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MUSCOVITE-PLAGIOCLASE EQUILIBRIA IN SILLIMANITE + QUARTZ BEARING METAPELITES, PUZZLE MOUNTAIN AREA, NORTHWEST MAINE

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ABSTRACT. The compositions of coexisting muscovite and plagioclase have been determined by electron probe analysis from 116 lower sillimanite (LSZ) through sillimanite + K-feldspar (SKZ) zone metapelites of the Puzzle Mt. Area, northwest Maine. In these samples, a 1 mole percent decrease in the albite content of plagioclase is accompanied by a decrease in the paragonite content of the coexisting muscovite of 0.3 percent in the LSZ and 0.1 percent in the SKZ.

Assuming P_{H_2O} = Ptotal = 3500 bars, the relation

$$\exp\left[\frac{\Delta\mu^\circ}{RT} + \frac{\Delta\bar{V}_s^\circ\,(P-1)}{RT}\right] = \left(\frac{X_{Pg}^{musc}}{X_{Ab}^{plag}}\right)\left(\frac{1}{f_{H_2O}}\right)\left(\frac{\gamma_{Pg}^{musc}}{\gamma_{Ab}^{plag}}\right)(X_{Al,Mg}^{musc})^2$$

was solved for a temperature of equilibration of each specimen. These temperatures have been used to determine:

1. That a thermal gradient of ~60° to 80°C per km existed during the final Acadian metamorphic event in the Puzzle Mt. Area.

2. That adjacent specimens containing apparently lower and higher grade AKNa-Ca mineral assemblages (that is, sill + musc + plag + qtz and sill + ksp + musc + plag + qtz) resulted from a f_{H_2O} approx 30 percent lower accompanying the equilibration of the K-feldspar-bearing specimens.

Due to uncertainties in the requisite thermochemical parameters (that is, $\Delta\mu^\circ$, Margules parameters, et cetera), temperatures derived from the equation above are, at best, relative values. Nonetheless, the results summarized above indicate that the compositions of coexisting muscovite and plagioclase can be used as a viable indicator of intensive parameter (T and/or f_{H_2O}) *variation* within a single, petrologically well characterized terrain.

NOTATION

A. Phases

musc	muscovite
plag	plagioclase
ab	albite
andy	andalusite
sill	sillimanite
ksp	K-feldspar
qtz	quartz
biot	biotite
st	staurolite
gar	garnet
ilm	ilmenite
po	pyrrhotite

B. Components

Mu	$KAl_3Si_3O_{10}(OH)_2$
Pg	$NaAl_3Si_3O_{10}(OH)_2$
Ab	$NaAlSi_3O_8$
An	$CaAl_2Si_2O_8$
Or	$KAlSi_3O_8$
Qz	SiO_2
Si	Al_2SiO_5

C. Metamorphic zones

LSZ	lower sillimanite zone
USZ	upper sillimanite zone
SKZ	sillimanite + K-feldspar zone

D. Other Notation

T	temperature (°K, exceptions noted)
P, Ptotal	lithostatic pressure (bars)
P_{H_2O}	H_2O pressure (bars)
f_{H_2O}	fugacity of H_2O (bars)

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X_i^α	mole fraction of component i in phase α
γ_i^α	activity coefficient of component i in phase α
W_i	margules parameter of component i
a_i	activity of component i
μ_i^α	chemical potential of component i in phase α (J/mole)
μ_i°	chemical potential of a pure phase (J/mole)
$\Delta\mu^\circ$	molar Gibbs energy of reaction involving pure phases (J/mole)
$\Delta\bar{V}_s^\circ$	molar volume change of solids at 298°K and 1 bar (J/bar)
ν_i	stoichiometric coefficient
M_i	moles (or grams) of component i
J	joule
ln	natural logarithm
R	universal gas constant (8.3147 J/ (°K mole)

INTRODUCTION

This report is part of an ongoing study of the metamorphism of pelitic schists from Northwest Maine initiated by the junior author. Much of the data presented here represents a portion of the compositional data compiled in a study of the metamorphism of the Puzzle Mountain area (fig. 1) by Cheney (ms and in preparation). The purpose of this contribution is to examine muscovite–plagioclase equilibria in sillimanite–quartz-bearing metapelites at metamorphic grades ranging from lower sillimanite (LSZ) through sillimanite + K-feldspar (SKZ) zones. In particular, we will focus on the interrelationships among the paragonite (Pg) content (Na/Na + K ratio) of muscovite, composition of the coexisting plagioclase, and the intensive parameters T and a_{H_2O}.

The usefulness of muscovite composition as a petrogenetic indicator in metamorphic rocks has recently been reviewed by Guidotti and Sassi (1976) and Thompson and Thompson (1976). By considering certain specific mineral assemblages within the subsystem Al_2O_3–K_2O–Na_2O–SiO_2–H_2O (the AKNa subsystem) bulk composition effects are minimized. Then, by a continuous reaction the Pg content of muscovite coexisting with sill + ab + qtz has been found to decrease with increasing metamorphic grade (increasing T or decreasing a_{H_2O}). Consequently, the Pg content of muscovite in such an assemblage can, with caution, be used qualitatively (compare Guidotti and Sassi, 1976) or quantitatively (compare A. B. Thompson, 1974; Chatterjee and Froese, 1975) to evaluate intensive parameter variation within a given metamorphic terrain.

Unfortunately, few natural pelites are adequately characterized by the AKNa subsystem because the plagioclase feldspar in these rocks has a significant An-content. General phase relationships within the AKNa–Ca subsystem, summarized by Evans and Guidotti (1966), indicate that the addition of An component to plagioclase coexisting with sill + qtz + musc results in a decrease in the Pg content of the coexisting muscovite. Therefore, the position of the continuous reaction, and hence muscovite composition, is functionally dependent on the activity of the Ab component in plagioclase in addition to P, T, and a_{H_2O}.

Our specific objectives are to evaluate the following with respect to the AKNa–Ca assemblages sill + musc + plag + qtz and sill + ksp + musc + plag + qtz:

1. The dependence of the Na/Na + K ratio of muscovite on the Ab content of coexisting plagioclase.
2. The change in this compositional dependence as a function of metamorphic grade.
3. The partitioning of Na between plagioclase and muscovite in order to evaluate the variation of intensive parameters within rocks from a single, well characterized metamorphic terrain, specifically the Puzzle Mountain area of Northwest Maine.

PETROLOGIC SETTING

The Puzzle Mountain area of Northwest Maine (fig. 1) is located on the northwest limb of the Merrimack synclinorium in what Moench and

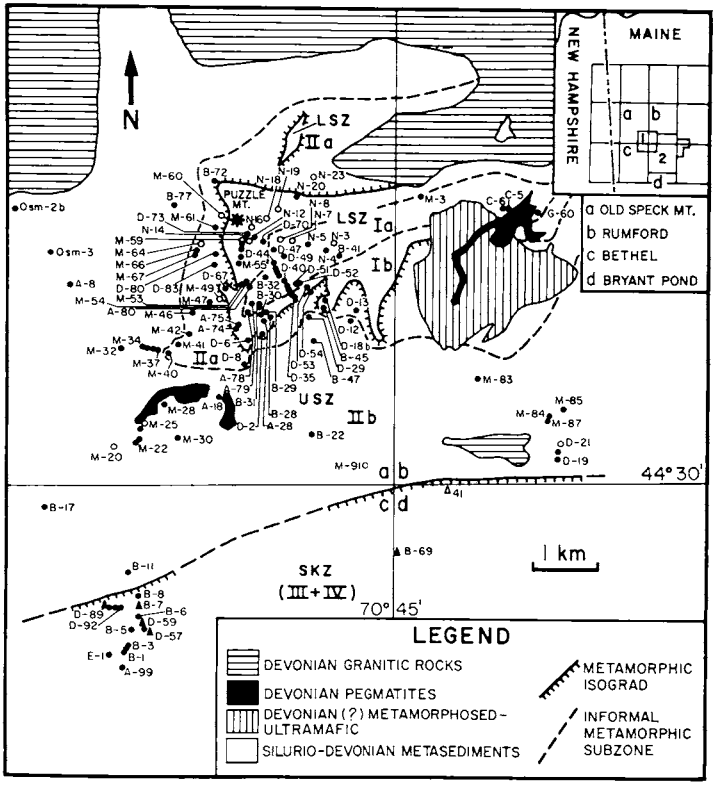

Fig. 1. Geologic map (modified from Moench and Hildreth, 1974, and Milton, ms) of the Puzzle Mountain area showing the distribution of specimens from which pertinent data have been obtained. Specimens containing sill + musc + plag + quartz are represented by closed circles, specimens from which only muscovite has been analyzed are represented by open circles, and specimens containing sill + ksp + musc + plag + qtz are represented by closed triangles (this study) and open triangles (Evans and Guidotti, 1966). Inset shows relation of (1) this study area to that of (2) Evans and Guidotti (1966).

Zartman (1976) call an orogenic transition zone between the singly deformed, low-grade terrain of northern Maine and the multiply deformed high-grade terrain of southern New England. The geologic history of this transition zone has been summarized by Guidotti (1970a, b, and 1974) and Moench and Zartman (1976) and thus need not be repeated in detail here. Suffice it to say that the pelitic schists of the Puzzle Mountain area are multiply deformed, polymetamorphic, Siluro-Devonian strata in which the metamorphic mineral assemblages reflect the last Acadian metamorphic event. This event was a post kinematic thermal overprinting related to the intrusion of 379 m.y. adamellites of the New Hampshire magma series (Moench and Zartman, 1976). The detailed studies of Cheney (ms), Cheney and Guidotti (1973), and Evans and Guidotti (1966) strongly suggest that the observed mineral assemblages, mineral compositions, and observed textural relations are consistent with a prograde sequence which closely approximated equilibrium.

The *apparent* prograde sequence produced by the final thermal event in the Puzzle Mountain area involves, sequentially, the gradual pseudomorphing of staurolite by coarse muscovite (+ biotite), the disappearance of staurolite from the pseudomorphs, disappearance of the pseudomorphs concomitant with the appearance of muscovite megacrysts in many specimens (compare Cheney and Guidotti, 1973), and finally the appearance of K-feldspar in sillimanite-bearing rocks. The two mineralogic discontinuities in this sequence form the basis for mapping two isograds, in that the disappearance of staurolite and the appearance of K-feldspar require reorientations of tie lines on the AFM and AKNa projections, respectively.

The isograd separating lower and upper sillimanite zones is based on the AFM terminal stability of staurolite by the reaction:

$$\text{st} + \text{qtz} + \text{musc} = \text{sill} + \text{biot} + \text{gar} + \text{H}_2\text{O} \qquad (1)$$

Typically, LSZ mineral assemblages contain staurolite and sillimanite, whereas staurolite is absent in the USZ (upper sillimanite zone). However, for the purpose of differentiating higher and lower grade portions of the LSZ and USZ, textural details of the above noted pseudomorphing process allow a general subdivision of each of these mineral facies into informal subzones. Division of the LSZ is based on the extent of development of muscovite (+ biotite) pseudomorphs after staurolite. Staurolite is unpseudomorphed or only partially so in the lower grade subzone. In the higher grade subzone (especially close to the isograd), it occurs mainly within polycrystalline pseudomorphs but still crosses grain boundaries between quartz and muscovite.

The isograd marking the disappearance of staurolite is defined by the last (highest grade) occurrence of staurolite observed to cross grain boundaries, especially between quartz and muscovite (as is implicit in the use of the AKFM projection). Above the isograd very small (0.01-0.03mm) blebs of staurolite persist in a few specimens as inclusions predominantly in quartz and plagioclase but also in muscovite. Subdivision

of the USZ is based on the lower grade subzone having specimens containing "inclusion" staurolite versus higher grade subzone rocks with no staurolite. Because this subdivision is based on the "last" (highest grade) occurrence of "inclusion" staurolite, the lower grade portion of the USZ contains many specimens without any staurolite.

The sillimanite + K-feldspar isograd considered here is the western extension of that studied by Evans and Guidotti (1966). It separates USZ and SKZ rocks and is based upon the initial occurrence of groundmass K-feldspar in sillimanite-bearing rocks. The isogradic reaction is:

$$\text{musc} + \text{plag} + \text{qtz} = \text{sill} + \text{ksp} + \text{H}_2\text{O}. \qquad (2)$$

Although theoretically a discontinuous reaction (in the AKNa system), addition of An (anorthite) component to plagioclase makes eq (2) a trivariant, continuous equilibrium (Evans and Guidotti, 1966). In fact, the "isogradic" assemblage (sill + ksp + musc + plag + qtz) persists above the isograd defined by the initial compatibility of sill + ksp and is intermingled with rocks containing the apparent USZ assemblage sill + musc + plag + qtz. Moreover, this mixture of K-feldspar and non K-feldspar-bearing SKZ assemblages cannot be rationalized simply by consideration of the An component in plagioclase (see Evans and Guidotti, 1966, p. 55 and below). Evans and Guidotti (1966), therefore, proposed that the mixture of assemblages was due (compare Chatterjee and Froese, 1975, p. 991) to a lower a_{H_2O} in the K-feldspar bearing rocks during equilibration, a conclusion substantiated below.

In summary, the assemblage sill + musc + plag + qtz occurs in LSZ through SKZ metapelites, and in the SKZ it is intermingled with the assemblage sill + ksp + musc + plag + qtz. Hence these AKNa–Ca assemblages can be classified as a function of mineral facies and informal subzone in the following manner:

LSZ	(I)	sill + musc + plag + qtz	(Ia) lower grade	(Ib) higher grade
USZ	(II)	sill + musc + plag + qtz	(IIa) lower grade	(IIb) higher grade
SKZ	(III)	sill + musc + plag + qtz		
	(IV)	sill + ksp + musc + qtz + plag		

(Assemblages IIb, III, IV correspond to assemblages II, III, IV respectively of Evans and Guidotti, 1966).

Figure 1 shows the distribution of specimens (and assemblages) from which mineral compositional data have been obtained. It can be seen that most assemblage Ia and assemblage IV specimens are from geographically restricted areas which approximate isothermal portions of the Puzzle Mountain terrain.

ANALYTICAL SETTINGS

The University of Wisconsin ARL-EMX electron microprobe was used to obtain compositional data from 111 muscovites and coexisting

TABLE 1
Chemical analyses of muscovite††

Specimen	SKZ-III														
No.	B8	B5	B11	D19	D21	D20b	M32	P85-R	P85-H	P85-Q	M30	P85-S	B17	M20	P85-U
SiO_2	46.53	44.76*	46.56	46.89	47.01	46.90	46.05	46.46	46.90	47.54	46.14	47.47	46.92	46.40	47.26
Al_2O_3	35.43	35.92	35.17	35.32	35.16	35.25	35.69	36.62	35.13	35.68	35.54	35.24	35.42	36.03	35.51
FeO	1.03	1.07	1.04	0.77	0.42	0.75	1.07	1.40	.69	.49	1.02	.41	1.04	0.89	.37
MgO	0.61	0.63	0.72	0.91	1.18	0.95	0.61	.89	1.03	.91	0.58	1.10	0.56	0.54	1.16
TiO_2	0.76	0.71	0.93	0.99	1.06	0.81	0.62	.79	.94	1.02	0.79	.99	0.75	0.78	.96
MnO	0.01	0.01	0.02	0.03	0.01	0.03	0.02	.00	.04	.01	0.01	.04	0.02	0.01	.02
K_2O	10.13	9.75	10.59	9.21	10.01	10.20	9.65	9.61	9.92	9.44	9.76	9.65	9.58	9.18	9.59
Na_2O	0.64	0.95	0.72	0.67	0.74	0.76	0.84	.88	.94	.92	1.00	1.03	1.04	1.04	1.09
BaO	n.d.	n.d.	0.15	n.d.	n.d.	n.d.	n.d.	--	--	--	0.15	--	n.d.	n.d.	--
H_2O	4.92	4.30	4.12	4.78	4.42	4.42	5.52	4.35	4.81	3.49	5.06	4.06	4.33	4.98	4.02
Formula--based on 22 Oxygen															
IV Si	6.178	6.038	6.163	6.193	6.192	6.191	6.143	6.100	6.186	6.212	6.143	6.219	6.176	6.140	6.190
Al	1.822	1.962	1.837	1.807	1.808	1.809	1.857	1.900	1.814	1.788	1.857	1.781	1.824	1.860	1.810
Tot. Al	5.543	5.713	5.467	5.497	5.459	5.465	5.611	5.622	5.463	5.488	5.575	5.443	5.558	5.620	5.481
VI Al	3.721	3.751	3.650	3.690	3.651	3.674	3.754	3.772	3.649	3.705	3.718	3.662	3.734	3.760	3.671
Fe	.114	.122	.121	.085	.047	.082	.120	.047	.076	.053	.114	.046	.115	.098	.040
Mg	.121	.126	.141	.179	.231	.168	.122	.174	.203	.177	.114	.225	.109	.107	.227
Ti	.076	.072	.043	.098	.105	.080	.062	.079	.068	.101	.079	.044	.074	.078	.094
Mn	.001	.001	.002	.003	.001	.003	.002	--	.005	.001	.001	.005	.003	.001	.002
Σ	4.033	4.071	4.007	4.055	4.035	4.029	4.060	4.072	4.001	4.036	4.026	4.035	4.044	4.044	4.034
XII K	1.716	1.478	1.788	1.636	1.685	1.717	1.662	1.610	1.669	1.571	1.657	1.612	1.608	1.584	1.602
Na	.165	.248	.185	.171	.190	.195	.217	.221	.238	.232	.257	.261	.266	.267	.277
Ba	--	--	.008	--	--	--	--	--	--	--	.008	--	--	--	--
Σ	1.881	1.926	1.981	1.807	1.875	1.912	1.959	1.831	1.907	1.803	1.922	1.873	1.874	1.851	1.879
Na/Na+K	.088	.129	.094	.095	.101	.102	.117	.121	.125	.129	.134	.139	.142	.144	.147
X_{Al}	.924	.923	.911	.910	.905	.912	.925	.926	.905	.918	.923	.910	.925	.930	.910
T °C - "B"	592	567	593	570		571	581	555	565	550	573	556	566		554
T °C - "C"	614	584	613	590		591	598	570	575	560	589	574	582		572

Specimen	SKZ--high grade - IIb								SKZ--low grade - IIa						
No.	M25	M36	M24	M21	M28	M31	M22	A8	M41	M49	M54	M67	M64	M69	M71
SiO_2	46.47	46.37	46.30	46.25	46.76	46.44	46.51	46.81	46.42	47.04	46.68	46.68	46.68	46.82	46.83
Al_2O_3	35.82	35.70	36.00	35.77	36.27	36.17	36.20	36.04	36.38	35.40	36.62	36.37	36.33	36.18	36.46
FeO	1.02	.71	.99	.61	.93	.97	.90	.98	.76	.74	.75	.67	.75	.78	.78
MgO	.63	.65	.61	.61	.54	.56	.57	.57	.54	.66	.55	.51	.54	.52	.55
TiO_2	.67	.66	.62	.89	.61	.81	.76	.79	.71	.48	.65	.62	.62	.68	.63
MnO	.01	.01	.02	.02	.01	.02	.02	.01	.01	.02	.01	.01	.02	.02	.01
K_2O	9.42	9.35	9.15	9.18	9.47	9.18	9.34	9.07	9.43	9.12	9.04	9.13	8.98	8.89	8.88
Na_2O	1.04	1.16	1.16	1.17	1.22	1.15	1.29	1.30	1.21	1.19	1.31	1.32	1.41	1.49	1.48
BaO	n.d.	.17	n.d.	n.d.	.21	n.d.	n.d.	n.d.	.21	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
H_2O	4.93	4.83	5.22	5.15	4.05	4.75	4.83	4.58	4.48	4.19	4.41	4.75	4.74	4.90	4.86
Formula-based on 22 Oxygen															
IV Si	6.153	6.133	6.140	6.138	6.144	6.140	6.135	6.163	6.119	6.157	6.127	6.148	6.147	6.150	6.139
Al	1.847	1.867	1.860	1.862	1.856	1.860	1.865	1.837	1.881	1.843	1.873	1.852	1.853	1.850	1.861
Tot.Al	5.591	5.616	5.627	5.595	5.617	5.626	5.624	5.592	5.652	5.617	5.665	5.646	5.639	5.625	5.657
VI Al	3.744	3.749	3.747	3.733	3.761	3.756	3.759	3.755	3.771	3.774	3.742	3.794	3.786	3.775	3.796
Fe	.114	.078	.109	.103	.102	.107	.099	.108	.084	.082	.082	.073	.083	.086	.086
Mg	.124	.127	.121	.120	.105	.109	.113	.112	.107	.130	.108	.101	.107	.102	.107
Ti	.076	.085	.072	.089	.060	.080	.075	.079	.071	.067	.064	.061	.061	.067	.062
Mn	.002	.001	.002	.002	.001	.003	.002	.001	.002	.002	.002	.001	.002	.002	.002
Σ	4.050	4.043	4.061	4.047	4.029	4.055	4.048	4.045	4.035	4.055	4.048	4.030	4.039	4.032	4.053
XII K	1.590	1.578	1.568	1.554	1.587	1.545	1.554	1.524	1.568	1.522	1.521	1.533	1.508	1.496	1.457
Na	.278	.298	.278	.302	.310	.301	.307	.332	.310	.303	.335	.339	.359	.331	.376
Ba	--	.009	--	--	.011	--	--	--	.011	--	--	--	--	--	--
Σ	1.868	1.885	1.846	1.856	1.908	1.846	1.861	1.856	1.889	1.825	1.856	1.872	1.867	1.827	1.833
Na/Na+K	.149	.159	.161	.163	.163	.163	.165	.179	.165	.166	.180	.181	.192	.203	.205
X_{Al} = Al VI	.924	.928	.927	.922	.933	.926	.928	.928	.934	.931	.937	.943	.938	.936	.936
T °C - "B"		561	561	540	550		563	557	556		558	556			553
T °C - "C"		536	536	526	544		577	570	569		571	566			565

Specimen	SKZ-low grade-IIb							SKZ-IIa + Ib						
No.	N23	M61	N3	N4	N7***	N8***	M56*	M53*	N5	M59*	N16*	N19*	N18*	M5-*	N14*
SiO_2	46.22	46.80	46.38	46.50	46.42	46.39	46.95	47.42	46.53	47.09	46.26	46.40	46.41	46.78	46.55
Al_2O_3	36.15	36.56	36.33	36.48	36.40	36.36	36.78	36.64	36.94	36.69	36.58	36.57	36.48	36.75	36.20
FeO	.70	.71	.73	.69	.75	.75	.72	.67	.77	.69	.59	.62	.60	.68	.73
MgO	.61	.52	.57	.57	.52	.51	.50	.48	.45	.53	.50	.49	.65	.46	.47
TiO_2	.62	.52	.51	.47	.54	.53	.48	.57	.44	.52	.60	.60	.53	.47	.53
MnO	.01	.02	.01	.01	.01	.00	.02	.02	.02	.01	.01	.01	.02	.01	.01
K_2O	8.81	8.52	9.12	8.91	8.90	8.79	8.80	8.64	8.55	8.72	8.72	8.65	8.70	8.53	8.39
Na_2O	1.63	1.67	1.36	1.44	1.54	1.64	1.57	1.62	1.64	1.66	1.68	1.74	1.70	1.74	1.88
BaO	.17	n.d.	.19	n.d.	.20	.20	n.d.	n.d.	n.d.	n.d.	.18	.17	.18	n.d.	.20
H_2O	5.16	4.74	4.87	5.00	4.78	4.94	4.25	4.46	4.80	4.15	4.96	4.82	4.82	4.65	5.13
Formula-based on 22 Oxygen															
IV Si	6.124	6.148	6.131	6.146	6.127	6.128	6.143	6.147	6.117	6.153	6.108	6.117	6.119	6.140	6.152
Al	1.876	1.852	1.869	1.864	1.873	1.872	1.857	1.853	1.883	1.847	1.892	1.883	1.881	1.860	1.848
Total Al	5.646	5.660	5.660	5.674	5.663	5.664	5.672	5.665	5.723	5.650	5.694	5.682	5.669	5.680	5.639
VI Al	3.770	3.808	3.791	3.810	3.790	3.792	3.815	3.812	3.840	3.803	3.802	3.799	3.788	3.815	3.791
Fe	.078	.078	.080	.076	.083	.083	.079	.073	.077	.075	.066	.068	.066	.075	.081
Mg	.120	.103	.113	.113	.102	.101	.097	.093	.088	.103	.098	.096	.122	.089	.092
Ti	.061	.052	.051	.046	.054	.053	.047	.056	.043	.051	.060	.060	.053	.046	.053
Mn	.001	.002	.001	.001	.001	.000	.002	.002	.002	.001	.001	.001	.002	.001	.001
Σ	4.030	4.043	4.036	4.046	4.030	4.029	4.040	4.036	4.050	4.033	4.027	4.024	4.036	4.036	4.018
XII K	1.489	1.428	1.537	1.500	1.499	1.482	1.469	1.444	1.434	1.453	1.468	1.455	1.463	1.428	1.415
Na	.418	.426	.350	.369	.394	.419	.398	.410	.417	.422	.429	.445	.434	.443	.482
Ba	.009	--	.010	--	.010	.010	--	--	--	--	.009	.009	.009	--	.010
Σ	1.916	1.854	1.897	1.869	1.903	1.911	1.867	1.854	1.851	1.875	1.906	1.909	1.906	1.871	1.907
Na/Na+K	.219	.230	.186	.197	.208	.220	.213	.221	.225	.225	.226	.234	.229	.237	.254
X_{Al}	.935	.942	.939	.942	.940	.941	.944	.948	.948	.94	.944	.944	.939	.948	.944
T°C - "B"		554		541			551	552	550					551	
T°C - "C"		566		553			561	563	560					561	

feldspars. The majority of muscovite analyses and all those of feldspar were partial analyses for K_2O, Na_2O, and CaO. To provide a check on the partial analyses, muscovite from 35 specimens was also analyzed by the oxide method of Bence and Albee (1968). The high level of correlation between the two sets of data is illustrated by an average deviation of 0.005 in the Na/Na + K ratio and the fact that only in four samples does the Na/Na + K ratio vary in excess of 0.010.

In addition, oxide compositional data from five USZ muscovite–plagioclase pairs were obtained using the University of Massachusetts automated ETEC electron microprobe. The method of analysis, procedures, and standards were similar for analyses done at both laboratories. The analytical details have been described in Cheney (ms) and Guidotti (1974).

Based on the analytical limits of the probe (see Albee, 1972, p. 3252-3254) and on our observed *maximum* relative (mean) standard deviations of 0.006 for K and 0.050 for Na in muscovite and 0.022 for Ca, 0.011 for Na, and 0.07 for K in plagioclase, we have assumed a minimum precision of plus or minus 1 mole percent (±0.010 in mole ratios) for the data employed in this study.

SYSTEMATIC MINERALOGY

This section summarizes observations and analytical data pertaining to minerals comprising AKNa–Ca assemblages Ia through IV. Most of the compositional data are summarized in tables 1, 2, and 4 which are based on table 8 and appendices A and B of Cheney (ms). However, samples of Cheney (ms) containing textural or chemical evidence of disequilibrium have been excluded; for example, specimens containing partially resorbed sillimanite. In addition, table 1 of this report contains analyses of some muscovites for which we have not determined the composition of the coexisting plagioclase. They are included in table 1, because they extend the data base for characterizing the regional compositional variation of muscovite in the Puzzle Mountain area.

Aluminosilicates.—Both sillimanite and andalusite occur in the pelites of the Puzzle Mountain area. Sillimanite, primarily as fibrolite, ranges from an average of 3 modal percent in the LSZ to 9 percent in the SKZ. Typical habits of fibrolite vary from radiating bundles of fibers in the LSZ to bundles of fibers parallel with the biotite and muscovite which serve to define the primary foliation in higher grade rocks. Less common are sporadic occurrences of coarsely crystalline sillimanite (up to 1.5 mm long). Such coarse sillimanite is more common in higher grade rocks but never replaces fibrolite as the dominant type.

Optically continuous, skeletal remnants of andalusite megacrysts occur mainly in LSZ specimens. As in other areas of Northwest Maine

TABLE FOOTNOTE

†† Roman numerals refer to assemblages specified in text. ††† Specimen does not contain sillimanite but is included here because of its apparent high Al bulk composition (C. F. Guidotti and Sassi, 1976). * Peculiar value. + Assemblage IB $X_{Al} = Al^{VI}/\Sigma VI$; T°C – "B" = calculated by method B – table 4, T°C – "C" = calculated by method C of table 4 but using M2 site composition for each individual specimen.

(see Guidotti, 1974), the occurrence of andalusite is not systematically related to specific mineral assemblages, isograds mapped on the basis of other phases, or systematic compositional relations among solid solution phases. Hence, andalusite is considered to be a metastable relic from an earlier metamorphism (Guidotti, 1970b).

Quartz and feldspar.—Quartz plus plagioclase comprise 45 to 55 percent of most rocks whereas primary K-feldspar occurs in only 5 SKZ specimens. All three minerals are typically confined to the groundmass. Exceptions include coarse plagioclase, K-feldspar, and polygonized quartz from the leucocratic portions of migmatitic gneisses occurring in the higher grade portions of the terrain.

Most plagioclase is unzoned, but some is weakly (rarely moderately) zoned. Such zoning is typically normal, but reverse zoning was observed in some specimens, especially in higher grade rocks. Although rim and core compositions determined for ten specimens confirm the variable and typically minor plagioclase zoning (see table 2), most plagioclase compositions used in this study represent rim compositions. The mole percent An in plagioclase from the 116 specimens is summarized with respect to metamorphic grade and mineral assemblage in tables 2 and 4 and illustrated on figure 2. Two points are pertinent here:

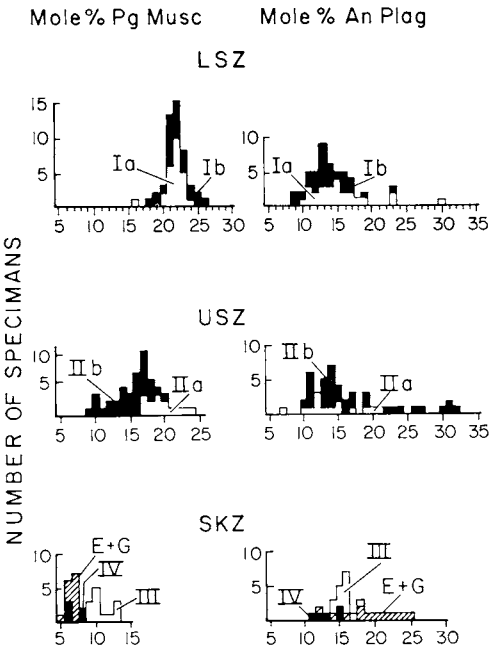

Fig. 2. (A) Mole percent paragonite in muscovite and (B) mole percent anorthite in plagioclase as a function of metamorphic grade. Roman numerals refer to assemblages designated in text. E & G refers to data from Evans and Guidotti (1966).

1. There is little systematic change in the composition of plagioclase, in terms of percent Ab or percent An, as a function of metamorphic grade. However, the percent Or is systematically higher in plagioclase from K-feldspar-bearing rocks (0.9-1.1 percent versus 0.2-0.9 percent).
2. Although overlap exists, plagioclase from assemblage IV tends to be slightly more sodic, averaging 13.0 percent An, than those from assemblage III which average 16.1 percent An.

This similarity and overlap in composition of plagioclase from SKZ specimens containing either assemblage III or IV was also reported by Evans and Guidotti (1966). However, the absolute compositions of plagioclase considered in that study were slightly, but systematically more calcic than those considered here (see fig. 2). This may reflect the slightly higher grade nature of the specimens studied by Evans and Guidotti (1966). In contrast to this report, few of the SKZ specimens considered by them were collected in the vicinity of the sill + ksp isograd.

Potassium feldspar varies modally from a trace to 15 percent. It is typically untwinned and rarely contains exsolution lamallae. In most specimens the K-feldspar is in physical contact with muscovite and sillimanite, although not necessarily at the same location. However, in two specimens the K-feldspar was confined, in trace quantities, to quartz/plagioclase-rich portions of the rock. The composition of K-feldspar, ranging from 11.5 to 14.6 percent Ab (averaging 13.1 percent Ab), is similar to that observed by Evans and Guidotti (1966) and Guidotti, Herd, and Tuttle (1973).

Muscovite.—In addition to the randomly oriented plates and laths of muscovite which comprise the previously noted polycrystalline pseudomorphs after staurolite, muscovite also occurs in two other textural habits: (1) foliation-defining groundmass laths are most common in the LSZ and decrease modally with increasing grade, becoming rare in the SKZ; and (2) muscovite megacrysts, up to 2 cm across, commonly lying at high angles to the foliation and occurring primarily in the SKZ and higher grade portion of the USZ. These megacrysts *appear* to form by recrystallization and coalescence of the muscovite in the pseudomorphs and perhaps by concomitant coalescence of groundmass folia.

Partial analyses of these coexisting but texturally different muscovites from 16 LSZ[1] and 16 USZ specimens from the Puzzle Mountain area were given by Cheney and Guidotti (1973). Figure 2 of that report established the virtual identity of TiO_2 and Pg contents for these coexisting, texturally different muscovites. In addition, the Pg content systematically decreases and weight percent TiO_2 systematically increases with increasing metamorphic grade. These observations substantiated Guidotti's (1968) suggestion that the pseudomorphs after staurolite are equilibrium

[1] Note that due to the definition of the LSZ-USZ isograd presented earlier in this report, two specimens (B45 and A75) included in the USZ of the earlier paper (Cheney and Guidotti, 1973) have been reassigned to the LSZ.

TABLE 2

Compositional data* and temperatures** derived from equation (14) for specimens shown on figure 1

2A. LSZ-Assemblage Ia

All specimens contain sill + st + biot + musc + plag + qtz ± ilm ± gar ± po ± graphite

	Musc	Plag				T – °C	
	%Pg	%Ab	%Or	%An	%Pg/%Ab	B	C
D40	23.5	89.8	.6	9.6	.262	551	562
D51	23.3	85.0	.5	14.5	.274	547	557
D37	23.2	88.2	.5	11.3	.263	550	56
M5	23.0	87.3	.7	12.0	.263	549	560
A78	22.6	83.7	.5	15.8	.270	546	559
D39b	22.5	86.9	.6	12.5	.259	549	560
D49	22.4	85.3	.4	14.3	.263	548	559
D38	22.4	86.7	.5	12.8	.258	549	561
D43	22.4	88.5	.3	11.2	.253	551	562
D47	22.3	88.4	.4	11.2	.252	551	562
B-4[4]	22.2	87.5	.4	12.1	.254	550	561
B-1	22.2	86.5	.5	13.0	.257	549	560
D50	22.0	84.2	.6	15.2	.261	547	558
A79b	22.0	85.4	.5	14.1	.258	548	559
B32	21.6	82.0	.4	17.6	.263	545	556
B31b	21.5	84.2	.5	15.3	.255	548	559
G60[3]	21.4	83.8	.5	15.7	.255	548	558
D41	21.4	85.3	.5	14.2	.251	549	560
D-2	21.2	82.2	.5	17.3	.258	546	557
A80	21.1	84.5	.6	14.9	.250	549	560
B33	20.7	80.3	.5	19.2	.258	545	555
C6	20.7	77.0	.5	22.5	.269	541	552
N-4	20.2	76.2	.4	23.4	.265	541	551
M3[4]	20.2	82.5	.5	17.0	.245	546	559
C5[3]	20.4	69.8	.5	29.7	.235	541	555

2B. LSZ-Assemblage Ib

	Musc	Plag				T – °C	
	%Pg	%Ab	%Or	%An	%Pg/%Ab	B	C
B29	25.6	86.4	.4	13.2	.296	546	557
M55	25.2	87.6	.5	11.9	.288	547	558
D53	25.0	90.4	.4	9.1	.277	551	561
M54	24.4	90.2	.4	9.4	.271	551	562
M57	23.8	87.0	.5	12.5	.274	548	559
M12	23.1	86.9	.4	12.7	.266	549	560
M58	23.1	84.2	.5	15.3	.274	546	557
M53	22.5	89.0	.4	10.6	.253	552	563
A26	22.1	86.6	.5	12.9	.255	550	561
D67	22.1	89.3	.6	10.2	.247	552	563
B45	21.8	89.0	.4	10.6	.245	553	564
D70[4]	21.8	87.0	.5	12.5	.251	550	561
N20	21.5	83.3	.4	16.3	.258	546	558
A74	21.2	85.5	.4	14.1	.248	550	561
D6	21.0	85.2	.5	14.3	.246	550	561
A75	21.0	83.8	.4	15.8	.251	548	559
M56	20.9	86.7	.4	12.8	.241	551	562
B30	20.8	87.5	.5	12.0	.238	552	563
D53	20.5	80.8	.5	18.7	.254	546	556
D35	20.5	83.4	.6	16.0	.246	548	559
B28	19.8	82.8	.5	16.7	.239	549	560
D18b	18.6	85.9	.6	13.5	.217	555	566
D2	18.5	82.2	.5	17.3	.225	551	562
D36	18.2	76.5	.6	22.9	.238	545	556

2C. USZ-Assemblage IIA

All specimens contain: sill + biot + musc + plag + qtz + ilm ± gar ± graphite ± po ± "remnant st"

	Musc	Plag				T – °C	
	%Pg	%Ab	%Or	%An	%Pg/%Ab	B	C
M61	24.4	92.4	.4	7.2	.264	553	565
D80	23.2	89.4	.4	10.2	.260	551	563
M47	21.6	87.2	.4	12.3	.248	551	563
D8	20.5	84.5	.5	15.0	.243	550	562
D83	19.9	85.9	.5	13.6	.232	552	564
M42	19.9	87.8	.4	11.7	.227	554	567
M67	18.7	86.2	.5	13.3	.217	555	567
D54	18.6	79.6	.6	19.8	.234	547	560
D13	18.5	80.3	.5	19.2	.231	549	561
M66	18.3	84.9	.5	14.7	.216	554	567
M40	17.5	88.2	.5	11.3	.198	560	572
M46	17.4	88.1	.4	11.5	.198	560	572
B72	16.7	78.4	.6	21.0	.231	551	563
		78.8	.6	20.6			
M41	16.5	82.2	.6	17.3	.201	556	568

2D. USZ-Assemblage IIB

All specimens contain sill + biot + musc + plag + qtz + gar ± cord ± po ± ilm or rutile ± graphite-

	Musc	Plag				T – °C	
	% Pg	%Ab	%Or	%An	%Pg /%Ab	B	C
D29	19.5	88.8	.5	10.7	.220	556	572
M35	19.2	85.3	.6	14.1	.225	553	569
A8	17.9	85.4	.3	14.4	.210	556	572
B77	17.8	85.4	.4	14.2	.208	556	572
M34	17.5	87.0	.5	12.5	.201	558	574
D12	16.8	72.7	.5	26.8	.231	543	558
B47	16.7	83.1	.5	16.4	.201	556	572
M22	16.6	88.8	.6	10.6	.187	563	579
M23	16.6	85.6	.5	13.9	.194	559	575
M36	16.6	85.7	.5	13.8	.194	559	575
Osm-3	16.5	85.2	.5	14.3	.194	559	575
M85	16.2	89.7	.5	9.8	.109	565	581
M24	16.1	86.2	.6	13.2	.187	561	577
M37	16.0	82.7	.5	16.8	.193	557	574
M86	15.7	86.9	.6	12.5	.181	563	580
M28	15.6	77.4	.6	22.0	.202	552	568
B22	15.5	82.2	.6	17.2	.189	558	575
		83.2	.7	16.1			
Osm-2b	15.3	80.3	.5	19.2	.191	557	573
M-83	15.3	86.8	.6	12.6	.176	564	580
P85-u[5]	14.9	76.4	.2	23.4	.195	553	569
		74.5	.2	25.3			
A18	14.3	83.7	.7	15.6	.171	564	581
B17	14.2	85.1	.2	14.7	.167	566	583
P85-s[5]	14.0	75.3	.4	24.3	.186	555	571
M30	13.7	88.0	.8	11.2	.136	572	589
P85-q[5]	12.8	67.9	.3	31.8	.189	550	566
		66.4	.2	33.4			
P85-h[5]	12.6	70.5	.3	29.2	.179	554	571
P85-r[5]	12.0	68.8	.4	30.8	.174	555	572
		66.7	.3	33.0			
M32	11.7	88.1	.6	11.3	.133	581	599
M67	11.0	88.5	.6	10.9	.124	586	604
D20-b	10.2	73.4	.6	26.0	.139	572	589
D20-a	9.6	81.0	.5	18.5	.119	586	604
		80.9	.5	18.6			
D19	9.5	68.6	.6	30.8	.138	570	587
		68.3	.6	31.1			
B11	9.4	85.0	.5	14.5	.111	593	611

2E. SKZ-Assemblage III

All specimens contain sill + biot + musc + plag + qtz ± gar ± ilm ± graphite ± po

	Musc	Plag				T – °C	
	%Pg	%Ab	%Or	%An	%Pg /%An	B	C
E5	12.9	81.1	.4	18.5	.159	567	584
E1	12.8	84.2	.6	15.3	.152	511	588
E2	12.8	84.7	.5	14.8	.151	572	589
E3	11.7	84.3	.6	15.1	.139	577	594
E1	10.9	83.7	.6	15.6	.130	581	598
D58	9.6	85.1	.6	14.3	.113	592	610
		84.9	.8	14.4			
A99	9.5	85.7	.6	13.7	.111	593	611
D91	9.4	83.6	.9	15.5	.112	591	609
		82.6	1.1	16.4			
D90-b	9.0	83.4	.7	15.8	.108	594	613
E6	8.9	84.9	.7	14.4	.105	597	615
E8	8.8	83.8	.5	15.4	.105	597	615
D90-a	8.5	83.2	1.0	15.8	.102	599	617
D92	8.5	83.8	.7	15.5	.101	599	618

2F. SKZ-Assemblage IV

All specimens contain sill + ksp + biot + musc plag + qtz ± gar ± ilm ± graphite ± po

	Musc	Plag				T – °C	
	%Pg	%Ab	%Or	%An	%Pg/%Ab	B	C
D57	8.4	84.6	1.0	14.5			
		88.1	1.0	10.9	.095	606	627
D59	7.7	86.1	.9	13.0	.089	611	632
B7	6.2	84.5	1.0	14.4	.073	628	650
D89	6.2	86.9	1.0	12.1	.071	631	654
B69	6.1	84.4	1.1	14.5	.072	629	652

features of a prograde metamorphic event superimposed on a preexisting metamorphic terrain.

Muscovite compositional data from all specimens considered here are listed in tables 1 (oxide analyses) and 2 (A-site compositions). These data are summarized as a function of metamorphic grade and assemblage in table 4 and on figure 2. Consideration of these data leads to the following generalizations:

1. The average mole percent Pg in muscovite systematically decreases from 21.7 percent in the LSZ to 16.5 percent in the USZ to 6.9 percent in the SKZ (assemblage IV only).

2. Within the LSZ there is no systematic change in the Pg content of muscovite with increasing grade. However, Pg contents of the lower grade muscovites are generally restricted to a rather narrow compositional range (21.0 and 24.0 percent Pg). This narrow compositional interval is consistent with our treatment below of these assemblage Ia rocks as having equilibrated at a relatively fixed P, T, and f_{H_2O}, that is, as if all were from a single outcrop.

3. *Within the SKZ*, assemblage IV muscovite is systematically more potassic than that of assemblage III, averaging 6.9 and 10.3 percent Pg respectively. A similar observation (see fig. 2) was made by Evans and Guidotti (1966, p. 55). They noted that this is the reverse of what is expected if intermixing of assemblages III and IV resulted simply from the stabilization of assemblage IV by the low An component in plagioclase.

Particularly important to arguments given below is the relative consistency shown in table 1 of M2 site (octahedral site) occupancy in muscovite from a given assemblage. Furthermore, excluding the A site, there is little change in the composition of muscovite as a function of metamorphic grade. This latter point is well shown by comparing average site occupancy of various cations from 13 LSZ and 21 USZ (assemblage IIb only) specimens of table 1:

Assemblage	$(Mg + Fe)^{VI}$	Ti^{VI}	Al^{VI}	Si^{IV}	Total Al
USZ (IIb)	0.237	0.082	3.720	6.161	5.558
LSZ (Ia and Ib)	0.176	0.052	3.804	6.132	5.672
Change with rise of grade	+0.061	+0.030	−0.084	+0.029	−0.114

TABLE FOOTNOTE

* All compositional data are in terms of mole percent. Where multiple plagioclase analyses are given, the top one is for the rim and the bottom is for the core.

** Temperatures were derived from eq (14) using the Margules parameters of Chatterjee and Froese (1975), (B) neglecting the M2 site correction, (C) using the M2 site correction (see tables 3 and 4).

3. Specimen does not contain sillimanite. It is included here because of its apparent high Al bulk composition (Guidotti, 1973). Hence the temperatures are upper limits for these specimens.

4. Specimen contains "remnant" andalusite.

5. Specimen location not shown on figure 1. The sample is from the "Bridgeman" drill core taken from the top of Whitecap Mountain 3.5 km N 45° E of specimen G60.

These relationships allow generalization of our limited M2 site data (and that of Evans and Guidotti, 1966, p. 37-38) to our summary data base given in table 4.

PARTITIONING

Introduction.—The theory of partitioning of independently variable phase components (actual phase components of Thompson,[2] 1959, p. 445-446) is an extension of the equilibrium conditions existing in a multiphase, heterogeneous, closed system described by Gibbs (reprinted 1961, p. 63-72). As developed by Thompson (1959, p. 449), the *completely general* criterion of equilibrium pertaining to the chemical potentials in such a system is:

That, for every stoichiometric equation among the actual phase components in the form

$$\sum_{i} \nu_i \, M_i = 0 \tag{3}$$

there exists an analogous relation among the chemical potentials vis a vis:

$$\sum_{i} \nu_i \, \mu_i = 0 \tag{4}$$

To be emphasized is that the general condition (4) includes the rather trivial case of the uniformity of μ_i among each and every phase in which component i is independently variable, that is:

$$\mu_i^{\alpha} - \mu_i^{\beta} = 0 \tag{5}$$

As pointed out by Albee (1965, p. 274), eq (5) is of limited use in geologic application due to crystallographic and stoichiometric constraints on the compositional variation of minerals. Consequently, Albee (1965, p. 275) introduced the concept of "exchange reactions" which are type (4) expressions relating the chemical potentials of actual phase components. The choice of "exchange components" (for example, KNa_{-1}; Brady, 1975) as actual phase components reduces type (4) conditions to type (5) relations among the chemical potentials when written on a cation basis (see Thompson and Thompson, 1976). Hence, the conditions of equilibrium among the chemical potentials of phase components, and consequently the functional form of the expression describing the partitioning of these components, are dependent on the actual components necessary and sufficient to describe the system of interest. Moreover, as discussed by Gibbs (reprinted 1961, p. 70-79), consideration of an additional phase does not alter the preexisting equilibrium conditions, although additional eqs (4) will be required to characterize completely

[2] As defined in Thompson (1959, p. 445), an actual component of a phase is any material of specified composition but unspecified physical state that may, independently of other components, be both added to *and* subtracted from a phase, in finite amounts, without destroying the homogeneity of the phase.

the equilibrium of the system. Thus, simple conditions of equilibrium such as (4), and the functional consequences thereof, remain valid in complex heterogeneous systems.

Plagioclase–muscovite equilibrium.—The plagioclase feldspars from assemblages I to IV can be approximated as Ab–An binary solutions inasmuch as the Or content never exceeds 1.2 mole percent (0.3 wt percent K_2O) and is less than 1.0 mole percent in non K-feldspar bearing specimens (table 2). Consequently, in these rocks, the Ab component in plagioclase is related to the Pg component in muscovite *only* by the stoichiometric equation:

$$0 = H_2O + (NaAlSi_3O_8)_{plag} + (Al_2SiO_5)_{sill} - (NaAl_3Si_2O_{10}(OH)_2)_{musc} - (SiO_2)_{qtz} \quad (6)$$

The corresponding equilibrium relation among the chemical potentials is:

$$0 = \Delta\mu = \mu_{H_2O} + \mu_{Ab}^{plag} + \mu_{Al_2SiO_5}^{sill} - \mu_{SiO_2}^{qtz} - \mu_{Pg}^{musc} \quad (7)$$

Substituting the relation

$$\mu_i = \mu_i^\circ + RT \ln a_i \quad (8)$$

for each phase in equation (7) yields

$$0 = \sum_i \nu_i \mu_i = \sum_i \nu_i \mu_i^\circ + RT \ln \left[\frac{a_{Ab}^{plag} \cdot a_{SiO_2}^{qtz} \cdot a_{H_2O}}{a_{Pg}^{musc} \cdot a_{Al_2SiO_5}^{sill}} \right]. \quad (9)$$

Adopting the conventional standard states, pure solids at T and 1 bar so that $a_{SiO_2} = a_{Al_2SiO_5} = 1$, ideal H_2O gas at T and 1 bar, and assuming that the molar change of the solids ($\Delta\bar{V}^\circ$) is independent of P and T, allows us to rewrite eq (9) as:

$$\Delta\mu^\circ + \Delta\bar{V}^\circ_s (P - 1) = RT \ln \frac{a_{Pg}^{musc}}{a_{Ab}^{plag} \cdot f_{H_2O}} \quad (10)$$

Substituting $(X_i^\phi \gamma_i^\phi)^N$ for a_i^ϕ (see Albee, 1972, p. 3263), where N is the number of crystallographic sites used in writing eq (6), γ_i^ϕ is the activity coefficient and x_i^ϕ is the mole fraction of component i in phase ϕ, results in:

$$\frac{\Delta\mu^\circ}{RT} + \frac{\Delta\bar{V}^\circ_s (P-1)}{RT} = \ln \left(\frac{X_{Pg}^{musc}}{X_{Ab}^{plag}} \right) \left(\frac{1}{f_{H_2O}} \right) \left(\frac{\gamma_{Pg}^{musc}}{\gamma_{Ab}^{plag}} \right) \quad (11)$$

The mole fraction quotient in eq (11) is defined as the $Kd_{Pg\text{-}Ab}^{musc\text{-}plag}$, and it is dependent upon P, T, and f_{H_2O}. This Kd is a more useful petrogenetic indicator than the commonly used Pg content of muscovite (compare Guidotti and Sassi, 1976, p. 110-118), because the Pg content of muscovite from assemblages such as I to III decreases, at a specified P, T, and f_{H_2O}, with decreasing Ab content (or increasing An content) of the plagioclase (see inset on fig. 3).

Stoichiometric relation (6) is the net transfer equilibrium (continuous reaction) governing the compositional variation of the solid solution phases from assemblages I to III. For assemblage IV, eq (6) describes the net transfer equilibrium pertaining to the sill + musc + plag tie plane, which is one of four unique tie planes that define a four phase volume (assemblage IV) in the AKNa–Ca subsystem (compare Evans and Guidotti, 1966, p. 52). The movement of the four phase volume, by reaction (2) with increasing T or decreasing f_{H_2O}, allows successively An richer plagioclase and correspondingly K-richer muscovites from assemblages such as III to coexist with ksp. Therefore, the muscovite–plagioclase tie lines in the four phase volume are parallel to those of the three phase tie planes. The composition of muscovite is related to the composition of plagioclase in assemblage IV in the same manner as in assemblages I to III even though the compositions of the solid phases in assemblage IV are a function only of the intensive parameters.

Data previously introduced have indicated that although the Pg content of muscovite decreases with increasing metamorphic grade, the An content of plagioclase (in assemblages I-III) seems to be largely independent of grade. These observed phase relations imply that the change in plagioclase composition resulting from the potassic enrichment of muscovite with grade is small. This point is further illustrated by balancing, *on a gram-atom basis,* eq (6) in the context of observed changes in muscovite composition from assemblages Ia through IV:

$$20\ K_{0.78}Na_{0.22}Al_3Si_3O_{10}(OH)_2 + 0.48\ SiO_2 = 2.08\ NaAlSi_3O_8$$
$$+ 1.28\ Al_2SiO_5 + 16.80\ K_{0.93}Na_{0.07}\ Al_3Si_3O_{10}(OH)_2 + 0.48\ H_2O$$

The relatively small amount of Ab component produced by the potassic enrichment of muscovite coupled with minor variation in the calcium content of pelites form a single terrain, likely explains the successful use of Pg content of muscovite as a qualitative monitor of metamorphic grade in petrologically well characterized terrains (see Guidotti and Sassi, 1976).

APPLICATION

From the preceeding discussion, the $Kd_{Pg\text{-}Ab}^{musc\text{-}plag}$ should vary in a systematic manner as a function of metamorphic grade, the composition of muscovite from assemblages I to III should fluctuate (at a specified P, T, and f_{H_2O}) with variation in the An content of the coexisting plagioclase, and the compositions of solid solution phases from assemblage IV should be dependent only on intensive parameters (primarily T and f_{H_2O}). Figure 3 illustrates these relations for muscovite–plagioclase pairs from the Puzzle Mountain area. The data portrayed on this figure can be summarized as follows:

1. Data points representing the approximately isothermal assemblage Ia muscovite–plagioclase pairs define a line over a relatively limited composition range. The slope of this line, 0.286, provides an estimate of the

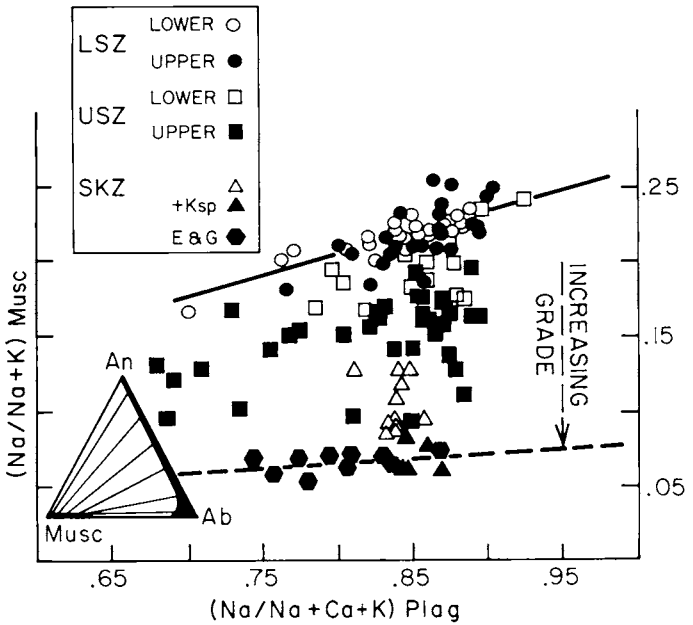

Fig. 3. Graphical illustration of the relation between the mole fraction Pg in muscovite and mole fraction Ab in plagioclase as a function of metamorphic grade. The solid line is a least squares fit for the LSZ assemblage Ia pairs (table 2A) only. The dashed line is a least squares fit for SKZ assemblage IV pairs (table 2F and Evans and Guidotti (E & G), 1966). The triangular inset is a schematic projection from sillimanite in the systems $CaO–Na_2O–K_2O–Al_2O_3–SiO_2–H_2O$.

compositional influence of plagioclase on coexisting muscovite. Hence a 10 mole percent decrease in the Ab (or increase in the An) content of plagioclase results in an approx 3.0 mole percent decrease in the Pg content of the coexisting muscovite. The assemblage Ia muscovite coexisting with albite in the Ca free system would contain, by extrapolation of the best fit line, 26.3 mole percent Pg.

2. As previously discussed, the dependence of muscovite composition on plagioclase composition can also be approximated from assemblage IV data points of this study combined with those of Evans and Guidotti (1966). The slope of a least squares line through these data points is 0.078. Thus at higher grade (the SKZ), a 10 mole percent decrease in the Ab content of plagioclase results in an approx 1 mole percent decrease in the paragonite content of the coexisting muscovite. Moreover the hypothetical muscovite coexisting with albite would contain approx 8 mole percent Pg. This is also the composition of muscovite at the AKNa isograd marking the initial compatibility of sillimanite and K-feldspar in the Ca-free system.

3. Although some overlap exists, there is clearly a sequential relation between the $Kd_{Pg\text{-}Ab}^{musc\text{-}plag}$ for individual specimens and increasing metamorphic grade.

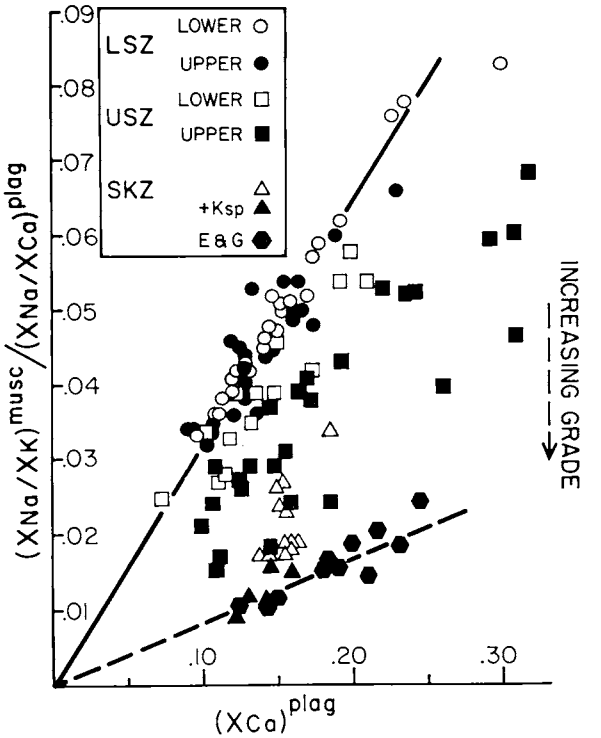

Fig. 4. Relationship between plagioclase and muscovite composition with changing metamorphic grade. The lines (as on fig. 4) are least squares fits which have been constrained to pass through the origin. The slope on these lines is equal to the Na/K ratio in muscovite divided by the mole fraction of albite in plagioclase.

4. SKZ specimens containing assemblage IV consistently plot to the high grade side of those specimens containing assemblage III. Most of the SKZ data from the Puzzle Mountain area are from an approximately isothermal area, therefore suggesting that assemblage IV specimens equilibrated at lower values of f_{H_2O} than assemblage III specimens as originally proposed by Evans and Guidotti (1966).

Figure 4 illustrates concisely the relationships among muscovite composition, metamorphic grade, and An content of plagioclase. The advantage of this diagram over the more conventional figure 3 is that, due to the choice of plotting parameters, figure 4 can be more easily interpreted in a fashion similar to Mg/Fe plots of Albee (1965, p. 276). Specifically, points representing plagioclase–muscovite pairs which equilibrated at a common P, T, and f_{H_2O} should, due to variation in the An content of the plagioclase, plot along a line radiating from the origin. Variations in P, T, or f_{H_2O} will be reflected by changes in the slope of these lines, principally due to variation in Pg content of the muscovite. Due to the removal of the compositional degree of freedom, data points representing

muscovite–plagioclase pairs from assemblage IV specimens will plot along a line if the compositional variation is due to the fluctuation in intensive parameters. Scatter of assemblage IV data points likely results from disequilibrium and/or analytical error. Note, however, that it is not generally possible to determine from figure 4 (or 3) if the scatter of data points from assemblages I to III reflects an equilibrium phenomenon (that is, locally variable f_{H_2O}) or disequilibrium. The well-defined linear trends on figure 4, relative to figure 3, are in a sense only apparent as the slopes of the lines on figure 4 are equivalent to $Na/K^{musc}/X_{Ab}^{plag}$ (the X_{An}^{plag} terms cancel), and this is related in a non-linear fashion to the slopes of the lines on figure 3 because $X_{Na}^{musc} = [(Na/K)/(1 + (Na/K))]$.

Evaluation of intensive parameters.—By assuming an asymmetric solution model for Mu–Pg solid solution, γ_{Pg}^{musc} can be related to polythermal, polybaric margules parameters (W_{Pg} and W_{Mu}) (Thompson, 1967, eq 80a) by the following relation:

$$\ln \gamma_{Pg}^{musc} = \frac{(X_{Mu}^{musc})^2}{RT} [W_{Pg}^{musc} + 2X_{Pg}^{musc} (W_{Mu}^{musc} - W_{Pg}^{musc})] \tag{12}$$

which upon substitution into eq (11) yields:

$$\left\{\frac{\Delta\mu^\circ}{RT} + \frac{\Delta\bar{V}_s^\circ (P-1)}{RT}\right\} =$$

$$\left\{ \ln X_{Pg}^{musc} - \ln X_{Ab}^{plag} - \ln \gamma_{Ab}^{plag} - \ln f_{H_2O} + \right.$$

$$\left. \frac{(X_{Mu}^{musc})^2}{RT} [W_{Pg}^{musc} + 2X_{Pg}^{musc} (W_{Mu}^{musc} - W_{Pg}^{musc})] \right\} \tag{13}$$

If the lithostatic pressure is known or estimated by an independent method, then the temperature dependent variables in eq (13) ($\Delta\mu^\circ$, ln f_{H_2O}, W_{Pg}^{musc}, and W_{Mu}^{musc}) can be approximated as linear functions of temperature in the form $\alpha/T + b$, as indicated in table 3. Thus eq (13) can be explicitly solved for the temperature of equilibrium of each sample if, in addition to the compositions of muscovite and plagioclase, we use Orville's (1972, p. 265) values for γ_{Ab}^{plag} (see table 3). However, as pointed out by Ferry (1976, p. 141), the activity expression for natural micas, used in quantitative expressions, requires modification to account for the deviation in M2 site composition (see table 4) from the idealized formula used in writing the stoichiometric eq (3) (see table 3). Hence, assuming ideal mixing over the M2 site of muscovite results in the substitution of $(X_{Pg}^{musc}\ \gamma_{Pg}^{musc})\ (X_{Al,M2})^2$ for a_{Pg}^{musc} then eq (13) becomes:

$$\left\{\frac{\Delta\mu^\circ}{RT} + \frac{\Delta V_s^\circ\,(P-1)}{RT}\right\} =$$

$$\left\{ \ln X_{Pg}^{musc} - \ln X_{Ab}^{plag} - \ln f_{H_2O} + \ln (X_{Al,M2}^{musc})^2 \quad + \right.$$

$$\left. \frac{(X_{Mu}^{musc})^2}{RT}\left[W_{Pg}^{musc} + 2X_{Pg}^{musc}\,(W_{Mu}^{musc} - W_{Pg}^{musc})\right] \right\} \qquad (14)$$

Temperatures derived from eq (14) will be maximum values for a specific specimen owing to the likelihood that $a_{H_2O} < 1$ during the equilibration of most metamorphic rocks. Moreover, these temperatures are relative values at best due to the uncertainty and arbitrary choice among the variables listed in table 3 (compare Eugster and others, 1972; Chatterjee and Froese, 1975; Pigage, 1976). For example, pseudobinary, isobaric, T–X diagrams have been derived from the polythermal-polybaric Margules parameters of Eugster and others (1972) by Thompson (1974) and a modified version of these parameters by Chatterjee and Froese (1975). From these diagrams, muscovite coexisting with sillimanite + albite + quartz should contain a maximum of approx 20 mole percent Pg. However, assemblage Ia muscovite averages 22 mole percent Pg, and the hypothetical muscovite from the Ca-free assemblage (Ia) would contain 26 mole percent Pg.

TABLE 3
Input parameters for eq (14)

Variable (Eqt. 13)	Value	Source	Comments
P	3500 bars	See text	
$\ln f_{H_2O}$	$-2{,}363.74/T + 10.22$	Burnham, Holloway and Davis (1969)	The T dependent equation was obtained by an isobaric least squares fit of $\ln f_{H_2O}$ versus $1/T^\circ K$ over a relatively narrow (500°C–700°C) T range.
$\ln X_{Ab}^{plag}$	1.0	Orville (1972)	Assumes coupled substitution of $CaAl^{+}{}_{-}NaSi$. Although Chatterjee used synthetic high albite (not analbite) and the plagioclase feldspars considered here are approximately low albites (Guidotti, 1970a), no structural state correction has been made (compare Pigage, 1976).
$\Delta V_s^\circ/R$	.0538 deg/bar	Chatterjee and Froese (1975)	
$\Delta\mu^\circ/RT$	$11{,}100.71/T - 20.47$	Chatterjee and Froese (1975), and Holdaway (1971)	The $\Delta\mu^\circ/T$ term is based on the work of Chatterjee (1972) and corrected to sillimanite on the basis of Holdaway's (1971) results.
$(W_{Pg}^{musc})/RT$	a) $1{,}695.70/T + .0854$	Eugster and others (1972)	
	b) $1{,}750.95/T + .0854$	Chatterjee and Froese (1975)	
$(W_{Mu}^{musc})/RT$	a) $2{,}317.03/T + .1990$	Eugster and others (1972)	
	b) $2{,}147.98/T + .1990$	Chatterjee and Froese (1975)	
$(X_{Al,M2})^2$	As per Table 5	Ferry (1976)	Assumes ideal mixing over the M2 site in muscovite. This term is necessitated in numerical calculations based on Chatterjee's (1972) work because synthetic stoiometric paragonites were used in the experiments (that is, $X_{Al,M2} = 1$)

Although the temperatures derived from eq (14) are relative values, they can be used to place qualitative limits on temperature gradients and variations of f_{H_2O} within a single metamorphic terrain.

Estimate of pressure.—Ideally, three unique stoichiometric equations such as (3) could be used to solve simultaneously for values of P–T and f_{H_2O}. However, uncertainties in thermodynamic parameters and solution models do not seem to warrant such a procedure. Consequently, we have assumed a pressure of 3500 bars which is predicated on Holdaway's (1971) aluminosilicate triple point and the following considerations:

1. The occurrence of remnant andalusite with sillimanite in several LSZ specimens suggests a maximum pressure equal to the aluminosilicate triple point; that is, approx 3.8 kb (Holdaway, 1971).
2. An estimate of the minimum pressure can be obtained by considering the intersection of the terminal stability of K-muscovite + quartz (a reaction that has not been observed in this area; Evans and Guidotti, 1966) with Holdaway's curve for the andalusite–sillimanite transition. Using the data of Chatterjee and Johannes (1974), this intersection occurs at approx 3.0 kb.

TABLE 4

Summary of analytical data as a function of metamorphic grade and AKNa–Ca mineral assemblage

		Musc		Plag			$T^{\circ}C$*		
Assemblage	No. of Samples	X_{Pg}	$[X_{(Al,M2)}]^2$	X_{An}	X_{Ab}	X_{Pg}/X_{Ab}	A	B	C
LSZ:									
Ia	25	.217	(.889)	.155	.840	.258	544	547	558
Ib	24	.218	(.889)	.138	.857	.254	546	549	560
Avg	49	.217	.889	.146	.849	.256	545	548	559
USZ:									
IIa	14	.194	.878	.142	.854	.227	550	552	565
IIb	33	.148	.847	.175	.820	.180	560	561	577
Avg	47	.162	.856	.165	.830	.195	556	557	572
SKZ:									
III	13	.103	.847	.161	.840	.123	586	585	603
IV	5	.069	(.821)	.130	.860	.080	623	620	643
E&G(IV)**	11	.066	.821	.197	.792	.083	619	615	638

* Temperatures calculated from equation (14) at 3500 bars as follows:

A. Using Margules parameters of Eugster and others (1972) and ignoring M2 site correction.

B. Using Margules parameters of Chatterjee and Froese (1975) and ignoring M2 site correction.

C. Using Margules parameters of Chatterjee and Froese (1975) and M2 site correction. Note that the LSZ average ($X_{Al,M2}$) was used for both assemblage IA and IB; the average assemblage I b correction was applied to assemblage III, and the average correction from Evans and Guidotti (1966) was used for assemblage IV.

** Data from Evans and Guidotti (1966).

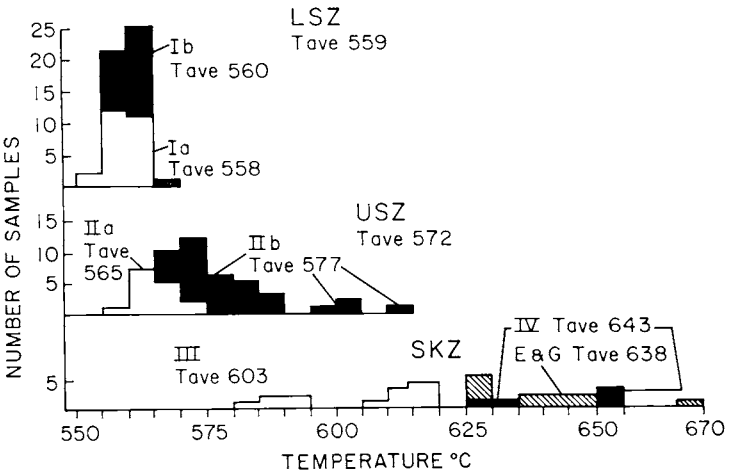

Fig. 5. Distribution of temperatures derived from eq (14) by method "C" (see table 4) as a function of mineral assemblage and metamorphic grade.

These pressures agree well with other pressure estimates for Acadian metamorphism in Northwest Maine. Particularly germane are the estimates of 3 to 4 kb by Burress (ms), assuming temperatures of 635° to 685°C for SKZ specimens from the Bryant Pond Quadrangle. Thus an *assumed* pressure of 3500 bars during metamorphism seems reasonable.

Results.—Temperatures derived from eq (14) are summarized for our average composition data by metamorphic grade and mineral assemblage in table 4. The temperatures derived from each specimen within a specific assemblage are listed in table 2 and summarized on figures 5 and 6.[3] The uncertainty in these temperatures is based solely on the plus or minus one mole percent error assumed for the probe analyses and varies from ± 3°C for assemblage Ia specimens to ± 15°C for assemblage IV specimens.

From table 4 and figure 5, the LSZ-USZ isograd should occur at approx 560°C, some 40° to 50°C lower than generally accepted for a pressure of 3500 bars (compare Guidotti, 1974). The lowest temperature group of assemblage III specimens (that is, those that presumably have equlibrated at a higher f_{H_2O}) average about 590°C. Hence, this value can be used in a relative sense as the temperature of the sillimanite + K-feldspar isograd.

Because the traces of the isograds are relatively parallel and dip northward at 10 to 15 degrees (Cheney, ms; Evans and Guidotti, 1966), the elevation difference between the isograds of approx 0.5 km can be used in conjunction with the relative temperature difference of approx 30°C to estimate a minimum thermal gradient of approx 60° ± 18°C per km during the final metamorphism in the Puzzle Mountain area. Using an average temperature of approx 600°C for all assemblage III

[3] The temperatures used throughout are those derived by method "C" of table 4.

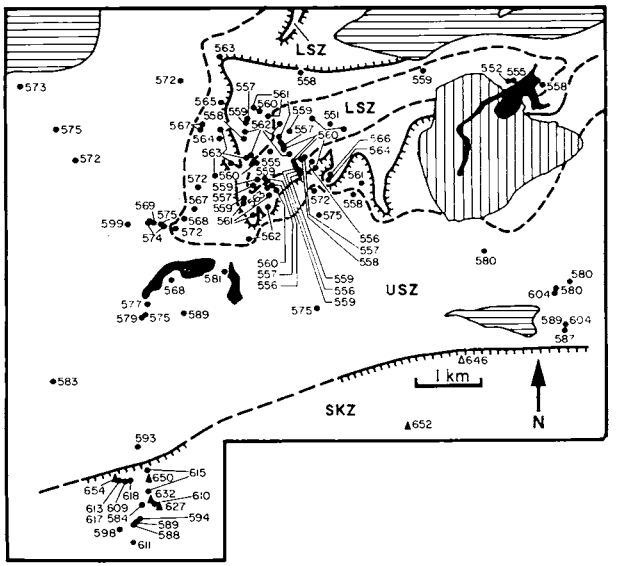

Fig. 6. Areal distribution of temperatures derived from eq (14) by method "C" (see table 4). Legend as on figure 1.

specimens results in an estimated thermal gradient of 80° ± 18°C per km. Although these thermal gradients seem high, they are consistent with the thermal nature of the metamorphism. Moreover, Burruss (ms), on the basis of "H_2O bearing inclusions on healed fractures in three Bryant Pond samples," has suggested that a thermal gradient of approx 70°C per km existed during the "retrograde evolution" of these particular samples.

Eq (14) can also be used to determine, in a qualitative manner, the difference in the equilibrium f_{H_2O} between assemblage III and IV specimens. Using the average assemblage III temperature of 600°C and solving eq (14) for f_{H_2O} but using the average assemblage IV compositions of muscovite and plagioclase (Puzzle Mountain data only) results in an approx 30 percent lower f_{H_2O} during the equilibration of the K-feldspar-bearing specimens relative to assemblage III specimens. This calculation quantifies the interpretation of Evans and Guidotti (1966) that, as previously discussed, the occurrence of intermixed assemblage III and assemblage IV specimens is most likely a function of locally variable, internally controlled f_{H_2O}.

SUMMARY AND CONCLUSIONS

The mole fraction quotient $X_{Pg}^{musc}/X_{Ab}^{plag}$ relating the actual Na components of muscovite and plagioclase can be considered as a P–T–f_{H_2O} dependent Kd. Analysis of this distribution coefficient has provided the functional dependence of muscovite A-site composition upon plagioclase composition for virtually the entire range in temperature over

which muscovite and sillimanite coexist. This compositional dependence is as follows:

1. Within the lower grade portion of the LSZ

$$\Delta(X_{Pg}^{musc})/\Delta(X_{Ab}^{plag}) = 0.3 \text{ percent Pg}/1.0 \text{ percent Ab} = 0.3$$

2. Within the SKZ, considering only K-feldspar-bearing specimens,

$$\Delta(X_{Pg}^{musc})/\Delta(X_{Ab}^{plag}) = 0.1 \text{ percent Pg}/1.0 \text{ percent Ab} = 0.1$$

Analysis of the $Kd_{Pg\text{-}Ab}^{musc\text{-}plag}$ can also be used, in a general fashion, to evaluate qualitatively equilibrium f_{H_2O} variations among specimens from the same metamorphic grade and T–f_{H_2O} variations from grade to grade. However, for a single sample or a small group of specimens, it is generally not possible to differentiate between intensive parameter variations and disequilibrium. Such differentiation should be considered in the context of an additional relation providing information on temperature variation among specimens, for example, qualitative temperatures derived from the Mg/Fe partitioning between garnet and biotite (Thompson, 1976; Goldman and Albee, 1977; Holdaway and Lee, 1977).

Relative temperatures derived by quantitative evaluation of eq (14) can be used to place limits on thermal gradients and f_{H_2O} variation among approximately isothermal specimens. In this context, we have determined that a thermal gradient of 60° to 80°C/km existed during the final metamorphism in the Puzzle Mountain area of Northwest Maine. This is consistent with the thermal nature of the metamorphism. Moreover, our calculations have quantified the observation of Evans and Guidotti (1966) that the mixture of K-feldspar and non-K-feldspar-bearing specimens characterizing *this* SKZ is due to an approx 30 percent lower f_{H_2O} of equilibration in the K-feldspar-bearing rocks.

The results summarized here indicate that the compositions of coexisting plagioclase and muscovite in sillimanite-bearing metapelites are a viable indicator of intensive parameter variation within a single petrologically well characterized metamorphic terrain.

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