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Synthesis and Upper Thermal Stability Limit of 2M-Margarite, $CaAl_2[Al_2Si_2O_{10}/(OH)_2]$

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With 4 Figures and 5 Tables in the text

Abstract

The univariant P_{H_2O}-T curve pertaining to the reaction

$$\text{margarite} \rightleftarrows \text{anorthite} + \text{corundum} + H_2O$$

has been experimentally determined by the method of reaction reversal, starting with a mixture of synthetically prepared pure phases. The equilibrium constant of the reaction may be expressed as $\log K = \log f^P_{T,H_2O} = 8.8978 - 4758/T + 0.0171\,(P-1)/T$, P in bars and T in °K.

On the basis of tabulated thermodynamic data of anorthite, corundum and H_2O (Robie and Waldbaum, 1968; Burnham et al., 1969), standard thermodynamic parameters of margarite are found to be:

$$\begin{aligned} S^0_{298.15} &= 63.1 \pm 2.6 \text{ cal deg-gfw}^{-1}, \\ H^0_{f,298.15} &= -1490.1 \pm 2.2 \text{ kcal gfw}^{-1}, \\ G^0_{f,298.15} &= -1398.3 \pm 3.0 \text{ kcal gfw}^{-1}, \\ V^0_{298.15} &= 3.0984 \pm 0.0012 \text{ cal bar-gfw}^{-1}. \end{aligned}$$

However, these data will require some modification in future, since the tabulated data for anorthite may not be correct (Thompson, 1974). To facilitate recalculation of the thermodynamic data of margarite owing to such change in the suggested thermo-chemical parameters of anorthite and/or corundum, appropriate Gibbs free energy difference functions are given.

Of the three most important dioctahedral mica endmembers muscovite, paragonite and margarite, the last-named has the lowest thermal stability. This accords well with available petrographic data.

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INTRODUCTION

Margarite, $CaAl_2[Al_2Si_2O_{10}/(OH)_2]$, is commonly regarded as a comparatively rare variety of dioctahedral mica. During the last ten years, however, it has been recognized as a fairly common phase in low- to medium-grade metamorphic rocks of various localities (SAGON, 1967, 1970; JONES, 1971; FREY and NIGGLI, 1972; HÖCK, 1972; FREY and ORVILLE, 1974 and others). Although mineral paragenetic data of margarite-bearing rocks are now available in fair amount (e.g. FREY and NIGGLI, 1972; HÖCK, 1972; FREY and ORVILLE, 1974), data bearing on the chemical composition of margarite in these rocks is still very rare. It seems certain, however, that margarite in these rocks form crystalline solutions with paragonite component (cf. JONES, 1971; ACKERMAND and MORTEANI, 1973), although more complex solution involving ephesite may also be fairly widespread (SCHALLER et al., 1967).

Stability relations of paragonite being fairly well established now (CHATTERJEE, 1970, 1972, 1973a), it seems necessary to study the stability and phase relations of margarite. The present paper deals with the upper thermal stability limit of synthetic 2M-margarite endmember, $CaAl_2[Al_2Si_2O_{10}/(OH)_2]$. In the system $CaAl_2Si_2O_8$-Al_2O_3-H_2O, the thermal stability of margarite is marked by the reaction

$$\underset{\text{margarite}}{CaAl_2[Al_2Si_2O_{10}/(OH)_2]} \rightleftarrows \underset{\text{anorthite}}{CaAl_2Si_2O_8} + \underset{\text{corundum}}{Al_2O_3} + H_2O\,.$$

From the experimentally determined P_{H_2O}-T data on this univariant four-phase curve, thermodynamic data for margarite have been extracted. Elucidation of common margarite-bearing assemblages with the help of compatibility relations of margarite, presented orally so far (CHATTERJEE, 1973b), will be the subject matter of a forthcoming paper, now in preparation.

PREVIOUS WORK

Early attempts to synthesize margarite include the works of EUGSTER and YODER (1954) and TU (1956). Both prepared margarite, but failed to establish its thermal stability limit due to extremely slow reaction rates. Since that time, VELDE (1971) has synthesized margarite and studied its upper thermal stability limit. Another pertinent work is that by STORRE and NITSCH (1973), who studied the stability relations of the assemblage quartz + margarite, using a natural margarite crystalline solution form Chester, Mass.

Preliminary results of the present work (CHATTERJEE, 1971) included three P_{H_2O}-T reversal brackets of the reaction margarite $\rightleftarrows$ anorthite + corundum

+ H_2O and unit cell data of synthetic margarite. Since then, better quality margarite could be synthesized; the data presented in this paper will supersede those given earlier.

EXPERIMENTAL METHODS

High Pressure Apparatus

Standard, cold seal, hydrothermal apparatus was used in this study. Approximately 20 mg of starting material along with ~ 5 mg deionize water was welded shut in thin-walled gold capsules, so that free water vapour was always present during the runs. The uncertainty of temperature measurement is believed to be $\pm 5°$ C, that of pressure measurement ± 100 bars. For further detail, the reader is referred to CHATTERJEE (1970, 1972).

In addition to this, solid-media piston-cylinder apparatus (BOYD and ENGLAND, 1960) was also used for some of the synthesis runs.

Starting Materials

Mararite was prepared on its own composition from a mixture of certified reagent-grade precipitated $CaCO_3$, synthetically prepared γ-Al_2O_3, SiO_2 and excess H_2O. The product of the first run usually contained fairly well crystallized margarite with some calcite left over. On recycling, the calcite vanished quantitatively and excellently crystallized margarite was obtained.

Nevertheless, synthesis runs on piston-cylinder apparatus did produce single-phase margarite in only one cycle. This is apparently due to (prefential?) loss of CO_2 through the lid of the capsule and consequent decrease of activity of CO_2 in the charge.

The synthetically prepared starting materials included 2M-margarite, anorthite and corundum, which will be briefly characterized in the next section.

Composition Studied

The upper thermal stability limit of mararite is given by the reaction margarite $\rightleftarrows$ anorthite + corundum + H_2O. To locate the P_{H_2O}-T univariant curve of this reaction, hydrothermal runs were conducted with mixtures of synthetically prepared margarite, synthetic anorthite and synthetic corundum. Two batches of condensed starting materials were synthesized on margarite composition ($CaO\ 2Al_2O_3 : 2SiO_2$ + excess H_2O): 1. a 2M-margarite and 2. a mixture of anorthite and corundum. The final mix contained 20% margarite and 80% anorthite + corundum, i.e. both the reactant and the products of the reaction to be studied. By noting the relative increase in the quantities of the products or the reactant, it was always possible to detect the direction of the reaction in a run.

Phase Identification

All run products were routinely investigated under polarizing microscope, followed by X-ray diffraction scanning. Due to the fine-grained nature of the run products, the X-ray diffraction method proved to be more reliable for detecting direction of the reaction.

The cell dimensions of the synthetic materials were obtained from X-ray diffractometer scans, using KI (a = 7.06516 ± 0.00010 Å) as internal standard for margarite and BaF_2 (a = 6.1971 ± 0.0002 Å) as internal standard for anorthite. Both these standards were calibrated beforehand against gem diamond. The techniques of measurement, indexing and data processing have been described in detail elsewhere (Chatterjee, 1974).

EXPERIMENTAL RESULTS

Phases Synthesized

Margarite: The synthetically prepared margarite crystallized as very fine-grained flaky material. When viewed with scanning electron microscope, they were found to be euhedral to subhedral platelets, up to 2 microns in size.

The X-ray diffraction pattern compares excellently with that of a natural 2M-margarite of near-endmember composition (Fig. 1). Indexed powder X-ray diffraction pattern has been reproduced in Table 1. A close sympathetic variation between observed and calculated intensities (after Borg and Smith,

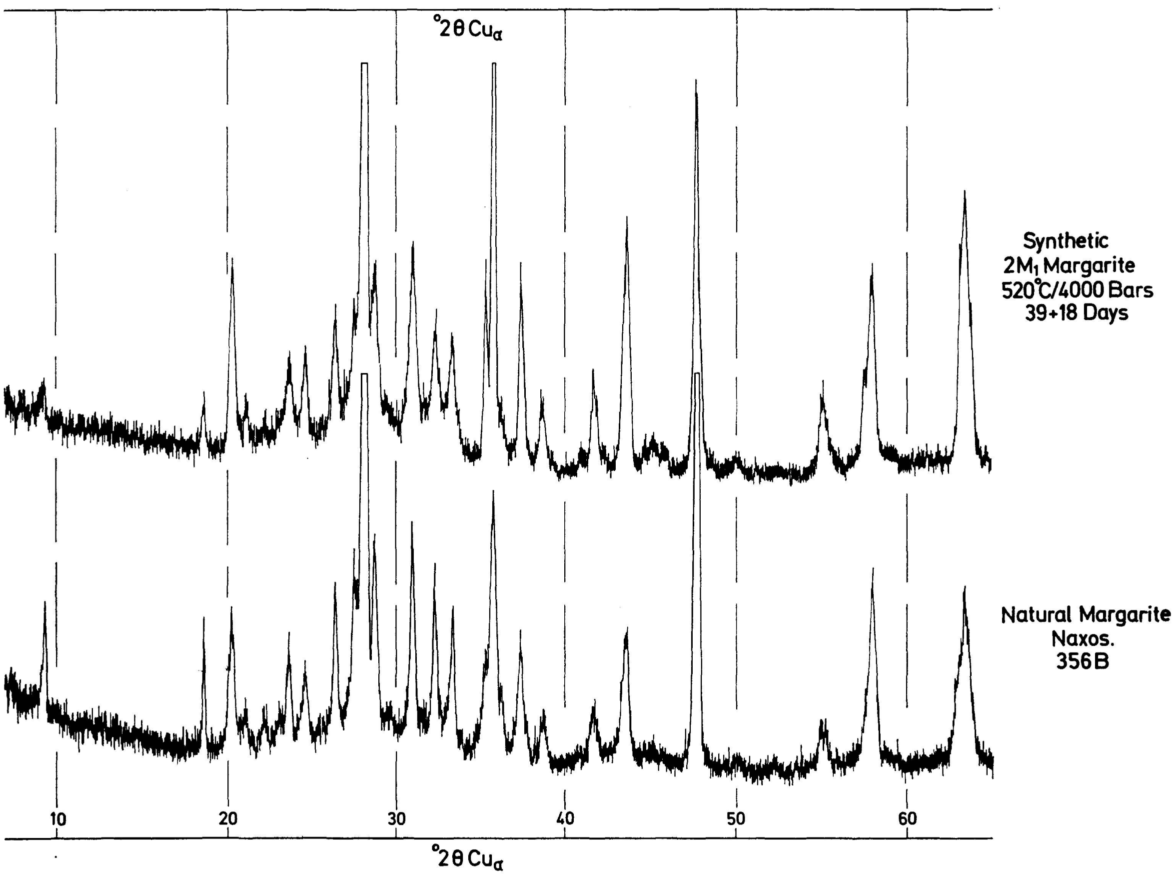

Fig. 1. X-ray diffraction patterns of synthetic and natural 2 M-margarite of near-endmember composition. Stronger non-basal reflections of the synthetic margarite is due to finer grains size (up to 2 microns), as compared to that of natural margarite.

Table 1. *Powder X-ray diffraction data for synthetic 2M-margarite prepared at 520° C / 4 kb P_{H_2O} / 39 + 18 days*

h	k	l	$d_{calc.}$ (Å)	$d_{obs.}$ (Å)	I/I_0 calc. Borg and Smith (1969)	I/I_0 obs.
0	0	2	9.539	9.563	8	4
0	0	4	4.769	4.771	1	4
1	1	0	4.406	4.403	19	13
−1	1	1	4.373	4.377	52	15
1	1	1	4.217	4.217	19	4
0	2	2	4.009	4.009	14	3
−1	1	3	3.771	3.774	30	8
0	2	3	3.629	3.629	26	9
−1	1	4	3.379	3.383	35	12
0	2	4	3.241	3.239	38	12
0	0	6	3.180	3.181	52	100
1	1	4	3.110	3.110	42	15
0	2	5	2.888	2.889	40	17
1	1	5	2.773	2.773	27	10
−1	1	6	2.686	2.687	25	10
−1	3	1	2.542	2.542	24	13
1	3	1	2.510	2.511	100	41
2	0	2	2.399	2.401	25	15
1	3	3	2.327	2.328	10	5
−2	2	1	2.209	2.208	2	1
−1	3	5	2.168	2.168	16	7
2	0	4	2.159	2.160	8	6
−2	0	6	2.085	2.085	21	13
1	3	5	2.074	2.075	35	18
0	4	4	2.005	2.005	9	2
−2	2	5	1.9804	1.9815	6	2
0	0	10	1.9077	1.9072	18	30
−1	3	9	1.6696	1.6687	9	6
2	0	8	1.6613	1.6610	6	5
−2	0	10	1.6011	1.6009	11	10
1	3	9	1.5925	1.5922	23	16
−3	3	1	1.4737	1.4730	41	18
0	6	0	1.4729	1.4730		
−1	3	11	1.4670	1.4675	29	21
2	0	10	1.4600	1.4601	14	11

Space group: C 2/c or Cc.

$a_0 = 5.1061 \pm 0.0010$ Å, $b_0 = 8.8373 \pm 0.0015$ Å, $c_0 = 19.1655 \pm 0.0031$ Å, $\beta = 95° 30.00' \pm 0.79'$, $V_0 = 860.85 \pm 0.18$ Å³.

Refinement on the basis of 34 observed diffraction lines. Standard error unit weight observed: 0.018° 2 θ.

1969) of the margarite diffraction lines is evident. The unit cell dimensions were refined by the method of least squares, using a computer program devised by Evans et al. (1963).

The unit cell dimensions of two synthetic and two natural margarites are given in Table 2. For the sake of comparison, unit cell data of a natural ephesite of near-endmember composition (Schaller et al., 1967) and those of a synthetic 2M-paragonite (Chatterjee, 1974) are also reproduced. While the cell sizes of natural margarite from Naxos (NAX 356 B) are virtually identical

Table 2. *Refined unit cell dimensions of synthetic and natural margarites and other related micas. Space group: C2/c (or Cc)*

Material	a (Å)	b (Å)	c (Å)	β	V (Å^3)	Lines[1])	SEUW[2])	Remarks
Synth. Margarite (Ma 1)	5.1061 (0.0010)[3])	8.8373 (0.0015)	19.1655 (0.0031)	95° 30.00′ (0.79′)	860.85 (0.18)	34	0.018	Prepared at 520° C/ 4 kb P_{H_2O}/39+18 days
Synth. Margarite (Ma 2)	5.1116 (0.0014)	8.8362 (0.0017)	19.1545 (0.0036)	95° 29.73′ (0.87′)	861.18 (0.23)	30	0.020	Synthesized at 620° C/ 11 kb P_{H_2O}/11 days
Nat. Margarite Naxos, Greece (NAX 356B)	5.1110 (0.0017)	8.8410 (0.0024)	19.1493 (0.0045)	95° 31.60′ (1.31′)	861.27 (0.30)	29	0.026	Contain >95 mole % margarite endmember (Chatterjee et al., 1974)
Nat. Margarite Chester, Mass. (Chatt. 1)	5.1219 (0.0024)	8.8603 (0.0033)	19.1607 (0.0067)	95° 20.73′ (2.36′)	865.75 (0.42)	27	0.034	Chemically inhomogeneous mixed crystal of margarite, paragonite and ephesite (Details in Chatterjee et al., 1974)
Synth. Paragonite	5.1304 (0.0010)	8.8927 (0.0015)	19.2698 (0.0028)	94° 13.21′ (0.98′)	876.77 (0.19)	39	0.018	(Chatterjee, 1974)
Nat. Ephesite South Africa	5.120	8.853	19.303	95° 5′	871.5			Near-endmember composition. Single crystal data of M. Ross in Schaller et al. (1967)

[1]) Number of lines used in cell dimension refinements.
[2]) Standard error unit weight observed, in degrees 2 θ.
[3]) Uncertainty quoted is standard error.

Table 3. *Hydrothermal run data bearing on the upper thermal stability limit of margarite*

Run No.	P_{H_2O} (kb)	Temperature (°C)	Run duration (days)	Condensed phases of run products
All runs conducted on CaO : 2 Al_2O_3 : 2 SiO_2 composition with excess water with a mix composed of 20% synth. margarite and 80% anorthite + corundum				
56	1	500	181	Trace Ma left, An+C increased[1])
59	1	490	182	No reaction detected
50	1	480	187	No reaction detected
58	1	470	182	Ma increased, An+C decreased
Reaction interval at 1 kb: 470°–500° C				
35	2	550	89	No Ma, all An+C
42	2	530	115	Ma decreased, An+C increased
51	2	520	137	Ma decreased, An+C increased
49	2	510	145	No reaction detected
57	2	500	142	Ma increased, An+C decreased
43	2	490	103	Ma increased, An+C decreased
Reaction interval at 2 kb: 500°–520° C				
39	4	600	70	No Ma, all An+C
34	4	580	81	No Ma, all An+C
54	4	570	103	No Ma, all An+C
36	4	560	95	Ma increased, An+C less
55	4	540	89	Ma increased, An+C decreased
Reaction interval at 4 kb: 560°–570° C				
31	6	640	52	No Ma, An+C only
32	6	620	58	No Ma, all An+C
52	6	610	89	No Ma, all An+C
38	6	600	62	Ma increased, An+C less
41	6	580	59	Ma only
Reaction interval at 6 kb: 600°–610° C				
45	7	660	59	No Ma, only An+C
53	7	650	79	No Ma, only An+C
47	7	640	80	No Ma, all An+C
48	7	630	80	No reaction detected
44	7	620	61	Ma increased, less An+C
Reaction interval at 7 kb: 620°–640° C				

[1]) Abbreviations used are: Ma: margarite, An: anorthite and C: corundum.

to those of synthetic margarite, the Chester material has a significantly larger cell size, suggesting solid solubility with substantial amounts of paragonite and/or ephesite endmembers. This is corroborated by chemical data (see SCHALLER et al., 1967; and in particular, CHATTERJEE et al., 1974).

Anorthite: The synthetic anorthite, prepared at 700° C/1 kb P_{H_2O}/2 days, showed a powder pattern, which could be indexed on the basis of space group P $\bar{1}$. The cell dimension refinement, based on 41 diffraction lines, gave: $a = 8.1796 \pm 0.0019$ Å, $b = 12.8736 \pm 0.0022$ Å, $c = 14.1720 \pm 0.0025$ Å, $\alpha = 93° \, 7.09' \pm 1.05'$, $\beta = 115° \, 52.48' \pm 1.09'$, $\gamma = 91° \, 14.71' \pm 0.99'$ and $V =$

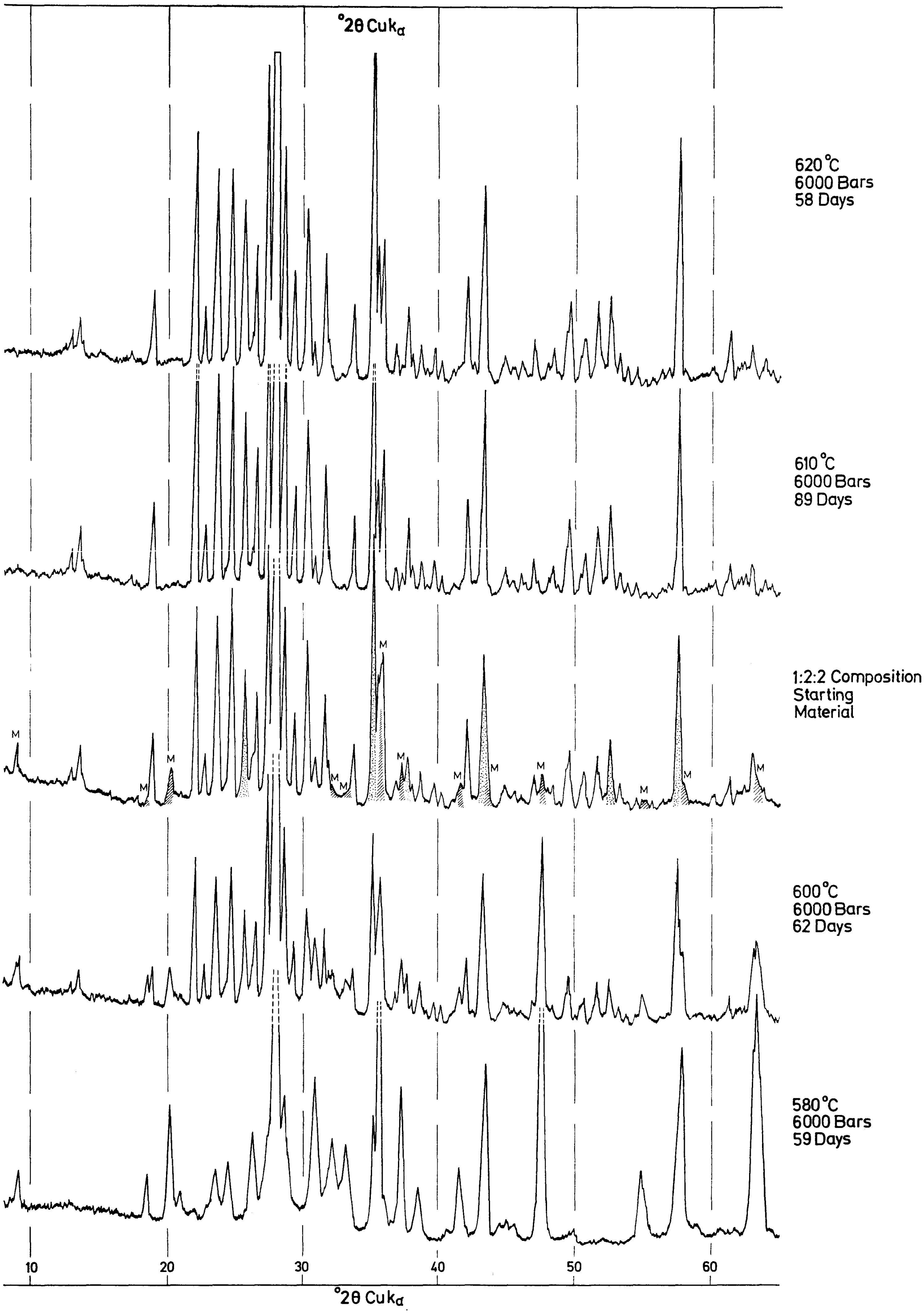

Fig. 2. X-ray diffractograms of condensed $CaO : 2Al_2O_3 : 2SiO_2$ starting material and run products at 6 kb P_{H_2O}. The run at 610° C shows complete decomposition of margarite, while that at 600° C a clear growth of the same phase. Note that the interpretation is based on *all* non-interfering margarite reflections. In the X-ray diffractogram of the starting material those margarite peaks (M) were hatched, which could be used in the interpretation of the direction of the reaction. The corundum peaks are stippled.

1339.0 ± 0.3 Å^3. Standard error unit weight observed was 0.012° 2 θ. These data agree very well with those published for synthetic anorthite by STEWART (1967).

The Reaction: Margarite ⇄ Anorthite + Corundum + H_2O

The run data bearing on the reversal of this reaction on a condensed CaO : $2Al_2O_3$: $2SiO_2$ mix, containing both the reactant and the products, are presented in Table 3. Extremely long runs (up to 187 days at 1 kb P_{H_2O}) were necessary for demonstrating reversal of reaction. In all, five reversal brackets could be obtained between 1 and 7 kb P_{H_2O}, including two ± 5° C brackets. Fig. 2 shows a series of X-ray diffractograms of run products at 6 kb P_{H_2O}, as an example of interpretation of direction of reaction (cf. Table 3).

The run data have been reproduced on a log f^P_{T,H_2O} vs 1/T° K plot (Fig. 3). The brackets shown in thinner lines are those representing log f^0_{T,H_2O}, i.e. after deducting the contribution of the volume change of the solids, ΔV_S, on the Gibbs free energy of the reaction. The thin straight line is a linear regression

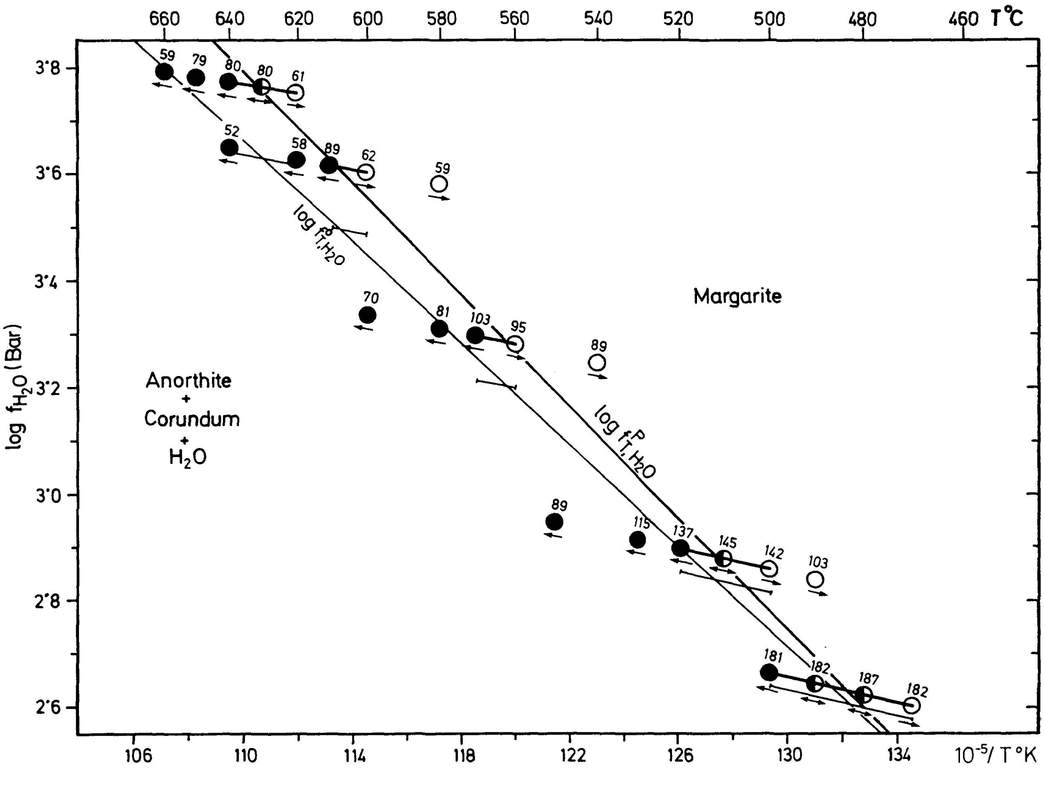

Fig. 3. A log f_{H_2O} vs 1/T °K plot of the run data of the univariant margarite-corundum-anorthite-H_2O equilibrium. Filled circles: growth of anorthite + corundum; half-filled circles: no reaction; and open circles: growth of margarite. The arrows also show the direction of the reaction. The figures at the top of each data point represent run durations in days.

Thicker brackets and linear regression fit are for the log f^P_{T,H_2O} vs 1/T °K data; the thinner ones for the log f^0_{T,H_2O} vs 1/T °K set.

fit to the log f^0_{T, H_2O} vs $1/T° K$ data, which justifies the assumptions (made in the next section) that the heat capacity of the reaction, $\Delta C_{p,r}$, is independent of T at least within the experimentally investigated range.

The P_{H_2O}-T brackets reported here compare favourably with VELDE's (1971) bracket for the same reaction at 2 kb P_{H_2O}, however, the agreement with his 1 kb P_{H_2O} bracket is unsatisfactory.

CALCULATION OF THERMOCHEMICAL PARAMETERS OF MARGARITE

Thermodynamic Data of Margarite

The univariant four-phase equilibrium curve experimentally determined in this study include only one phase, namely margarite, whose thermodynamic data are not yet available from direct calorimetric measurements. Therefore, the experimental P_{H_2O}-T data will be used now to extract thermodynamic data of margarite.

The basic principle underlying these calculations and the method of data reduction has been outlined before (CHATTERJEE, 1970). The format of calculation used tacitly assumes that:

1. All phases involved in the reaction are pure phases of unit activity. This certainly holds true for the solids and possibly also approximately for the vapour present during the runs described above.
2. The volume difference of solids, ΔV^0_s, is independent of pressure P, so that the $\int_1^P \Delta V_s dP$ term may be set equal to $\Delta V^0_s(P-1)$. This assumption is also tenable (as discussed by FISHER and ZEN, 1971, p. 299) in this case, especially because the ΔV_s term contributes very little to the Gibbs free energy within the range of pressure considered here.
3. $\Delta C_{p,r}$ of the reaction is independent of temperature, T. This has already been demonstrated in the last section on the basis of log f^0_{T, H_2O} vs $1/T° K$ plot (Fig. 3).

The standard molar entropy S^0_{298}, the standard molar enthalpy of formation from the elements $H^0_{f,298}$, and the standard molar Gibbs free energy of formation from the elements $G^0_{f,298}$ were calculated for margarite as follows:

$$S^0_{298} = 63.1 \pm 2.6 \text{ cal deg-gfw}^{-1},$$
$$H^0_{f,298} = -1490.1 \pm 2.2 \text{ kcal gfw}^{-1},$$
$$G^0_{f,298} = -1398.3 \pm 3.0 \text{ kcal gfw}^{-1}.$$

The uncertainties given are two standard errors.

Gibbs Free Energy Difference Functions

It should be noted that the thermodynamic data given above will hold true only if the tabulated data for anorthite, corundum and water are correct. Unfortunately, this does not seem to be so at present. One reason for this is that the reference state for Al in the calorimetric data differ for anorthite and for corundum. Thus, the anorthite data are related to gibbsite (BARANY, 1962, p. 13), which in its turn is tied to $AlCl_3 \cdot 6H_2O$ (BARANY and KELLEY, 1961, p. 4). The latter shows a significant deviation from the corundum state. As such, adjustment of the anorthite data with respect to corundum is necessary (THOMPSON, 1973b). Therefore, the margarite data presented above will also have to be adjusted accordingly. To facilitate such data adjustment, Gibbs free energy difference functions, as suggested by D. R. WALDBAUM and extensively used by THOMPSON (1973a, 1973b), are given below. These were computed from basic thermochemical data of solids reproduced in Table 4 and thermodynamic data of water (BURNHAM et al., 1969) as recast by FISHER and ZEN (1971, Table 1).

Table 4. *Basic thermodynamic data of the solid phases, used in computing the Gibbs free energy difference functions X and Y*

Phases	S^0_{298} (cal deg-gfw^{-1})	$S^0_{f,298}$ (cal deg-gfw^{-1})	$H^0_{f,298}$ (cal gfw^{-1})	$G^0_{f,298}$ (cal gfw^{-1})	V^0_{298} (cal bar-gfw^{-1})	Source
Margarite	63.11 (2.61)[2]	−308.105 (2.61)	−1490136[1] (2170)	−1398274[1] (2990)	3.0984[3] (0.0012)	This study
Anorthite	48.45 (0.30)	−180.024 (0.30)	−1009300 (1150)	− 955626 (1160)	2.4089 (0.0012)	ROBIE and WALDBAUM, 1968
Corundum	12.18 (0.03)	−74.854 (0.03)	− 400400 (300)	− 378082 (310)	0.61126 (0.00017)	ROBIE and WALDBAUM, 1968

[1]) In order to retain internal consistency, the data have *not* been rounded off on the basis of uncertainty.
[2]) The uncertainty quoted correspond to *two* standard errors in each case.
[3]) Averaged from the unit cell data of synthetic margarite, given in Table 2.

The thermodynamic formalism underlying the computation of Gibbs free energy difference functions has been outlined by THOMPSON (1973a, 1973b). Two different Gibbs free energy difference functions X and Y were computed (Table 5), which are defined as follows:

$$X \equiv G^0_{f,298,An} - G^0_{f,298,Ma} = -G^0_{f,298,C} + \Delta S^0_{f,s} \Delta T - \Delta V^0_s \Delta P - G^{*P}_{T,H_2O}$$

and $$Y \equiv G^0_{f,298,An} + G^0_{f,298,C} - G^0_{f,298,Ma} = +\Delta S^0_{f,s} \Delta T - \Delta V^0_s \Delta P - G^{*P}_{T,H_2O}.$$

To give an example of the use of these difference functions, we may wish to adjust the $G^0_{f,298}$ of margarite based upon values of $G^0_{f,298}$ of anorthite and corundum, as suggested by THOMPSON (1973b).

Table 5. *Gibbs free energy difference functions X and Y (defined in the text) related to the univariant margarite-anorthite-corundum-H_2O equilibria*

Equilibrium Temperature (°C)	Equilibrium H_2O-Pressure (bars)	G^{*P}_{T,H_2O} (cal gfw^{-1}) (after FISHER and ZEN, 1971, Table 1)	X (cal)	Y (cal)
485	1000	−40042	+442687	+64605
510	2000	−38543	+442596	+64514
565	4000	−35543	+442680	+64598
605	6000	−33129	+442552	+64470
630	7000	−31764	+442595	+64514
		Average	+442622 ±59	+64540 ±59

In order to be internally consistent with THOMPSON's (1973b, Table 5) data set A, $G^0_{f,298}$ of margarite will have to be −1406.246 Kcal gfw^{-1}, while $G^0_{f,298}$ of margarite will be −1402.240 Kcal gfw^{-1} to be consistent with his data set B.

It should be pointed out, however, that this method of data adjustment based upon Gibbs free energy difference functions is meaningful as long as changes only in $G^0_{f,298}$ of a solid phase is indicated. Should a different value for $S^0_{f,298}$ be found to apply than that used in the computation of the difference function, this method will become vulnerable.

CONCLUSIONS

A P_{H_2O}-T plot of the thermal stability limits of the three important dioctahedral mica endmembers muscovite (CHATTERJEE, 1973, unpublished), paragonite (CHATTERJEE, 1970) and margarite (this study) has been reproduced in Fig. 4. It is seen that muscovite has the highest and margarite the lowest thermal stability limit. In a general way, this agrees with the petrographic data available to date.

However, the thermal stability limits of the mica endmembers have only limited application to natural rocks for two reasons:

1. The micas are almost always found in natural rocks in mineral assemblages, in which they coexist with other minerals. Therefore, not the thermal stability limits but the compatibility relations of the micas would apply to natural rocks.
2. The mica endmembers usually form crystalline solutions with each other in nature. As such, compatibility relations of appropriate mica crystalline solutions will be of interest.

In a forthcoming paper, now in preparation, compatibility relations of margarite as well as their possible geologic applications will be discussed in detail.

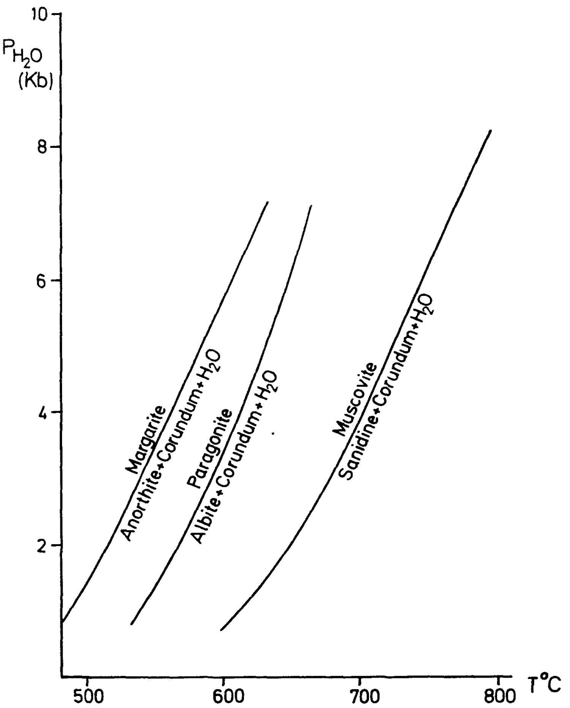

Fig. 4. Comparison of upper thermal stability limits of dioctahedral mica *endmembers* margarite (this study), paragonite (Chatterjee, 1970) and muscovite (Chatterjee, 1973, unpublished).

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