0.07}Mg_{0.01})(Si_{2.58}Al_{1.13}Mg_{0.29})O_8.$$

The largest value of $(Fe, Mg)Al_2Si_2O_8 = 10.8$ (mol%) is found in An-rich (An_{91}) plagioclase of the Allende (CV3) chondrite, whereas that in An-poor plagioclase is 9.8 (mol%) in An_{12} plagioclase of the Plainview (H5) chondrite. Because of the coexistence of higher amount of Mg, the pure component $FeAl_2Si_2O_8$ could not be found in the meteorites. The formula in An_{91} plagioclase is obtained as follows (Table 7):

$$(Ca_{0.84}Na_{0.08}Mg_{0.10}Fe_{0.10})(Si_{2.27}Al_{1.49}Mg_{0.24})O_8.$$

In contrast to the higher contents of $Ca(Fe, Mg)Si_3O_8$ and $(Fe, Mg)Al_2Si_2O_8$ in

Table 7. Chemical compositions of plagioclases showing representative maximum content of each component obtained in the analyzed meteorites.

Samples	ALH-77307,85 -1 (C3)	Allende -8 (CV3)	Juvinas eucrite	Allende -3 (CV3)
Weight percent oxides:				
SiO_2	56.46	49.31	46.61	50.07
Al_2O_3	20.86	27.58	34.41	23.77
FeO*	1.89	0.25	0.71	0.42
MnO	0.03	0.04	0.00	0.06
MgO	4.36	5.01	0.08	2.60
CaO	12.90	16.96	16.43	21.04
Na_2O	3.19	0.92	0.86	1.20
K_2O	0.08	0.00	0.02	0.00
Total	99.79**	100.07	99.12	99.18***
Cations per 8 oxygens:				
Si	2.583	2.265	2.153	2.354
Al	1.125	1.493	1.873	1.317
Fe	0.072	0.010	0.027	0.017
Mg	0.297	0.343	0.006	0.182
Mn	0.001	0.002	0.000	0.002
Ca	0.632	0.835	0.813	1.060
Na	0.283	0.082	0.077	0.109
K	0.005	0.000	0.001	0.000
Total	4.998	5.030	4.950	5.041
End-member (mol %)				
$KAlSi_3O_8$	0.5	0.0	0.1	0.0
$NaAlSi_3O_8$	28.3	8.0	7.7	7.9
$CaAl_2Si_2O_8$	33.9	57.7	81.3	65.3
$(Fe,Mg)Al_2Si_2O_8$	7.7	**10.8**	3.3	2.8
$Ca(Fe,Mg)Si_3O_8$	**29.3**	23.5	0.0	11.6
$[\ ]Si_4O_8$	0.3	0.0	**7.6**	0.0
$M(MT)O_8$	0.0	0.0	0.0	**12.3**
An(mol %)	68.7	91.1	91.2	90.6
An(Al,Si) (mol %)	21.3	58.9	86.1	43.5
Fe/(Fe+Mg)	0.20	0.02	0.82	0.09

*Total iron oxide as FeO.
**Analysis includes Cr_2O_3=0.02(wt%).
***Analysis includes Cr_2O_3=0.02(wt%).

meteoritic plagioclases, the largest values of $[\ ]Si_4O_8$ and $M(MT)O_8$ in this study are at most 7.6 and 12.3 (mol%), respectively. The largest value of $[\ ]Si_4O_8$ is found in An_{91} plagioclase of Juvinas eucrite, as shown in the following formula (Table 7):

$$(Ca_{0.81}Na_{0.08}Fe_{0.03}Mg_{0.01}Al_{0.02}[\ ]_{0.05})(Si_{2.15}Al_{1.85})O_8.$$

In An-poor plagioclase, the higher content of $[\ ]Si_4O_8$ is 5.5 (mol%) in An_9 plagioclase of ALH-769,75 (L6) chondrite. The detailed discussion on $[\ ]Si_4O_8$ for the formation process as excess silica (BEATY and ALBEE, 1980) is not useful in meteorites, because meteoritic plagioclase having maskelynites or microcrystalline phases show easily more than 10 mol% of component $[\ ]Si_4O_8$ and analyzed data without compositional

restriction (*i.e.* total cations more than one percent). Almost all exsolved regions have the component M (MT) O_8; that is, excess M atoms. The highest content of M (MT) O_8(=12.3 mol%) is found in An_{91} plagioclase of the Allende (CV3) chondrite, as shown in the formula of M $(M_{0.169}T_{3.831})$ O_8 (see Table 7), whereas that in An-poor plagioclase is 4.9 (mol%) in An_{11} plagioclase of the Holbrook (L6) chondrite shown as the formula $M(M_{0.056}T_{3.944})O_8$. The result of the existence of $M(MT)O_8$ indicates that (1) removable M atoms make the pure component $M(MT)O_8$ in the low-temperature field, or (2) more than two (metastable?) phases in the exsolved individuals make the charge-balanced pseudo-component M (MT) O_8. The detailed discussion will be published in the coming paper. In this paper, the component M (MT) O_8 is assumed as one of the end-members in the exsolved plagioclase, mainly because excess M atoms are surely required for charge-balancing (Fig. 5).

3.2. *End-member abundances in Y-75 chondrites*

Although the data of Y-75 chondrites listed in Table 8 are almost maskelynite, monomict or microcrystalline determined both by the petrographic observation and by chemical constraint of more than one percent of the total numbers of cations (see MIÚRA, 1984), the interesting relationships are found among the petrologic type, the An-content and the Fe/(Fe+Mg) ratio. MIÚRA (1984) pointed out that the increased petrologic type, together both with increased Fe substitution and with decreased An-content in the Y-75 chondrites (L4–5), indicates the increase of degree of crystallization from petrologic types 4 to 6 (see Table 8; MIÚRA, 1984). Table 8 shows that there is no

Table 8. End-member abundances and the Fe/(Fe+Mg) ratios of grains with analyzed data of plagioclase(-like) grains in the Y-75 Antarctic meteorites.

Sample No.	Type and group	An (mol %)	An (Al,Si) (mol %)	End-member abundances (mole)♯ K	Na	Ca	FmAl	CaFm	MV	M(MT)	Fe/(Fe+Mg) ratio
Y-75128,92	L4(83%)	29.1	30.5	6.9	59.1	7.2	6.3	9.8	0.0	0.7	0.70
		15.8	8.4	3.4	52.7	10.5	0.8	0.0	32.6	0.0	0.50
Y-75119,91	L4(67%)	11.0	6.4	6.3	65.9	9.0	1.4	0.0	17.4	0.0	0.64
Y-75102,74	L4(67%)	14.2	−1.3	2.1	54.6	9.4	2.1	0.0	31.8	0.0	0.76
Y-75133,93	LL4(67%)	10.1	30.3	6.0	53.8	9.4	2.0	0.0	28.8	0.0	0.81
Y-75124,91	L5(67%)	10.1	−0.4	0.3	61.8	7.0	7.0	0.0	23.9	0.0	0.84
Y-75135,93	L5(67%)										
(maskelynite		10.9	33.2	1.1	75.6	9.4	2.3	0.0	11.6	0.0	0.89
in matrix)	[4]*	(28)**	(18)	(6)	(13)	(4)	(12)		(18)		(8)
(crystal in matrix)		10.4	17.2	1.1	81.5	9.6	2.9	0.0	4.9	0.0	0.83
(crystal in chondrule)		10.4	32.2	1.0	71.6	8.5	1.6	0.0	17.3	0.0	0.69
Y-75258,97	LL6	8.4	6.9	3.6	84.6	7.7	0.6	0.3	1.0	2.2	0.99
	[10]	(6)	(19)	(8)	(28)	(6)	(3)	(5)	(23)	(22)	(1)

*Number of analyzed data used for discussion are shown in square brackets.

**Numbers in parentheses are standard deviation referring to the last decimal place of the analyzed data used for discussion.

♯K=$KAlSi_3O_8$, Na=$NaAlSi_3O_8$, Ca=$CaAl_2Si_2O_8$, FmAl=$(Fe,Mg)Al_2Si_2O_8$, CaFm=$Ca(Fe,Mg)Si_3O_8$, MV=[ ]Si_4O_8, M(MT)=$M(MT)O_8$.

such a remarkable effect of the end-member abundances from petrologic types 4 to 6. The result is quite reasonable if the grains in the Y-75 chondrite (except Y-75258,97) are monomict or microcrystalline. The discussion on the end-member abundances, therefore, should be applied to plagioclase crystals, whereas the value of Fe/(Fe+Mg) would be useful for any structural and compositional states of plagioclase (-like) grains in meteorites. However, the largest values of the end-member abundances are Ca(Fe, Mg)Si_3O_8=9.8 (mol%) and [ ]Si_4O_8=32.6 (mol%) in Y-75128,92 (L4 (83%)), (Fe, Mg) $Al_2Si_2O_8$=7.0 (mol%) in Y-75124,91 (L5(67%)), and M(MT)O_8=2.2 (mol %) in Y-75258,97 (LL6). The value of [ ] Si_4O_8=32.6 (mol%) indicates that the grain in Y-75128, 92 (L4 (83%)) is not normal plagioclase crystal (*i.e.* total cations, 4.733 per 8 oxygen). Although the EPMA analytical data of Fe+Mg substitution, An-content, and end-member abundances may be informative to progressive crystallization from maskelynite to plagioclase crystal probably within the same meteoritic bodies of the Y-75 chondrites, further examples will be required to suggest the results of the probable meteorite shower.

3.3. End-member abundances in chondrule and matrix (Table 9)

MIÚRA (1984) pointed out that plagioclase crystal in the chondrule of the Y-75135, 93 (L5(67%)) chondrite has a lower value of Fe/(Fe+Mg) (=0.69), and that the crystal in the matrix has a relatively higher value (0.83), though the values in the maskelynite are variable (0.77–0.99). Higher contents of [ ]Si_4O_8 and (Fe, Mg) $Al_2Si_2O_8$ in chondrule and matrix, respectively, are found in plagioclase crystal of the Y-75135,93 chondrite (*cf.* Table 8). However, a regular relationship of the end-member abundances between chondrule and matrix is difficult to obtain in plagioclase crystals in the Y-74640, 81 (H6) and Y-75258,97 (LL6) chondrites. This is mainly because the plagioclase crystals between chondrule and matrix in chondrite with petrologic type 6 are difficult to clearly distinguish from each other, and because the different chemical sources between chondrule and matrix might be mixed up in the formation of each chondrite. But the

Table 9. Average values of the end-member abundances and the Fe/(Fe+Mg) ratios of chondrule and matrix in the analyzed meteorites.

Sample	Y-74640,81(H6)		Y-75258,97(LL6)	
Component(mol %)	Chondrule[5]*	Matrix[7]	Chondrule[4]	Matrix[3]
$KAlSi_3O_8$	0.8(6)**	1.2(6)	3.5(6)	3.1(5)
$NaAlSi_3O_8$	87.1(9)	86.3(15)	86.3(11)	85.3(16)
$CaAl_2Si_2O_8$	9.3(7)	9.2(7)	7.6(4)	7.5(5)
$(Fe,Mg)Al_2Si_2O_8$	1.0(3)	0.9(5)	0.6(4)	0.7(5)
$Ca(Fe,Mg)Si_3O_8$	0.2(2)	0.7(6)	0.6(6)	0.0
[ ]Si_4O_8	1.4(12)	1.5(15)	0.2(3)	0.6(9)
M(MT)O_8	0.2(4)	0.2(4)	1.2(8)	3.8(27)
An	9.8(6)	10.2(5)	8.4(4)	8.1(6)
An(Al,Si)	10.1(36)	8.1(13)	5.9(12)	7.7(23)
Fe/(Fe+Mg) ratio	0.90(13)	0.99(2)	0.98(2)	0.99(1)

*Numbers of analyzed data used for discussion are shown in square brackets.

**Numbers in parentheses are standard deviation referring to the last decimal place of the analyzed data used for discussion.

lower ratio of Fe/(Fe+Mg) in chondrule is still held in meteoritic plagioclase crystals in this study.

3.4. *End-member abundances of Plainview chondrite*

MIÚRA (1984) reported that an An-content *vs.* Fe/(Fe+Mg) diagram is useful in expression of intermediate (or transition) state between maskelynite and plagioclase crystal. The Plainview (H5) chondrite consists of three different regions; that is, Fe/(Fe+Mg)=0.42, 0.62 and 0.92 on the average (Table 10), whereas maskelynite has $<$0.38 Fe/(Fe+Mg) in composition of Plainview. Table 10 shows that the end-member abundances of [ ] Si_4O_8 and M (MT) O_8 are higher in the Fe-rich region of Fe/(Fe+Mg)=

Table 10. Average values of the end-member abundances in the An-content and Fe/(Fe+Mg) diagram of the Plainview(H5) chondrite.

Region Component(mol %)	1 [4]*	2 [5]	3 [8]	Average [17]
$KAlSi_3O_8$	4.6(19)**	4.6(5)	5.6(11)	5.1(13)
$NaAlSi_3O_8$	75.4(26)	76.7(35)	78.8(22)	77.4(31)
$CaAl_2Si_2O_8$	5.4(41)	4.3(26)	8.9(18)	6.7(34)
$(Fe,Mg)Al_2Si_2O_8$	6.5(23)	5.4(29)	1.2(13)	3.7(32)
$Ca(Fe,Mg)Si_3O_8$	7.0(35)	7.1(35)	2.4(18)	4.8(37)
[ ]Si_4O_8	1.0(15)	1.4(17)	1.7(24)	1.3(19)
$M(MT)O_8$	0.1(1)	0.5(5)	1.4(21)	1.0(18)
An	13.7(20)	12.3(16)	11.7(23)	12.3(22)
An(Al,Si)	5.7(5)	3.6(31)	4.0(23)	4.3(24)
Fe/(Fe+Mg) ratio	0.42(3)	0.62(6)	0.92(5)	0.71(22)

*Numbers of analyzed data used for discussion are shown in square brackets.
**Numbers in parentheses are standard deviation referring to the last decimal place of the analyzed data used for discussion.

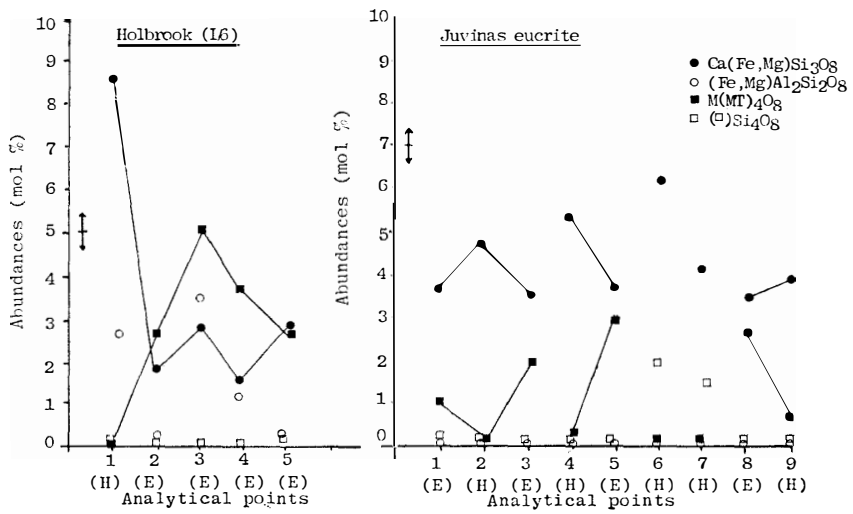

Fig. 5. End-member abundances of plagioclase in homogeneous (H) and exsolved (E) regions in the Holbrook chondrite and the Juvinas eucrite. Solid line shows the change of the regions (H or E) within the same grain.

0.92, whereas those of An, (Fe, Mg) $Al_2Si_2O_8$, and Ca (Fe, Mg) Si_3O_8 are higher in the Mg-rich region of Fe/(Fe+Mg)=0.42. It is also found in this study that the analyzed data of maskelynite and crystals in chondrule are plotted in Mg-rich regions in the Plainview (H5) chondrite (see Fig. 3 of MIÚRA (1984)), though further examples will be required. Figures 3 and 4 show that the considerable variation of the components (esp. Ca(Fe, Mg)Si_3O_8 and (Fe, Mg) $Al_2Si_2O_8$) is characteristic of the Plainview (H5) and may suggest a mixture of various parent materials.

Table 11. Average values of the end-member abundances in the exsolved and homogeneous regions of the Holbrook chondrite and the Juvinas eucrite.

Sample	Holbrook	chondrite	Juvinas	eucrite
Region	exsolved	homogeneous	exsolved	homogeneous
Component(mol %)	[4]*	[1]	[4]	[5]
$KAlSi_3O_8$	4.7(7)**	4.1	0.7(6)	0.5(5)
$NaAlSi_3O_8$	80.5(16)	76.8	16.4(44)	12.6(47)
$CaAl_2Si_2O_8$	7.8(8)	7.8	77.1(47)	81.2(44)
$(Fe,Mg)Al_2Si_2O_8$	1.3(14)	2.6	0.0	0.0
$Ca(Fe,Mg)Si_3O_8$	2.2(6)	8.7	3.7(4)	4.8(8)
$[\ ]Si_4O_8$	0.0	0.0	0.0	0.7(7)
$M(MT)O_8$	3.5(9)	0.0	2.1(7)	0.2(2)
An	10.5(2)	8.8	82.5(49)	86.9(52)
An(Al,Si)	0.5(5)	0.0	76.5(49)	81.6(53)
Fe/(Fe+Mg) ratio	0.80(9)	0.53	0.89(9)	0.72(21)

*Numbers of analyzed data used for discussion are shown in square brackets.
**Numbers in parentheses are standard deviation referring to the last decimal place of the analyzed data used for discussion.

Fig. 6. Microphotograph of the Juvinas eucrite showing exsolved (No. 8 in Fig. 5) and almost homogeneous (No. 9 in Fig. 5) regions. No. 8: $An_{77}Or_{1.7}$, $Ca(Fe,Mg)Si_3O_8=3.3$ (mol %), $M(MT)O_8=2.5$, $CaAl_2Si_2O_8=71.6$, No. 9: $An_{79}Or_{1.6}$, $Ca(Fe,Mg)Si_3O_8=4.0$ (mol %), $M(MT)O_8=0.5$, $CaAl_2Si_2O_8=74.3$.

3.5. End-member abundances of exsolved plagioclase

The exsolved plagioclase crystal has a small amount of $M(MT)O_8$; that is, 2.1 and 3.5 (mol%) on the average in the Juvinas eucrite and the Holbrook chondrite, respectively (Table 11). Figure 5 shows the detailed end-member abundances in homogenous (designated as H) and exsolved (designated as E) grains of the both crystals. Although a definite relationship among all the end-members is slightly difficult to obtain because of various states of crystallization, the amount of $Ca(Fe, Mg)Si_3O_8$ is relatively higher than that of $M(MT)O_8$ in homogeneous crystal of plagioclase, especially in the Holbrook chondrite, as shown in Fig. 5. The higher content of $M(MT)O_8$ in the exsolved regions of compositional zoning or twinned crystal (Fig. 6) is characteristic of the exsolved plagioclase, whereas the small amount of Ca (Fe, Mg) Si_3O_8 still exists in the exsolved regions. However, Table 11 shows that the contents of (Fe, Mg) $Al_2Si_2O_8$, [ ]Si_4O_8 and An are not regular between H and E regions in this study. Anyhow, it is found in Table 11 and Figs. 5 and 6 that excess M atoms and relatively higher iron-content is chemically required for the exsolution. The X-ray study of the exsolved meteoritic plagioclase will be discussed in the coming paper.

4. Conclusions

The results of this study are summaried in the following:

1) Significant amounts of iron, magnesium and Na (and/or Ca) vacancy were observed in meteoritic plagioclases of the Holbrook (L6), Willard (L6), Plainview (H5), ALH-77307,85 (C3) and Allende (CV3) chondrites and the Juvinas eucrite. Seven components were used in this study; that is, $KAlSi_3O_8$, $NaAlSi_3O_8$, $CaAl_2Si_2O_8$, (Fe, Mg)$Al_2Si_2O_8$, $Ca(Fe, Mg)Si_3O_8$, [ ]Si_4O_8 and $M(MT)O_8$, where [ ] means Na and/or Ca vacancy, M=Na, K, Ca, and T=Al, Si. The amounts of $Ca(Fe, Mg)Si_3O_8$ and (Fe, Mg)$Al_2Si_2O_8$ significantly decreased with an ordinary An-content.

2) The end-member abundances of plagioclase were obtained in the nine meteorites (Table 6). The existence of components $Ca(Fe, Mg)Si_3O_8$ and (Fe, Mg)$Al_2Si_2O_8$ easily supported by the regression analyses in Tables 2 and 3, and Tables 4 and 5, respectively.

3) The highest contents of $Ca(Fe, Mg)Si_3O_8$ and (Fe, Mg) $Al_2Si_2O_8$ were 29.3 (mol%) and 10.8 in the ALH-77307,85 (C3) and Allende (CV3) chondrites, respectively. The highest values of [ ]Si_4O_8=7.6 (mol%) and M (MT) O_8=12.3 were found in the Juvinas eucrite and the Allende (CV3) chondrites (Table 7).

4) Although the petrologic types increased from 4 to 5 as the value of Fe/(Fe+Mg) relatively increased in seven Y-75 chondrites from Y-75128,92 (L4 (83%)), Y-75119,91 (L4 (67%)), Y-75102,74 (L4 (67%)), Y-75133,93 (LL4 (67%)), Y-75124,91 (L5(67%)), Y-75135,93(L5 (67%)) to Y-75258,97(LL6), there was no such a remarkable change of the end-member abundances from petrologic types 4 to 6. This shows that the variation of end-member abundances can be found in the plagioclase crystal of chondrite with types 5 to 7.

5) Lower Fe/(Fe+Mg) ratio is characteristic of meteoritic plagioclase in chondrule. The distinct relation of the end-member abundances between chondrule and matrix is difficult to obtain in plagioclase crystals of the Y-74640,81 (H6) and Y-75258,97

(LL6) chondrites (Table 9), though higher contents of [ ]Si_4O_8 and (Fe, Mg) $Al_2Si_2O_8$ in chondrule and matrix, respectively, were found in plagioclase crystals of the Y-75135,93 (L5 (67%)) chondrite (Table 8).

6) Plainview (H5) chondrites with three different values of Fe/(Fe+Mg) (*i.e.* 0.42, 0.62 and 0.92 on the average; MIÚRA, 1984) showed that the end-member abundances of [ ] Si_4O_8 and M (MT) O_8 are higher in the Fe-rich regions of Fe/(Fe+Mg)=0.92, whereas those of An, (Fe, Mg) $Al_2Si_2O_8$ and Ca(Fe, Mg)Si_3O_8 are higher in the Mg-rich region of Fe/(Fe+Mg)=0.42 (Table 10). It is found in Figs. 3 and 4 that a considerable variation of the components is characteristic of Plainview (H5) and might suggest a mixture of various parent materials.

7) The exsolved plagioclase crystal has a small amount of M(MT)O_8; that is, 2.1 and 3.5 (mol%) on the average in the Juvinas eucrite and the Holbrook chondrite, respectively (Table 11). The higher content of M(MT)O_8 is found in the exsolved plagioclases (Fig. 5), whereas the higher amount of Ca(Fe, Mg)Si_3O_8 is observed in the homogeneous regions. Further study of X-ray analytical microscopy is required to support the result that excess M atoms and relatively higher iron-content is characteristic in the exsolution of plagioclase.

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References

BEATY, D. W. and ALBEE, A. L. (1980): Silica solid solution and zoning in natural plagioclase. Am. Mineral., **65**, 63–74.

BRYAN, W. B. (1974): Fe-Mg relationships in sector-zoned submarine basalt plagioclase. Earth Planet. Sci Lett., **24**, 157–165.

LONGHI, J., WALKER, D. and HAYS, J. F. (1976): Fe and Mg in plagioclase. Proc. Lunar Sci. Conf., 7th, 1281–1300.

MIÚRA, Y. (1984): Computer simulation of anomalous composition of Mg-Fe plagioclase in meteorite. Mem. Natl Inst. Polar Res., Spec. Issue, **35**, 226–242.

MIÚRA, Y. and MATSUMOTO, Y. (1981): Classification of several Yamato-75 chondrites (III). Mem. Natl Inst. Polar Res., Spec. Issue, **20**, 53–68.

MIÚRA, Y. and MATSUMOTO, Y. (1982): Classification of several Yamato-75 chondrites (IV). Mem. Natl Inst. Polar Res., Spec. Issue, **25**, 1–16.

MIÚRA, Y. and TOMISAKA, T. (1984): Shachôseki no atarashii tan-seibun ni tsuite (On new end-members of plagioclase). Nihon Kôbutsu Gakkai 1984-nen Kai Kôen Yôshishû (Abstract with Program, Mineral. Soc. Japan, May 27, 1984, Tokyo), 147.

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