

Lithium aluminosilicate occurrences in pegmatites and the lithium aluminosilicate phase diagram

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Abstract

The minerals eucryptite (LiAlSiO_4), spodumene ($\text{LiAlSi}_2\text{O}_6$), and petalite ($\text{LiAlSi}_4\text{O}_{10}$) are important constituents of lithium pegmatites, yet the stability relations of these minerals are poorly understood. Natural occurrences and available experimental data have been utilized to construct a P - T diagram for phases in the binary system LiAlSiO_4 - SiO_2 . The calculated lithium aluminosilicate stability relations in the degenerate $(n+3)$ -phase multisystem eucryptite, spodumene, β -spodumene (β - $\text{LiAlSi}_2\text{O}_6$), petalite, and quartz imply that eucryptite + quartz is stable at low P and T , spodumene is stable at high P , spodumene + petalite is stable at moderate P and T , petalite is stable at moderate T and low P , and β -spodumene is stable only at high T . The proposed lithium aluminosilicate phase diagram accounts for the stable occurrence of eucryptite + quartz at low P and T , and for the observed replacement of petalite by spodumene + quartz (at relatively high P and decreasing T) or by eucryptite + quartz (at relatively low P and decreasing T). The phase β -spodumene is probably stable only well above the lithium pegmatite solidus, thus accounting for the absence of β -spodumene in nature.

Introduction

The three lithium aluminosilicates, eucryptite (LiAlSiO_4), spodumene ($\text{LiAlSi}_2\text{O}_6$), and petalite ($\text{LiAlSi}_4\text{O}_{10}$) have long been of interest to mineralogists, igneous petrologists, and economic geologists (*e.g.*, Julien, 1879; Brush and Dana, 1880) (Table 1). Much of what is known about the geology of these minerals stems from field studies of pegmatites conducted during World War II, when the wartime demand for mica and beryl was high. The results of these and subsequent investigations are summarized by Cameron *et al.* (1949, 1954), Page *et al.* (1953), Jahns (1955), Černý (1975), and Heinrich (1975). In addition, studies of pegmatites in the Soviet Union have provided a large body of information pertaining to the geology of lithium pegmatites and occurrences of the lithium aluminosilicates; much of this work is summarized by Fersman (1931), Ginzburg (1955), Beus (1960), Vlasov (1961, 1964-66), Gordienko (1970), and Ovchinnikov and Kuz'menko (1976).

Of the three lithium aluminosilicates, spodumene

is by far the most widespread and abundant. Petalite is second in abundance and distribution, and it constitutes the principal lithium aluminosilicate in several pegmatite districts. Some of the important localities and modes of occurrence of spodumene and petalite are summarized by Heinrich (1975). Eucryptite is the rarest and least abundant of these three minerals; however, it has now been reported from at least twenty localities (Table 2). The rarity of eucryptite may be due in part to the fact that it is difficult to identify (its name stems from the Greek *eu kryptos*, meaning well-concealed). It commonly occurs in extremely fine-grained intergrowths with albite or quartz, and its optical and physical properties are virtually identical to those of quartz (*e.g.*, Černý, 1972).

Pegmatitic deposits of spodumene and petalite are mined throughout the world for use in glass ceramics and in the manufacture of numerous lithium-based compounds (Heinrich, 1975; Vine, 1976; Singleton, 1979). The only known economic deposit of eucryptite is in pegmatites of Bikita, Zimbabwe (Cooper, 1964). In addition to lithium ores, ores of beryllium, niobium, tantalum, tin, cesium, rubidium, bismuth, and other rare elements commonly

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Table 1. Names and compositions of phases discussed in this paper

Phase	Abbreviation used	Composition
eucriptite	Ecr	$\alpha\text{-LiAlSiO}_4$
spodumene	Spd	$\alpha\text{-LiAlSi}_2\text{O}_6$
petalite	Pet	$\text{LiAlSi}_4\text{O}_{10}$
quartz	Qtz	SiO_2
β -eucriptite	Bec	$\beta\text{-LiAlSiO}_4$
β -spodumene	Bsp	$\beta\text{-LiAlSi}_2\text{O}_6$

are concentrated in the inner, lithium aluminosilicate-bearing zones of pegmatites (Vlasov, 1961); the richest of these ores typically occur in albite or lepidolite masses that appear to have formed at the expense of spodumene-bearing pegmatite (Beus, 1960; Vlasov, 1961). Therefore, the processes responsible for the formation and destruction of lithium aluminosilicates also may be responsible for the dramatic concentration of rare elements in these zones. Studies of lithium aluminosilicate stabilities may yield information pertinent to rare-metal mineralization and thus may aid future geochemical exploration for these elements.

The purpose of this study is to define the conditions of formation and alteration of the lithium aluminosilicates in terms of pressure, temperature, cooling history, and alteration by late fluids. The melting and crystallization equilibria of systems containing lithium aluminosilicates have been studied extensively at low pressures; phase relations near the solidus in the system $\text{NaAlSi}_3\text{O}_8\text{-LiAlSiO}_4\text{-SiO}_2\text{-H}_2\text{O}$ at 2 kbar $P_{\text{H}_2\text{O}}$ are discussed by Stewart (1978). In contrast, the subsolidus stabilities of the lithium aluminosilicates have received relatively little attention, even though lithium aluminosilicates, especially spodumene, commonly display a variety of replacement textures and assemblages that result from late-stage pegmatitic processes (e.g., Brush and Dana, 1880; Graham, 1975; Jahns and Ewing, 1976; Chakoumakos, 1978). This paper will focus on the subsolidus stabilities of the three lithium aluminosilicates named above. Virgilite, $\text{Li}_x\text{Al}_x\text{Si}_{3-x}\text{O}_6$ (French, Jazek, and Appleman, 1978), and bikitaite, $\text{LiAlSi}_2\text{O}_6 \cdot \text{H}_2\text{O}$ (Hurlbut, 1958; Phinney and Stewart, 1961; Drysdale, 1971) will not be considered here. This paper presents a qualitative P - T phase diagram for the portion of the lithium aluminosilicate system $\text{LiAlSiO}_4\text{-SiO}_2$ (Table 1) that is relevant to geologic

conditions. In a following paper (London and Burt, 1982b) we will consider the effects of added components in the melt or coexisting fluid (as opposed to solid solution components) on lithium aluminosilicate stabilities. The preliminary results of experimental phase equilibrium studies in the system $\text{LiAlSiO}_4\text{-SiO}_2\text{-H}_2\text{O}$ are available in London (1981).

Nomenclature

The numerous pegmatite classification schemes (e.g., Cameron *et al.*, 1949; Jahns, 1955; Brötzen, 1959; Beus, 1960; Vlasov, 1961; Uebel, 1977) have established a fair degree of uniformity in the terminology applied to the types and internal features of pegmatite bodies, yet there are some significant differences among these models in their proposed crystallization sequences and zonal assemblages. These discrepancies stem largely from a lack of uniformity in the criteria used to distinguish the primary or secondary (subsolidus or replacement) nature of pegmatite minerals. Pegmatite textures

Table 2. Reported occurrences of eucriptite

Locality	References	Association
Boulder County, Colorado USA	USNM #128648*	fine-grained aggregate; associated phases unknown
Branchville, Connecticut, USA	Brush and Dana, 1880	Ecr+Alb after Spd
Center Strafford, New Hampshire, USA	Mrose, 1953	Ecr+Alb
Etta mine, South Dakota, USA	Schwartz & Leonard, 1926	unconfirmed
Harding mine, New Mexico, USA	Jones, 1964 Mrose, 1953	massive Ecr; Ecr+Qtz after Spd
Hiddenite, North Carolina, USA	USNM #10014	tentative; Ecr-Spd-Qtz fracture filling
Kings Mt., North Carolina, USA	Leavens <i>et al.</i> , 1968	Ecr+Qtz fracture filling
Pala dist., California, USA	Jahns, 1979	association not reported
White Picacho dist. Arizona, USA	Burt <i>et al.</i> , 1977	Ecr+Alb after Spd
Nakina, Ontario, Canada	Mandarino & Harris, 1968	Ecr-Spd
Tanco mine, Manitoba Canada	Cerný, 1972, 1975	Ecr+Anl**; massive Ecr; Ecr+Qtz after Spd
Bikita, Zimbabwe	Hurlbut, 1957, 1958, 1962	Ecr-Qtz-Bik** after Pet; Ecr+Qtz after Pet; massive Ecr
Gatumba, Rwanda	von Knorring, 1966	Ecr+Alb after Spd
Mollefontein, S. Africa	Schröder & Hoffman, 1957	association not reported
Salisbury dist., Zimbabwe	Grubb, 1973	Ecr-Spd-Pet-Qtz
Central Kazakstan, USSR	Ovchinnikov & Kuz'menko, 1976	association not reported
Kalbinsk region, USSR	Ginzburg, 1959	Ecr+Alb after Spd
Kola Peninsula, USSR	Sosedko & Gordienko, 1957	Ecr+Alb after Spd
Siberia, USSR	Lugovskii <i>et al.</i> , 1977	Ecr-Spd-Pet-Qtz
Soviet Central Asia	Rossovskii, 1971	Ecr+Qtz after Spd and/or Pet
Nanling Ranges, S. China	Chao, 1964	with Lib, Lpt** <i>etc.</i>
Londonderry, West Australia	USNM #146420	Ecr+Qtz after Pet

* USNM = U.S. National Museum, Smithsonian Institution, Washington, D.C., USA

** Anl = analcime, Bik = bikitaite, Lib = liberite, Lpt = lepidolite

are extremely complex and varied, and recognition of the paragenetic sequence consequently is ambiguous and subjective.

In this and the following paper (London and Burt, 1982b) inferred equilibrium assemblages are indicated by mineral names separated by plus signs (+); mineral associations that probably do not or may not constitute a single paragenesis are represented by mineral names separated by hyphens (-). The term *primary* is applied to a mineral or an assemblage that generally appears to have crystallized directly from a melt (with or without the participation of an aqueous fluid phase). The granitic border zones and most of the extremely coarse-grained pegmatitic units are interpreted as primary in origin. In general, minerals are regarded as *secondary* or *replacement assemblages* only if recognizable evidence of replacement (*e.g.*, pseudomorphism, crystallographically continuous relicts of a host phase, embayment, *etc.*) is present.

Natural occurrences

Spodumene and petalite usually form extremely large primary crystals in the interior portions of zoned lithium pegmatites. In such occurrences, both minerals typically are embedded in masses of quartz or of quartz plus other minerals (*e.g.*, albite, microcline, amblygonite-montebrazite). In contrast, eucryptite seldom, if ever, forms as a primary phase. Relatively coarse (up to 2 cm in dimension) subhedral crystals of eucryptite described by Hurlbut (1962) from Bikita, Zimbabwe, may be primary, although they do not approach the giant dimensions of the other two lithium aluminosilicates.

Primary spodumene and petalite frequently exhibit partial to complete pseudomorphism by a variety of fine-grained mineral assemblages. Although alteration may be pervasive, it usually is sporadic within individual pegmatites and even within single host crystals. In most cases, this replacement along cleavage surfaces and crystal boundaries of the host lithium aluminosilicates can be clearly attributed to pegmatitic processes and not to subsequent regional metamorphism or local hydrothermal alteration (*e.g.*, London and Burt, 1982a). Alteration of primary spodumene or petalite may be essentially isochemical or may involve cation exchange. This paper considers only isochemical replacement processes; the following paper (London and Burt, 1982b) treats lithium aluminosilicate alterations involving cation exchange.

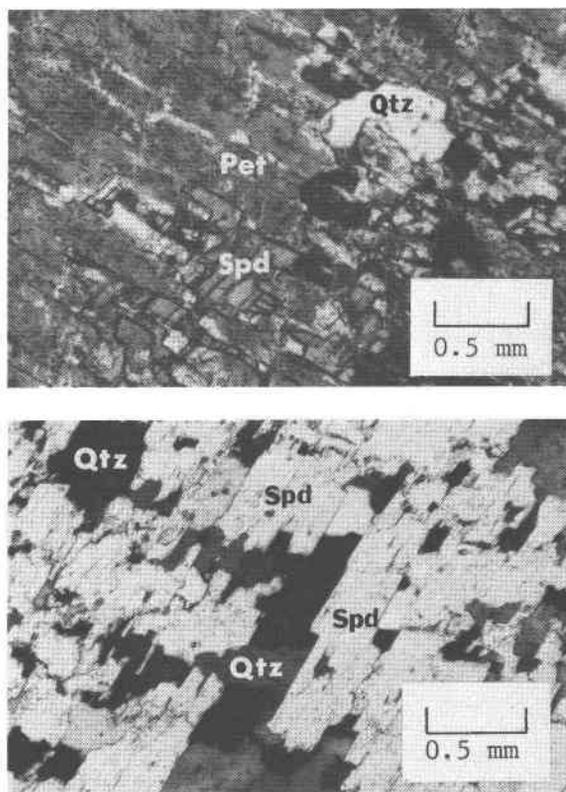


Fig. 1. Photomicrographs showing (top) the alteration of petalite to spodumene + quartz from Bikita, Zimbabwe (specimen courtesy of C. S. Hurlbut, Jr., and Carl A. Francis, Harvard University), and (bottom) spodumene + quartz replacement of petalite from the Tanco pegmatite, Bernic Lake, Manitoba, Canada. Crossed polars.

Isochemical replacement

The most common isochemical replacement of lithium aluminosilicates involves the breakdown of petalite to a symplectic or fibrous intergrowth of spodumene + quartz (Fig. 1). Černý and Ferguson (1972) and Černý (1975) have reviewed the eight localities from which this isochemical replacement of petalite has been reported. In addition to the textural similarities of the symplectic or fibrous intergrowths, descriptions from several of these localities indicate that the long or *c* axes of spodumene grains or fibers are oriented parallel to the {001} cleavage in the host petalite. In samples from the Tanco pegmatite in Manitoba, Černý and Ferguson (1972) observed (optically) a crystallographic orientation of the secondary spodumene with respect to petalite, such that c_{spd} is parallel to b_{pet} .

Eucryptite, which is chemically and structurally similar to the feldspathoids nepheline and kalsilite,

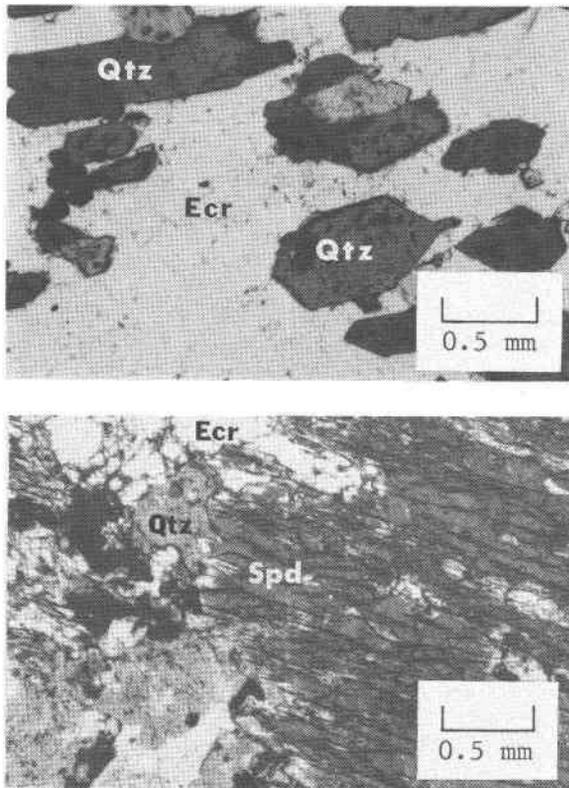


Fig. 2. Photomicrographs showing (top) the intergrowth of eucryptite + quartz after petalite from Bikita, Zimbabwe (specimen courtesy of C. S. Hurlbut, Jr., and Carl A. Francis, Harvard University), and (bottom) eucryptite + quartz replacement of spodumene from the Harding pegmatite, Taos County, New Mexico. Crossed polars.

surprisingly occurs intergrown with quartz after petalite (Fig. 2) from Bikita, Zimbabwe (Hurlbut, 1962) and from pegmatites in the Central Soviet Union (Rossovskii, 1970). Estimates by Rossovskii, and our own observations from thin sections of Bikita samples, indicate that the intergrowths are approximately 60–80 vol% eucryptite and 40–20 vol% quartz. Černý (1975) has described a eucryptite + quartz intergrowth that appears to have replaced spodumene at the Tanco pegmatite, and we have observed eucryptite + quartz intergrowths after spodumene from the Harding pegmatite, Taos County, New Mexico (Fig. 3). In addition, well-formed crystals of eucryptite and quartz occur as fracture fillings in pegmatite from Kings Mountain, North Carolina (*e.g.*, Smithsonian specimen USNM 119971) and from Hiddenite, North Carolina (*e.g.*, tentative identification of Smithsonian specimen USNM 10014). These natural occurrences led Leavens *et al.* (1968) and Burt *et al.* (1977) to

propose that eucryptite + quartz is a stable assemblage under some geologic conditions.

Fluid inclusion studies

The available data on the homogenization temperatures (*i.e.*, minimum filling temperatures) of multiphase fluid inclusions suggest that spodumene or spodumene-bearing pegmatite crystallizes in the range of 300°–650°C (Sheshulin, 1963; Bazarov and Motorina, 1969; Brookins *et al.*, 1979; Taylor *et al.*, 1979). Sheshulin (1963) identified two generations of spodumene in pegmatites from the Soviet Union; homogenization temperatures were 500°–650°C for first-generation spodumene, and 290°–390°C for second-generation spodumene. Estimates of filling pressure for saline or CO₂-rich inclusions indicate pegmatite crystallization pressures in the range of 1.5–2.5 kbar (Bazarov and Motorina, 1969; Brookins *et al.*, 1979; Taylor *et al.*, 1979). The results of these studies imply that spodumene (\pm quartz) is

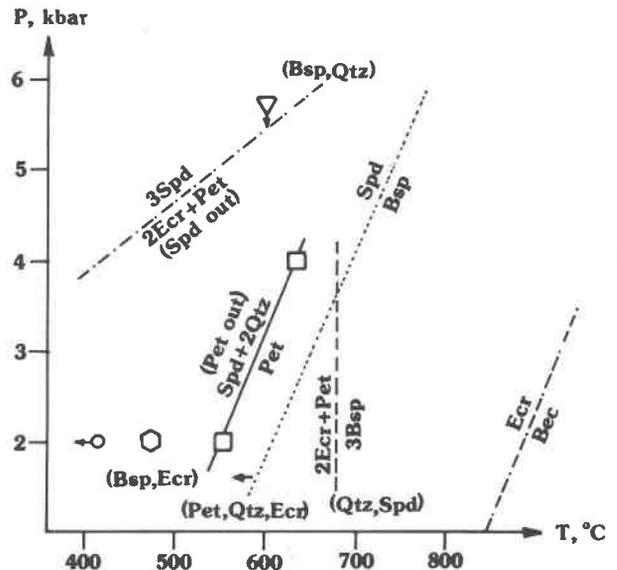


Fig. 3. Summary of the principal experimental work on the lithium aluminosilicate system under geologic conditions (several kilobars of pressure). Dot-dash line and triangle: spodumene synthesis experiments of Shternberg, *et al.* (1973); dashed line: stability limit of petalite reported by Stewart (1963); dotted line: spodumene- β -spodumene inversion determined by Munoz (1969); dash-dash-dot line: the α -eucryptite- β -eucryptite inversion reported by Isaacs and Roy (1958); solid line and squares: reversal of reaction (Bsp,Ecr) reported by Stewart (1963); hexagon: reversal of reaction (Bsp,Ecr) reported by Munoz (1971); circle: spodumene + quartz \rightarrow petalite, unreversed reaction (D. J. Drysdale, unpublished data, personal communication, 1980). Equilibrium boundaries for the reactions probably lie in the directions indicated by arrows.

Table 3. Cell dimensions and calculated molar volumes for eucryptite and β -eucryptite

Source	a_0 , Å	c_0 , Å	v^0 , joules/bar
<u>eucryptite</u>			
Bikita, Zimbabwe (Hurlbut, 1962)	13.480(5)	9.010(5)	4.744
Kings Mountain, North Carolina (Leavens <i>et al.</i> , 1968)	13.490(7)	9.016(7)	4.755
Tanco mine, Bernic Lake, Manitoba (Černý, 1972)	13.472(1)	9.000(1)	4.733
synthetic (Tien and Hummel, 1964)	13.51	9.07	4.797
synthetic (Winkler, 1948)	13.53	9.04	4.796
synthetic (Rieder, 1971)	13.520(4)	9.035(4)	4.786
synthetic (Stewart, 1960)	13.476(3)	9.003(1)	4.738
<u>β-eucryptite</u>			
Buerger (1948)	10.55	11.22	5.428
Winkler (1948)	5.27 ($\frac{1}{2}a_0$)	11.25	5.432
Tien and Hummel (1964)	5.249 ($\frac{1}{2}a_0$)	11.186	5.359
Pillars and Peacor (1973)	10.497(3)	11.200(5)	5.364

stable to pressures at least as low as 2 kbar at low to moderate temperatures.

Calorimetric and experimental data

Construction of the lithium aluminosilicate phase diagram in P - T space is hampered by a lack of relevant thermochemical data and experimental work in the system LiAlO_2 - SiO_2 . The stability relations among the lithium aluminosilicates in quartz-saturated and undersaturated systems have received considerable attention from ceramists (*e.g.*, Hatch, 1943; Roy *et al.*, 1950, Tien and Hummel, 1964) because of the low thermal expansion and fluxing properties of lithium aluminosilicate glasses and ceramics. Most of this work was conducted at atmospheric pressure. The principal investigations under geologic conditions (several kilobars of pressure) are those of Isaacs and Roy (1958), Skinner and Evans (1960), Stewart (1960, 1963, 1978), Phinney and Stewart (1961), Edgar (1968), Munoz (1969, 1971), Drysdale (1971, 1975), Grubb (1973), and Shternberg *et al.* (1973). The thermochemical properties of spodumene, β -spodumene, and β -eucryptite have been determined from solution and adiabatic calorimetry by Barany and Adami (1966) and Pankratz and Weller (1967); the thermochemical properties of petalite have been determined calorimetrically by Bennington *et al.* (1980) and by Hemingway (unpublished data, 1980). Although many aspects of lithium aluminosilicate stabilities have been elucidated, these experimental studies have not provided a coherent and geologically relevant lithium aluminosilicate phase diagram

in P - T space. There are significant discrepancies and apparent conflicts in much of the experimental work (discussed below). Also, none of the previous studies has accounted for the stable association of eucryptite + quartz, even though this assemblage has been reported from several localities (Table 2).

Considerable confusion surrounds the thermochemical properties and structural state of eucryptite. There are two principal forms of LiAlSiO_4 . One is the natural, low-temperature form known as eucryptite, low-eucryptite, or α -eucryptite. Its dimorph is a synthetic, high-temperature form referred to as high-eucryptite or β -eucryptite. The crystal structures of eucryptite and of β -eucryptite are analogous to those of α -quartz and β -quartz, respectively, (*i.e.*, they are stuffed derivatives of the silica minerals: Buerger, 1954). Eucryptite possesses space group symmetry $R3$ (Mrose, 1953; Hurlbut, 1962) whereas that of β -eucryptite is $P6_422$ (Winkler, 1948). The nature of the polymorphic transition, which has not been studied in detail, appears to be different from that of α - and β -quartz. It may be a first-order reconstructive transformation, inasmuch as β -eucryptite is metastably preserved at room temperature (Pillars and Peacor, 1973). In addition, the molar volumes of eucryptite and β -eucryptite are quite different (Table 3). The data presented in Table 3 serve to clearly identify the LiAlSiO_4 polymorphs studied by other workers.

The thermochemical data reported for "eucryptite" by Robie *et al.* (1978) are derived from the calorimetric work of Barany and Adami (1966) and of Pankratz and Weller (1967) and thus belong to β -

Table 4. Molar volume and thermochemical data on phases in the system $\text{LiAlSiO}_4\text{-SiO}_2^*$

Phases	V^0 , joules/bar	S^0 , joules/mole-K	$\Delta H_{f,298}^0$, kjoules/mole
Ecr	4.797(5)**	90(2)***	-2138(3)***
Bec	5.363(5)	103.80(80)	-2123.300(1.980)
Spd	5.837(2)	129.30(80)	-3053.500(2.790)
Bsp	7.825(4)	154.40(1.20)	-3025.300(2.790)
Pet	12.840(5)****	270(1) [†] 232.200(92) ^{††} 232.9(1) ^{†††}	-4832(2) [†] -4886.5(12.5) ^{††}
Qtz	2.2688(1)	41.46(20)	-910.700(1.000)

*Data from Robie et al. (1978), except as follows:
 **from Tien and Hummel (1964)
 ***calculated as explained in the text
 ****calculated from X-ray data (JCPDS file card 14-90)
 †calculated from the work of Stewart (1963) as explained in the text
 ††from Bennington et al. (1980)
 †††unpublished data from B. S. Hemingway (personal communication, 1980)

eucriptite. Nevertheless, the thermochemical properties of eucriptite can be estimated by utilizing the work of Isaacs and Roy (1958). They have bracketed the reaction $\alpha\text{-LiAlSiO}_4 = \beta\text{-LiAlSiO}_4$ and report a transformation reaction slope of 23.5 bars/K. Using the molar volume data of Tien and Hummel (1964) and assuming a constant $\Delta V_{\alpha-\beta} = 0.56$ joules/bar (Table 4), then the Clausius-Clapeyron equation, $dP/dT = \Delta S/\Delta V$, may be solved for ΔS , and $\Delta S_{\alpha-\beta} = 13.19$ joules/mole-K. Subtraction of this quantity from $S^0_{298.15}$ for β -eucriptite (Robie et al., 1978) yields an estimate for $S^0_{298.15}$ of eucriptite (Table 4). From the experimentally determined reaction line for the eucriptite- β -eucriptite transition, Isaacs and Roy (1958) determined that $\Delta H_{\alpha-\beta} = 15.7 \pm 3$ kJ/mole; subtraction of this quantity from $\Delta H_{f,298.15}^0$ for β -eucriptite (Robie et al., 1978) provides an estimate for $\Delta H_{f,298.15}^0$ for eucriptite (Table 4).

Values for $S^0_{298.15}$ and $\Delta H_{f,298.15}^0$ of petalite can be derived from the experimental work of Stewart (1963) on the eucriptite-absent (Bsp,Ecr) reaction: $\text{Spd} + 2\text{Qtz} = \text{Pet}$. Stewart (1963) gives two reaction reversals that define a line with slope 23.5 bars/K. Using this information and data from Table 4, $S^0_{298.15}$ and $\Delta H_{f,298.15}^0$ of petalite have been calculated from the above Clausius-Clapeyron equation and from the equation, $\Delta G = \Delta H - T\Delta S + (P-1)\Delta V$, assuming that ΔV and ΔS are constant over a wide range of P and T . Other workers have presented experimental results involving reaction (Bsp,Ecr) that differ significantly from those of Stewart (1963). Stewart reported a reversal of reaction (Bsp,Ecr) at 550°C and 2 kbar $P_{\text{H}_2\text{O}}$, whereas Mu-

noz (1971) gave an equilibrium temperature of 470°C at the same pressure, and D. J. Drysdale (unpublished data; personal communication, 1980) observed that petalite is stable down to temperatures of 440°C or lower for this same reaction at 2 kbar pressure (Fig. 3). While this discrepancy may not change the fundamental relationship of quartz-spodumene-petalite equilibria, it would have a drastic effect on the thermochemical properties of petalite as derived from Stewart's work. Recent calorimetric determinations for $S^0_{298.15}$ of petalite by Bennington et al. (1980) and by B. S. Hemingway (unpublished data, personal communication, 1980) are in close agreement (Table 4), and both differ substantially from the value derived from the work of Stewart (1963). The value for $S^0_{298.15}$ of petalite reported by Bennington et al. (1980) has been used here in reaction slope calculations involving petalite. In addition, the work of Stewart (1963) and Munoz (1971) on reaction (Bsp, Ecr) appears to be in conflict with the results of synthesis experiments (reported as an unreversed reaction boundary) by Shternberg et al. (1973) involving reaction (Bsp,Qtz) (Fig. 3). This conflict underscores the need for further experimental work in the lithium aluminosilicate system.

The lithium aluminosilicate phase diagram

The lithium aluminosilicate phase diagram (Fig. 4) involves the (n+3)-phase multisystem eucriptite, spodumene, β -spodumene, petalite, and quartz (Table 5). Figure 4 is drawn for temperatures sufficiently low that β -eucriptite is unstable (*cf.* Isaacs and Roy, 1958). In addition, β -spodumene is assumed to be stoichiometric, although high-temperature β -spodumene- β -quartz solid solutions have been reported by numerous experimentalists (*e.g.*, Hatch, 1943; Roy, et al., 1950; Skinner and Evans, 1960; Munoz, 1969, 1971; Stewart, 1978). The assumption of stoichiometry in β -spodumene permits the use of calorimetric data compiled by Robie et al. (1978). This degenerate multisystem contains five possible stable or metastable binary invariant points in P - T space. These and one topology of all possible univariant reactions in this system are presented in Figure 4. The topology shown implies that the assemblage spodumene + petalite is stable (*i.e.*, invariant points [Spd] and [Pet] are metastable). An alternative configuration (not shown) would interchange stable and metastable invariant points, and by "the stable-metastable correspondence" (Burt, 1978), would imply that the assemblage quartz +

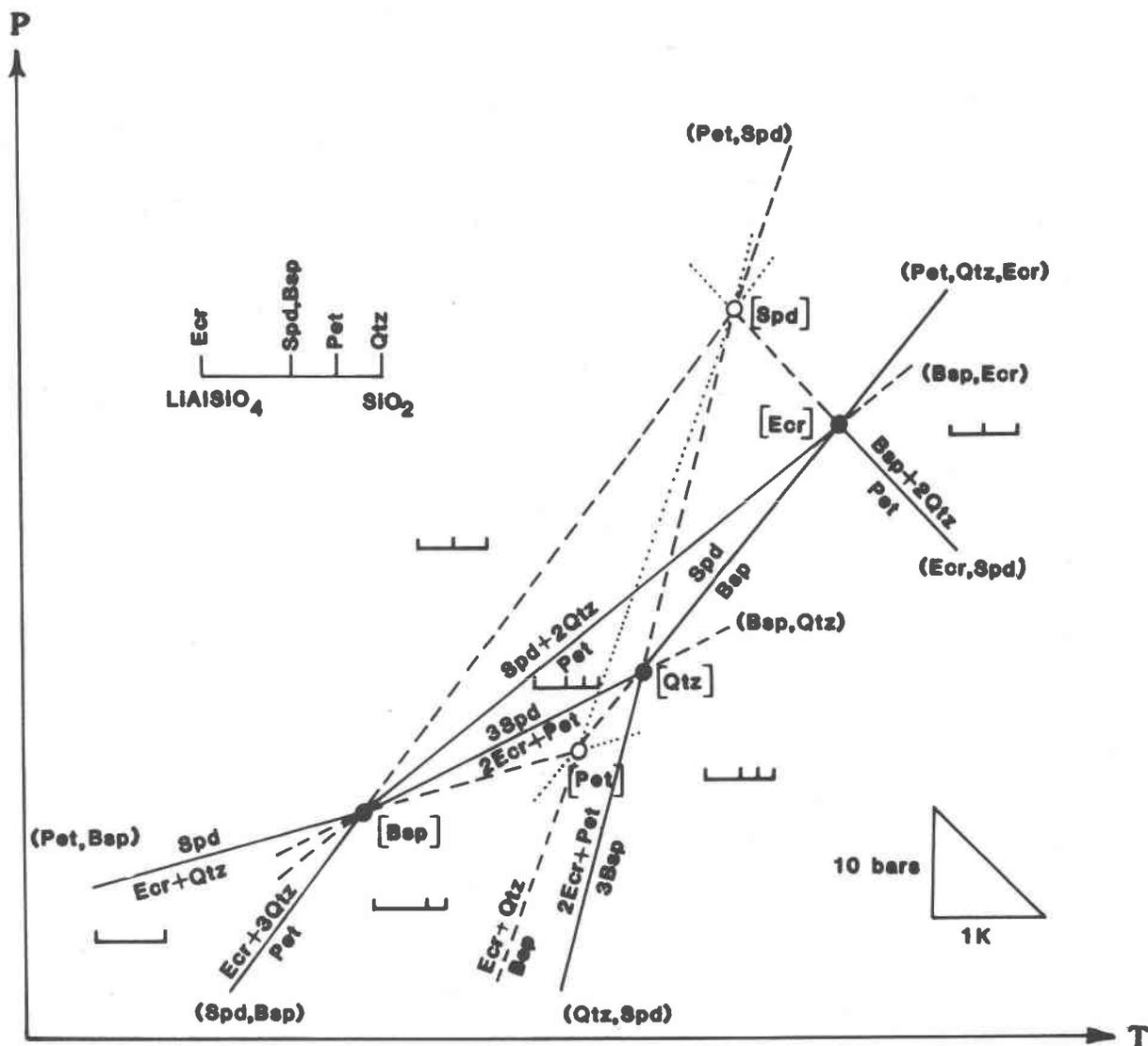


Fig. 4. P - T phase diagram illustrating the stable topology of univariant reactions in the $(n+3)$ -phase multisystem eucryptite, spodumene, β -spodumene, petalite, and quartz in the system LiAlSiO_4 - SiO_2 . Solid lines: stable reactions; dashed and dotted lines: metastable reactions; solid circles: stable invariant points; open circles: metastable invariant points. The triangular scale marker in this and the following diagram indicates the relative slopes of reaction lines. Positions of phases on the binary LiAlSiO_4 - SiO_2 are indicated in the upper left portion of this diagram. See Table 1 for abbreviations of phase names.

eucryptite + β -spodumene (not known in nature) is stable. The configuration shown in Figure 4 is therefore assumed to be the stable one.

The univariant reactions around point [Bsp] involve only the naturally occurring phases, and thus the topology of reactions about this point is particularly relevant to geologic environments (Fig. 4). The configuration of phases on the binary join LiAlSiO_4 - SiO_2 (Fig. 4) implies four possible univariant reactions at invariant point [Bsp]. Reaction slope calculations based on the data of Table 4 indicate that

these four reactions have positive slopes in P - T space. The uncertainties in the thermochemical properties of eucryptite and the discrepancies reported for the 2 kbar reversal of reaction (Bsp,Ecr) make the precise calculation of the location of point [Bsp] a meaningless effort. Our feeling, based on the natural occurrences discussed above, is that the point [Bsp] occurs at considerably lower P and T than does the aluminosilicate triple point (Holdaway, 1971). Natural occurrences and the limited experimental work then suggest that the

Table 5. Univariant reactions in the system $\text{LiAlSi}_4\text{-SiO}_2$ (Fig. 4)

Phases absent	Reactions	ΔV_{298}^0	ΔS_{298}^0	dP/dT bars K
(Pet,Bsp)	$\text{Ecr} + \text{Qtz} = \text{Spd}$	-1.23	-2.16	+1.8
(Spd,Bsp)	$\text{Ecr} + 3\text{Qtz} = \text{Pet}$	+1.23	+17.82	+14.4
(Bsp,Ecr)	$\text{Spd} + 2\text{Qtz} = \text{Pet}$	+2.46	+19.98	+8.1
(Bsp,Qtz)	$2\text{Ecr} + \text{Pet} = 3\text{Spd}$	-4.92	-24.30	+4.9
(Pet,Spd)	$\text{Ecr} + \text{Qtz} = \text{Bsp}$	+0.76	+22.94	+30.2
(Pet,Qtz,Ecr)	$\text{Spd} = \text{Bsp}$	+1.99	+25.10	+12.6
(Qtz,Spd)	$2\text{Ecr} + \text{Pet} = 3\text{Bsp}$	+1.05	+51.00	+49.0
(Ecr,Spd)	$\text{Bsp} + 2\text{Qtz} = \text{Pet}$	+0.48	-5.12	-10.7

four univariant lines define a spodumene field at relatively high P and moderate T , a petalite field at high T and low to moderate P , a eucryptite field at low to moderate P and T , and a petalite + spodumene field over a narrow range of moderate to high P and T . Inasmuch as the bulk compositions of lithium-rich zones in pegmatites normally fall between petalite and quartz on the binary $\text{LiAlSi}_4\text{-SiO}_2$ (Stewart, 1978), the quartz-saturated phase diagram (Fig. 5) is appropriate for showing the stability fields of the lithium aluminosilicates in nature.

Reactions among the lithium aluminosilicates may be useful indicators of pressure and temperature. Shternberg *et al.* (1973) suggested that spodumene may be useful as a depth indicator, and Figures 4 and 5 indicate that reaction (Pet,Bsp) may constitute a geobarometer in that it establishes a minimum crystallization pressure for spodumene-bearing pegmatites. The ferric iron analogue of spodumene, $\text{LiFe}^{3+}\text{Si}_2\text{O}_6$, has been synthesized at moderate to very low pressure (Barrer and White, 1951; Drysdale, 1965), and substitution of iron and other elements in spodumene might therefore extend its stability field to quite low pressures (Appelman and Stewart, 1968). Natural spodumenes, however, usually contain less than two or three mole percent of impurities (Heinrich, 1975; Deer *et al.*, 1978), and these impurities exist largely in the form of micro-inclusions of other minerals (Graham, 1975). Therefore, the activity of $\text{LiAlSi}_2\text{O}_6$ in natural spodumene probably is near unity.

Implications for the crystallization of lithium pegmatites

Allowing for considerable uncertainty in the derived thermochemical data (Table 4) and for ionic

substitutions that make the system non-binary, the lithium aluminosilicate phase diagram here proposed (Figures 4 and 5) explains some commonly observed field associations and experimental results. The breakdown of petalite to β -spodumene reported from hydrothermal runs with bulk compositions between $\text{LiAlSi}_4\text{O}_{10}$ and SiO_2 (*e.g.*, Roy *et al.*, 1950; Skinner and Evans, 1960; Stewart, 1963, 1978; Munoz, 1969) may occur via reactions (Pet, Qtz, Ecr) and (Qtz, Spd) (Table 5 and Figure 4) at moderate T and low to moderate P . However, the topology of reactions depicted in Figure 4 suggests that petalite is stable to high T for bulk compositions from $\text{LiAlSi}_4\text{O}_{10}$ to SiO_2 (Rossovskii, 1967; Rossovskii and Matrosov, 1974; *cf.* also Stewart, 1963, and Bennington *et al.*, 1980, p. 13). The absence of β -spodumene phases in nature may be explained by the fact that the bulk compositional range of lithium-rich zones in pegmatites falls between $\text{LiAlSi}_4\text{O}_{10}$ and SiO_2 (Stewart, 1978), and reaction (Ecr, Spd) (Table 5 and Figure 5) probably lies at sufficiently high T that β -spodumene, if produced, would react completely with excess silica to form petalite above the lithium pegmatite solidus (see Jahns and Burnham, 1958).

The proposed stability relations among the naturally occurring lithium aluminosilicates are illustrated at invariant point [Bsp] in Figure 5. With decreasing T , petalite may break down to spodumene + quartz at moderate to high P (via reaction (Bsp,Ecr)) or to eucryptite + quartz at moderate to

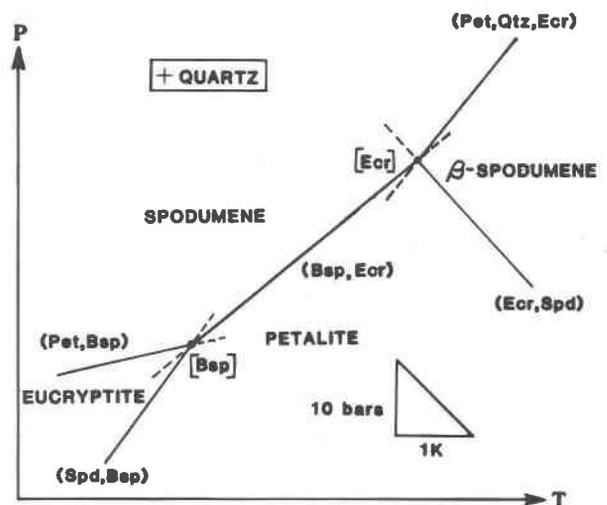


Fig. 5. P - T phase diagram illustrating the stability fields of the lithium aluminosilicates in (natural) quartz-saturated environments.

low P (via reaction (Spd,Bsp)). The lithium aluminosilicate stability relations presented in Figures 4 and 5 explain the isochemical replacement of petalite reported from numerous localities and account for the observed compatibility of eucryptite + quartz under geologic conditions of relatively low P and T . Based on existing thermochemical data, the proposed reaction (Pet,Bsp) should be highly pressure-dependent and would represent a minimum pressure boundary for crystallization of spodumene-bearing pegmatites and granites. Ionic substitutions in natural spodumene may extend its stability to somewhat lower pressures at the expense of eucryptite + quartz. These factors probably are largely responsible for the general absence of eucryptite + quartz pseudomorphs after spodumene and for the rarity of eucryptite + quartz assemblages. The following paper (London and Burt, 1982b) considers other chemical processes that appear to operate in lithium pegmatite evolution and that would also inhibit or preclude the formation of eucryptite + quartz.

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