

## **David B. Stewart (1978): Petrogenesis of lithium-rich pegmatites**

David London

University of Memphis

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### **Introduction**

David B. Stewart spent his entire career with the U.S. Geological Survey. He pursued field studies of the geology of coastal Maine early on, and later turned more to experimental petrology at the U.S.G.S. eastern regional headquarters in Reston, VA. Stewart coauthored several papers on the chemistry, crystal structure, and optical properties of the alkali feldspars. He might have observed pegmatites that are common in the metamorphic rocks of Maine. As a long-time friend of J.J. Norton, Stewart might have gained familiarity with the lithium-rich pegmatites of the Black Hills, SD. However he acquired his interest in pegmatites, he was among the first to investigate the stability relations among the lithium aluminosilicates, including spodumene ( $\alpha$ -LiAlSi<sub>2</sub>O<sub>6</sub>, a clinopyroxene), petalite (LiAlSi<sub>4</sub>O<sub>10</sub>, a phyllosilicate) and eucryptite ( $\alpha$ -LiAlSiO<sub>4</sub>, a feldspathoid) in a geological context. The lithium aluminosilicate system has had a long history of experimental study in the ceramics industry, but all such work has been at one atmosphere of pressure.

Stewart conducted most of his experimental research on the lithium aluminosilicates in the late 1950s. His first published results (Stewart, 1960, 1964) were an abstract and an obscure article (meaning, not widely known and difficult to obtain) from a professional meeting. It would be another 14 years before his work would appear in a widely circulated professional journal:

- Stewart, D.B. (1978) Petrogenesis of lithium-rich pegmatites. *American Mineralogist*, **63**, 970-980.

That article, the principal subject of this essay, is posted in PDF along with this essay by permission of the Mineralogical Society of America. Readers should refer to Table 3 and Figures 2 and 4 of Stewart's (1978) article, which are central to the discussion in this essay.

### **The Compositions of Lithium-Rich Pegmatites**

Stewart (1978) began by compiling calculations of the bulk compositions of lithium-rich pegmatites with the aim of estimating their Li<sub>2</sub>O content. He recognized the variability of zoned pegmatites as a problem in making that estimate. Zoned pegmatites are notoriously difficult to sample accurately, and the lithium-rich zones, all of which are late interior units, might be incompletely sampled, and might represent variable proportions of the entire pegmatite body. Unzoned spodumene pegmatites – in particular, Kesler's (1961) analyses of the deposits at Kings Mountain, NC – were more significant indicators of a spodumene-saturated bulk composition. Stewart's (1978) Table 3, which includes data from five pegmatites, showed them to contain 1.5 wt% Li<sub>2</sub>O with almost no deviation. There was no difference in composition (among any components) between pegmatites that contained spodumene and those that contained petalite, and whether zoned or unzoned.

## The System $\text{LiAlSiO}_4\text{-NaAlSi}_3\text{O}_8\text{-SiO}_2\text{-H}_2\text{O}$

Stewart's (1978) principal experimental contribution to this article was his Figure 4, the liquidus diagram for the system  $\text{LiAlSiO}_4$  (eucryptite)- $\text{NaAlSi}_3\text{O}_8$  (albite)- $\text{SiO}_2$  (quartz)- $\text{H}_2\text{O}$  at 200 MPa of  $\text{H}_2\text{O}$  pressure (i.e., all experiments were saturated in an aqueous solution, which Stewart, 1978, termed as "gas"). A eutectic (the lowest temperature at pressure at which a melt is in equilibrium with an invariant mineral assemblage) in the system was found at  $640^\circ \pm 10^\circ\text{C}$  for the assemblage of albite-quartz-petalite-melt-"gas". The lithium content of melt in equilibrium at the eutectic was 2.2 wt%  $\text{Li}_2\text{O}$  and varied little as a function of composition along the liquidus field boundaries of petalite or eucryptite. In terms of the weight percentage of mineral components, Stewart (1978) reported the eutectic composition as  $\text{Ab}_{34}\text{Qtz}_{27}\text{Pet}_{39}$  or  $\text{Ab}_{34}\text{Qtz}_{42}\text{Spd}_{24}$ . Relative to the eutectic composition on the albite-quartz sideline, the ternary eutectic liquid in the Li-saturated system contained 60% more quartz component, and Stewart (1978) observed that silica was more soluble than feldspar-forming components in the "gas". Stewart (1978) noted that the temperature of the eutectic is nearly  $100^\circ\text{C}$  lower than the eutectic on the albite-quartz- $\text{H}_2\text{O}$  sideline, and he speculated that the addition of potassium to the system would lower the eutectic by another  $35^\circ\text{C}$ , which would push the lowest melting or crystallization temperature to  $\sim 605^\circ\text{C}$  for a eutectic composition of albite-orthoclase-quartz-petalite-melt-"gas". Additional components would foster an even lower solidus temperature; for example, at the  $\text{H}_2\text{O}$  pressure of 200 MPa, the solidus of the Macusani obsidian, which is rich in boron and fluorine in addition to lithium, is  $450^\circ\text{C}$  (London et al., 1989).

Stewart (1978) drew a comparison between the experimental data of his Figure 4 and the bulk compositions of the pegmatite samples in his Figure 2. The correspondence between them is good. In Figure 2, the bulk compositions of the natural pegmatites plot closer to a feldspar component, and the compositions of the lithium aluminosilicate zones closer to quartz, relative to the experimental field boundaries and the eutectic. However, together they are close to the experimentally determined compositions and straddle the eutectic composition of Stewart's (1978) Figure 4.

More important than the details of Stewart's (1978) liquidus diagram was his demonstration that the lithium aluminosilicates, petalite or spodumene, can be stable magmatic phases in a quartz-rich pegmatite assemblage. Heretofore, the lithium aluminosilicates, like other exotic minerals, were widely believed to have formed by a subsolidus replacement process (e.g., Schaller, 1933). Jahns (1953) sought to counter that hypothesis with his textural assessment of crystal sizes and orientations. Stewart (1978) proposed that the assemblage of quartz-albite-spodumene or petalite (zone 6 of Cameron et al., 1949), with or without K-feldspar (zone 5 of Cameron et al., 1949), would be "*the last that can reasonably be interpreted to have originated by the crystallization of molten silicate magma*" (p. 970). In my opinion, this was the most significant contribution from Stewart's (1978) article.

## Petrogenesis of Lithium-Rich Pegmatites

Stewart (1978) used the data he had collected to frame his views on the origins of the lithium-rich pegmatites. He observed that:

*“Li-rich pegmatites occur farthest from the granite, and constitute one percent at most of the mass or number of pegmatites present. The Li-rich pegmatites occur immediately adjacent to or in sillimanite-grade metamorphic rocks, but not within related granites.”* (p. 970)

from which he concluded:

*“Fractional crystallization is a superficially attractive process to invoke for the generation of Li-rich magma. However, granites associated with lithium pegmatites typically contain less than 100 ppm Li... and even very perfect fractionation that would yield typical Li-rich pegmatite with ~ 7,100 ppm Li (= 1.53 weight percent Li<sub>2</sub>O) would require at least 70 times as much parental magma as pegmatite. Extreme mechanical problems arise in extracting such a small fraction of rest liquid. Alternate suggestions have been made (Norton, 1973, p. 369), and my present opinion is that anatexis is the hypothesis most consistent with geological and experimental evidence... Partial melting of Li-enriched metasediments would yield Li-rich magma at temperatures 75 °C (or more) lower than those at which granitic magmas of minimum melting compositions in the system Or-Ab-Qz-H<sub>2</sub>O could form... The irregular distribution of Li-rich pegmatites farthest of all pegmatite varieties from related granite and the sparsity of Li-rich pegmatites within related granites are in accord with an anatectic origin.”*(p. 978-979)

Throughout the history of the study of pegmatites, the prevalent understanding of most geologists, many of them very firmly grounded in field geology and in geochemistry, has been that pegmatites are late-stage residual melts (commonly termed “rest” liquids, meaning the rest of what’s left after extensive crystallization of the major mineral assemblages) that are derived by the fractional crystallization of larger masses of granitic magma. The derivation of pegmatite-forming melts from much larger masses of granitic magma was a foundational concept in the model of Jahns and Burnham (1969, and see essay #9 of this series). Pegmatites inherit their chemical attributes from those granites, and in turn, the granites mimic their source regions in ways that are traceable through their minor- and trace-element compositions and sometimes by their isotopic signatures (e.g., Černý 1991a,b; Černý et al., 2005, 2012, to name but a few). The vast majority of the lithium-rich pegmatites and the peraluminous granites with which they are associated possess the elemental characteristics of muscovite-rich schists and gneisses. Despite this trilogy of association, alternative viewpoints have also circulated since the beginning of study in the 19<sup>th</sup> century (see Jahns, 1955, and Chapter 2 of London, 2008, for reviews). An origin by direct anatexis is one of them. Stewart (1968) was one of the first of the few contemporary geologists to espouse it. That concept presumes that the first-formed melt from the same type of mica-rich metamorphic rocks that give rise to a granite is the same as the last chemically complex liquid that might fractionate from that granite. In other words, fractional crystallization and partial melting are identically reversible processes that give identical compositions of melt at the same fraction of melt to solids at the same temperatures. If there was any doubt about the fallacy of this assumption, it should have been dispelled as early as the first experimental work of Bowen (1913), who demonstrated that the compositions of plagioclase derived from partial melting experiments were wholly different from the compositions of crystals that formed by the crystallization of an undercooled melt; consequently, the melt compositions evolved along wholly different trajectories. This was the foundational evidence of Bowen’s concept of fractional crystallization and the evolution of chemically distinct melt compositions (a disequilibrium process) as distinct from the equilibrium relations of the liquidus diagram, which

he and others ascertained through partial melting experiments. There is a wealth of other evidence of all types, grounded in theory, experimentation, and the analysis of migmatites, obsidians, and trapped melt inclusions, that invalidate the anatectic model that Stewart (1978) endorsed. Some of the reasons were presented in essay #10 of this series. Other constraints have been discussed in section 9.7.1 of London (2018) and by Černý et al. (2005, 2012). Černý et al. (2005) began their consideration of the anatectic model for pegmatites (p. 357) with this opening remark: “*Anatectic proposals, applied to the origin of individual LCT [for lithium-cesium-tantalum] pegmatites, collide with numerous problems...*” Černý et al. (2005) then went on to outline ten significant conflicts of the anatectic model with other known aspects of granite-pegmatite geology.

***The compositions of anatectic melts.*** The anatectic model carries a connotation that in the process of anatexis, all of the exotic and fluxing components present in the source rocks would partition into a tiny fraction of melt, thereby facilitating the formation of the melt at low temperatures. This would be the means by which the first-formed melts would possess the high flux content and low solidus temperatures of the last-formed melts that are generated by fractional crystallization (e.g., see “interior glass pools with topaz” from experiment PEG-46 in Table 8 of London and Morgan, 2017). Proponents of the model have failed to explain how the whole-rock budget of exotic trace elements could or would migrate to a miniscule fraction of melt. It is also a chicken-or-egg problem of what comes first – the fluxes or the melt – because the melt cannot exist at the low temperatures of the model without the fluxes, and the fluxes reside in the metamorphic minerals until they have melted. Bea (1996) rationalized the compositions of anatectic melts, represented by the leucosomes (white lenses) in migmatites, by an observation that the minerals involved in melting do so more or less congruently, meaning that the concentrations of trace elements in the minerals are the same as their contribution to the melt. This is because the minerals dissolve at their surfaces into the melt; the partition coefficients for their trace elements, therefore, are unity in the relation to the proportion of the mineral that is melting. In that case, the first-formed melts that are derived from the anatexis of muscovite-bearing schists and gneisses would possess about the same trace-element concentration as the minerals that are involved in melting, which would be in the parts per million. Bea (1996) demonstrated this to be the case for migmatites. In a unique study of mineral-hosted glass inclusions that represented the actual, first-formed melts derived from muscovite schists, those glasses possessed low concentrations in single-digit to about a hundred parts per million of the important trace elements found in pegmatites (Acosta-Vigil et al., 2017), as predicted by Bea (1996). These concentrations of trace elements are anywhere from one hundred to ten thousands times below the threshold values for crystallizing rare-element minerals such as tourmaline, spodumene, beryl, pollucite, etc. at the solidus temperatures of granitic melts (e.g., see London, 2018).

What applies to the trace-element budgets of metamorphic rocks is equally relevant to the melting reactions of the major minerals. When heated to their solidus temperature, they melt in the proportions of a minimum or eutectic melt for the minerals that participate in the melting reaction. In every experimental study of the melting of muscovite-rich rocks at elevated H<sub>2</sub>O pressures, the first indications of melting occur at temperatures that are normally ascribed to the solidus of the granite system, close to 700°C (e.g., see Brown, 1994, London et al., 2012, and

citations therein). The first-formed melts are granitic, and they are not saturated in any of the rare minerals found in pegmatites.

***The relation of pegmatites to metamorphic grade.*** Černý (1991a) and London (2018) made this important distinction: the lithium-rich pegmatites lie in rocks near the staurolite-sillimanite isograd but generally on the low-temperature side of it at conditions that would place the peak of metamorphism of the host rocks in the upper greenschist facies of metamorphism at  $\leq 500^{\circ}$ - $550^{\circ}$ C. A large proportion of the rare-element pegmatites are hosted by greenstones, which are the medium-grade metamorphic products of basalts. This environment is nearly  $150^{\circ}$ - $200^{\circ}$ C below the minimum melting temperature of muscovite-albite-quartz schists and gneisses (e.g., Kerrick, 1972; London et al., 2012).

Stewart's (1978) anatectic model implied that the host rocks to the lithium pegmatites were at their peak of metamorphism when the pegmatite-forming melts were generated, albeit at temperatures too low to produce granitic liquids. However, the zoned lithium-rich pegmatites possess the textural features of quenched melts (London, 2008), and their temperatures of crystallization are in the range of  $375^{\circ}$ - $450^{\circ}$ C (London, 2018; London et al. 2020). If granitic melts – even highly fractionated melts like the Macusani obsidian – crystallized in this range of temperatures, then their host rocks, no matter what their mineralogical record of metamorphism, had to have been at temperatures of a normal continental geothermal gradient, which is about  $150^{\circ}$ - $200^{\circ}$ C at depths of  $\sim 6$ - $7$  kilometers, at the time of pegmatite emplacement. This is because the initial temperature of crystallization along the margins of a pegmatite-forming melt at its final emplacement is approximately the midpoint between the temperature of the melt and the temperature of the host rocks upon emplacement. For example, a temperature of crystallization at  $400^{\circ}$ C (London et al., 2020) is the midpoint between a magmatic temperature of  $650^{\circ}$ C (the liquidus temperature of the  $H_2O$ -saturated d Li-rich Macusani obsidian at 200 MPa: London et al., 1989) and a host rock temperature of  $150^{\circ}$ C, a value representative of a normal continental geothermal gradient at a depth of  $\sim 6$ - $7$  kilometers. The metamorphic host rocks reach temperatures of  $150^{\circ}$ - $200^{\circ}$ C by cooling upon uplift long after they achieved their peak metamorphic mineral assemblage. Therefore, the relationship of pegmatites to the peak metamorphic grade of their host rocks is a moot point. The distribution of pegmatites is unrelated to the metamorphic grade of their host rocks as much as it is a reflection of the geothermal gradient that exists long after those host rocks have cooled from their peak of metamorphism during uplift.

***Extraction and migration of pegmatite-forming melts.*** An implication of the anatectic model is that pegmatite-forming melts crystallized *in situ* in the rocks from which they were derived. In that case, the melts would have cooled and crystallized over millions of years in the course of uplift of their hosts. Such slowly cooled melts would not produce the textures of rapidly chilled melt bodies that are present in common and rare-element pegmatites (e.g., see Figure 9 and related text in London et al., 2020). Alternatively, pegmatite-forming melts that crystallized rapidly at temperatures hundreds of degrees below their liquidus would have to have migrated kilometers from their source into much cooler host rocks. As Stewart (1978) noted, the Li-rich pegmatites represent minute volumes of melt, and such small melt volumes could not have migrated from their source into rocks that were several hundreds of degrees cooler. The small batches of pegmatite-forming melt that they represent do not possess enough heat to remain fluid

over the distance of several kilometers. The lithium-rich pegmatites are, however, commonly the distal part of a field of thousands of common pegmatite bodies, which represent a much more voluminous batch of extracted melt.

As part of Stewart's (1978) argument, he compared the average lithium content of the end-members – the lithium pegmatites and the global granite average compositions – and he observed that the lithium pegmatites represent  $\sim 1/70$ , or 14% of the whole<sup>1</sup>. He is correct in his suggestion that it would be mechanically difficult to extract such a small fraction of melt from an otherwise crystalline granitic pluton. However, by only comparing the end points, Stewart (1978) left out everything in between, which includes the high-level and highly fractionated stocks of “pegmatitic granites” (e.g., Goad and Černý, 1981) and the system of thousands of dikes that comprise 99% of the pegmatite field. In personal remarks to me, Černý's retort to Stewart (1978) was to this effect: if one adds the rare elements of the pegmatite dike system back to their presumed granite source, then the granite source has an adequate budget of those rare elements to make all of the observed pegmatites. Duh.

In a study of beryllium geochemistry, London (2015) used a Rayleigh fractionation model to account for the derivation of beryl-saturated pegmatites. Starting with a Be value representative of Stewart's (1978) granite average ( $\sim 6$  ppm Be), about 70% of crystallization would bring the residual melt to the Be content of the small pegmatitic granite stocks ( $\sim 18$  ppm Be). That remaining fraction of melt (30%) is extractable from the crystalline mush (e.g., Bachman and Bergantz, 2004) to form the pegmatitic granites as cupolas on much larger batholiths. With another 70% of crystallization of the pegmatitic granite stock, the extraction of the remaining 30% of its residual melt lies at the beryl-saturation threshold in pegmatites (30 ppm: Evensen and London, 2002), but only if the pegmatite-forming melts are substantially undercooled when they begin to crystallize. Thus, London (2015) proposed that the derivation of pegmatite-forming melts is an episodic and protracted process of extraction. Sweetapple and Collins (2002) have documented that this sequence of events happens over many millions of years of magmatic evolution in a belt of granite batholiths, evolved plutons, and the pegmatites they spawn.

### The Lithium Aluminosilicate Phase Diagram

Donald M. Burt, my graduate advisor, produced a Schreinemaker analysis of the lithium aluminosilicate phase diagram (Burt et al., 1977; London and Burt, 1982) in the system  $\text{LiAlSiO}_4\text{-SiO}_2$  containing the phases  $\alpha$ -eucryptite,  $\alpha$ -spodumene, petalite, and quartz (Figure 1).

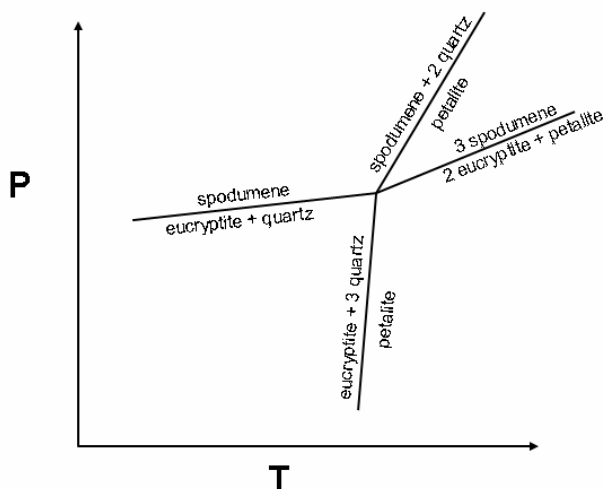


Figure 1

The published results of experiments by Stewart (1964) and Muñoz (1969) appeared to be in error (too high in temperature at pressure) on the location of the reaction  $\text{spodumene} + 2 \text{ quartz} = \text{petalite}$  at low pressure based on my thermodynamic calculations of the pressures and temperatures of the reaction boundaries. In 1980, therefore, I set off for Washington, DC, to conduct the experiments that would validate the topology and add actual pressures and temperatures to the reaction boundaries. I had sponsors at the Smithsonian Institution (Daniel E. Appleman), at the Carnegie Geophysical Laboratory (Hatten S. Yoder, Jr.), and Dave Stewart sponsored my work at the U.S.G.S. headquarters in Reston. Stewart might have been vexed that I showed up to reinvestigate his work of the late 1950s, especially as his own paper on lithium minerals finally appeared in press just two years earlier. He could have refused to cooperate with me when I (an aspiring Ph.D. applicant) suggested to him that one of his published reaction boundaries was incorrect in its location in pressure and temperature. Instead, Stewart not only opened the experimental lab to me, but also let me examine all of his experimental products and notes on results, and we discussed those at length. I found the source of the error: in his experiments with petalite, he had added so much water that all of the quartz dissolved into the aqueous solution, and his system was no longer quartz-saturated as he intended. He had slipped onto the metastable extension of the quartz-absent reaction of  $3 \text{ spodumene} = \text{petalite} + 2 \text{ eucryptite}$  at higher temperature. Stewart accepted the correction, which was discussed in the final publication of my experimental work<sup>2</sup> (London, 1984), with humility and a characteristically wry quip<sup>3</sup> in his review of the manuscript. Dave Stewart, however, was an impeccable scientist for whom getting the right answer was more important than defending his own work.

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## Footnotes

<sup>1</sup> London (2019) estimated the mass of granitic magma that would be necessary to produce the Tanco pegmatite based on the concentrations of cesium in the pegmatite and the partitioning of cesium between minerals and melt. The volume of the Tanco pegmatite is ~ 0.06 cubic kilometers. The volume of granitic melt necessary to produce the cesium concentration in that volume is 9 cubic kilometers. The ratio of granite to pegmatite, therefore, is 150.

<sup>2</sup> From London (1984), p. 997:

*“The resultant slope of the reaction  $Pet = Spd + 2 Qtz$  (Table 3) is considerably shallower than the value reported by Stewart (1963: 23.5 bar/°C), and extends the stability field of petalite to much lower T at low P. Discrepancies in the reported experimental results of this reaction at 2 kbar (cf. Stewart, 1963; Munoz, 1971) appear to be due to differences in experimental technique. In this study, quartz saturation was maintained in all subsolidus fields. The petalite bulk composition employed by Stewart (1963) and Munoz (1971), apparently became quartz undersaturated (i.e., shifted toward spodumene) in runs into the petalite stability field, with the result that small amounts of spodumene would persist stably with petalite until the reaction  $3 Spd = Pet + 2 Ecr$  that is terminal to spodumene was encountered at higher T. The reaction boundaries reported by Stewart (1963) and Munoz (1971) may be for the quartz-absent eucryptite-producing reaction cited above. In this regard, it is significant that Stewart's (1978) evidence for silica leaching into the aqueous phase is based in part on the appearance of small amounts of eucryptite in these runs.”*

<sup>3</sup> Stewart was tall, with a deep, gruff voice, direct and blunt in his demeanor, and brilliant, all meaning that he could easily be intimidating to a graduate student. The author of his obituary, however, described him correctly: *“He did not suffer fools easily, but this attribute was offset by a quick wit and disarming self-deprecation. He had a wonderful sense of humor. He was a warm and loyal friend, welcoming and hospitable, and an attentive mentor to many younger colleagues.”* see <https://www.legacy.com/us/obituaries/washingtonpost/name/david-stewart-obituary?id=6051555>