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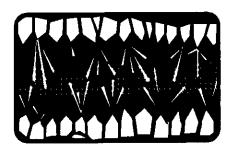
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# Articles



# Rare-element Granitic Pegmatites. Part I: Anatomy and Internal Evolution of Pegmatite Deposits

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#### INTRODUCTION

Granitic pegmatites are one of the classic sources of a broad spectrum of rare metals: many exotic elements were discovered in pegmatite minerals, and Li, Rb, Cs, Be, Ga, Sc, Y, REE (rare earth elements), Sn, Nb, Ta, U, Th, Zr and Hf have been extracted from them, Industrial minerals, such as ceramic and dental feldspar, optical quartz and fluorite, petalite and refractory spodumene, sheet and crushed micas, and ceramic amblygonite, also are valuable, as is mining of gernstock and collectibles. High concentrations of raremetal mineralization and high purity of most of the industrial minerals, combined with their coarse grain size, are the main factors favouring pegmatite exploitation. A potential for simultaneous extraction of a variety of ore and industrial minerals also makes pegmatites attractive exploration targets.

The high technology signature of pegmatite ore deposits is particularly favourable in the present market conditions. Consumption of rare elements is steadily increasing in a variety of industrial technologies, and this trend will undoubtedly continue into the future. Beside the already classic uses of lithium in storage batteries, ceramic ware and smelting of aluminum ores, beryllium

bronze can be quoted in applications requiring high resistance to metal fatigue, other beryllium alloys are used in aircraft, satellite and space-vehicle frames, and beryllium metal is used in heat shields, rocket motors, aircraft and space-shuttle brake discs, inertial navigation systems and X-ray windows. Compounds of gallium serve in the manufacture of opto-electric devices such as lightemitting diodes, photodiodes, laser diodes and solar cells, fibre optics and a wide range of consumer electronic products, including computers and communication processors of electronic signals. Tantalum is also used in computers and communication technology in space, defence and industrial fields, and in capacitors of solid-state circuits. The chemical industry and production of prosthetic devices also consume a sizeable proportion of tantalum supply. These examples of technological uses of a few metals are just a tip of the iceberg of growing industrial applications of rare elements.

Pegmatite mining operations compete today against several new deposit types with large tonnages of low-grade ore. It is only the largest pegmatite deposits which can be exploited with mechanized operation that are economically viable on the present market, and the number of these sources is rather restricted (Table 1). However, extensive populations of small mineralized pegmatites are abundant, and mining at communal to family scale contributes a significant proportion of world production of some commodities, e.g., Be and Ta in Brazil, India and numerous African countries. Consequently, pegmatites require a rather thorough treatment in the present series on ore deposit models because of their diversity in geological setting, composition and economic significance. Also, understandably enough, the mining community is not overly familiar with pegmatite deposits in general, and with recent developments in their petrology in

For these reasons, principal classifications of granitic pegmatites will be reviewed here first, to clarify nomenclature, geological affiliation and geochemical features of individual categories. Descriptive characteristics and genetic models will be approached separately at three levels: individual pegmatites are treated in Part I, whereas local cogenetic groups and large-scale populations of pegmatite fields, belts and provinces are dealt with in Part II (Černý, 1991c). Such a treatment will not only maintain a "reversed continuity", beneficial to gradual build-up of a preferred genetic model, but will also allow space necessary for comments on controversial views and gaps in our present knowledge.

Pegmatite research was originally descriptive, and aimed at the mineralogy and structure of individual bodies. Attempts at petrological interpretations (such as those of Niggli, Fersman, Landes and Schaller) were very restricted during the first half of the 20th century. Systematic field studies by the United States Geological Survey in the 1940s and 1950s (Cameron, Heinrich, Jahns, Norton, Page and others) significantly boosted investigation of individual deposits and mapping of pegmatite districts. They also developed a generalized system of describing internal structure of individual bodies, and led to several genetic concepts of internal pegmatite evolution. The 1950s to 1970s were marked by an extensive effort of Soviet geologists (Beus, Ginsburg, Gordiyenko, Kuzmenko, Nedumov, Rossovskyi, Shmakin, Solodov, Vlasov and others), which generated classifications, exploration criteria and genetic interpretations of large-scale pegmatite populations. During the same period, modern petrology of pegmatite crystallization was initiated in the United States by Jahns, Burnham and Stewart. This has been maintained into the 1980s by Brown, Burt, Ewing, Foord and London. All of this was originally small-scale activity by a few isolated individuals. However, pegmatite research has started gaining momentum in recent years, thanks to extensive input from studies of the petrology of granites and rhyolites. Today, a sizeable international group of investigators contributes to our understanding of different aspects of the pegmatite process. Theoretical and experimental petrology and detailed field studies, coupled with meticulous mineralogy and geochemistry, crystal-chemical and crystallographic research, isotope systematics and exploration of virgin pegmatite populations, are the main lines of investigation that have expanded during the last decade. This late start into the "contemporary" petrology is the main reason why some fundamental problems of pegmatite petrology — a process encompassing magmatic, supercritical, hydrothermal and gaseous phenomena in unique chemical systems — are still debated and questioned. The gaps in our knowledge, and subjects to be examined in the future, will become self-evident in the following synthesis.

#### **TERMS OF REFERENCE**

The subjects of the present study are granitic pegmatites of the rare-element class which carry the qualitatively most diversified, and quantitatively most enriched, assemblages of rare metals. This category was recognized and defined by Ginsburg et al. (1979) and Ginsburg (1984) as part of a four-fold subdivision, which introduced geological law and order into the jungle of rock types that were indiscriminantly called pegmatites, and that had defied attempts at a uniform genetic interpretation. Table 2 and Figure 1 (modified from Černý, 1989a, 1990) illustrate the rea-

Manono-Kitololo, Zaire

Minas Gerais and Ceara states, Brazil

Table 1

sons behind these difficulties: lumping together vastly different products of fundamentally different processes, located in diverse crustal environments, just could not work.

This classification outlines the principal conditions of pegmatite formation in four crustal settings, and provides the basic tool of decision-making at the most general continental scale of exploration strategy.

The class of rare-element pegmatites is subdivided from a paragenetic-geochemical viewpoint into a number of pegmatite types and subtypes, shown in Table 3 (Černý, 1982a, 1989a, 1990, 1991b). An unorthodox combination of different criteria is required to define them: bulk chemistry, geochemical signature of accessory minerals, internal structure, and P-T conditions of crystallization are employed to differentiate the spectrum of categories. Thus, the sequence from rare-earth to complex type is based on bulk plus trace composition, whereas subtypes of the complex pegmatites are defined in part by P-T conditions and in part by chemical

(modified from Sinclair, 1991 (includes references)).

Production reserves of some pegmatite deposits

potentials of subordinate anions. In contrast, the albite-spodumene type differs from the spodumene subtype of complex pegmatites mainly by internal structure, but the albite type is separated on the basis of bulk composition.

This subdivision of the rare-element pegmatite class provides orientation among, and within, diverse pegmatite populations on a broad to local regional scale. It rationalizes regional search for deposits of specific rare metals.

Table 4 presents a breakdown of rareelement pegmatites into three families — LCT, NYF and mixed — to emphasize petrochemical differences among the types outlined above (Černý, 1991a). The pegmatite families are characterized by their bulk composition, typical assemblages of trace elements, and types of granites associated with them. This subdivision is instrumental in petrogenetic studies, pointing out different crustal sources of the silicate matrix and its content of rare elements.

Deposit	Production/Reserves	Comments
Tanco, Manitoba	2.1 Mt, 0.216% ${\rm Ta_2O_5}$ 7.3 Mt, 2.76% ${\rm Li_2O}$ (in spodur 0.3 Mt, 23.3% ${\rm Cs_2O}$ 0.9 Mt, 0.20% BeO 0.1 Mt, 2.50% ${\rm Rb_2O}$	Pre-production reserves mene + petalite)
Preissac-Lacorne area, Quebec	19 Mt, 1.25% Li₂O	Pre-production reserves from the former Quebec Lithium property
FI (J.MLit), Yellowknife, NWT	13.9 Mt, 1.19% Li <sub>2</sub> O	"Identified paramarginal resources"
Thor (Echo-Tanco), Yellowknife, NWT	8.4 Mt, 1.5% Li <sub>2</sub> O	"Identified paramarginal resources"
Violet, Herb Lake, Manitoba	5.9 Mt, 1.2% Li <sub>2</sub> O	Reserves
Nama Creek, Georgia Lake, Ontario	3.9 Mt, 1.06% Li <sub>2</sub> O	Reserves
Lac la Croix, Ontario	1.4 Mt, 1.3% Li <sub>2</sub> O	Reserves
Madawaska (Faraday), Ontario	4.5 Mt, 0.09% U <sub>3</sub> O <sub>8</sub>	Production 1956-1964 and 1976-1982
Tin-spodumene belt, North Carolina	26 Mt, 1.5% Li 30.5 Mt, 1.5% Li <sub>2</sub> O	Measured and indicated reserves, Foote Mineral Co. Reserves, Lithium Corporation of America
Bikita, Zimbabwe	10.8 Mt, 3.0% Li <sub>2</sub> O	Total lithium reserves
Kamativi, Zimbabwe	100 Mt, 0.114% Sn 100 Mt, 0.603% Li <sub>2</sub> O	"Maximum inferrable reserves of a single pegmatite"
Uis, Namibia	87 Mt, 0.134% Sn	Reserves in 1983
Greenbushes, Australia	28 Mt, 0.114% Sn 28 Mt, 0.043% ${\rm Ta_2O_5}$ 28 Mt, 0.031% ${\rm Nb_2O_5}$ 33.5 Mt, 2.55% ${\rm Li_2O}$	Proven and probable reserves

35 Mt, 1.3% Li<sub>2</sub>O

106 Mt, 0.04% BeO

Proven reserves

Estimated in situ ore

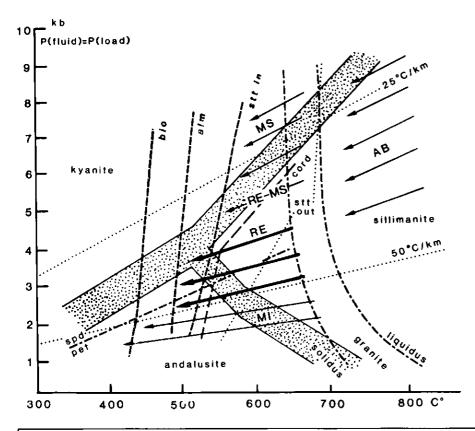


Figure 1 P-T fields of environments hosting pegmatite populations of the abyssal (AB), muscovite (MS), transitional rare-element + muscovite (RE-MS), rare-element (RE) and miarolitic (MI) classes, modified after Ginsburg et al. (1979) and Černý (1990). Arrows indicate regional fractionation trends relative to metamorphic grades of the host rocks. Aluminosilicate boundaries from Robie and Hemingway (1984), cordierite in metapelites after Schreyer and Seifert (1969), spodumene-petalite from London (1984), granite liquidus-solidus after Jahns (1982), other boundaries from Winkler (1967, 1976).

Class	Family*	Typical Minor Elements	Metamorphic Environment	Relation to Granites	Structural Features	Examples			
Abyssal	_	U,Th,Zr,Nb,Ti,Y, REE,Mo poor (to moderate) mineralization	(upper amphibolite to) low- to high-P granulite facies; ~4-9 kb, ~700-800°C	none (segregations of anatectic leucosome)	conformable to mobilized cross-cutting veins	Rae and Hearne Provinces, Sask. (Tremblay, 1978); Aldan and Anabar Shields, Siberia (Bushev and Koplus, 1980); Eastern Baltic Shield (Kalita, 1965)			
Muscovite	_	Li,Be,Y,REE,Ti, U,Th,Nb>Ta poor (to moderate)** mineralization; micas and ceramic minerals	high-P, Barrovian amphibolite facies (kyanite-sillimanite); ~5-8 kb, ~650-580°C	none (anatectic bodies) to marginal and exterior	quasi- conformable to cross- cutting	White Sea region, USSR (Gorlov, 1975); Appalachian Province (Jahns et al., 1952); Rajahstan, India (Shmakin, 1976)			
Rare- element	LCT	Li,Rb,Cs,Be,Ga,Sn, Hf,Nb≷Ta,B,P,F poor to abundant mineralization; gemstock; industrial minerals	low-P, Abukuma amphibolite (to upper greenschist) facies (andalusite- sillimanite); ~2-4 kb, ~650-500°C	(interior to marginal to) exterior	quasi- conformable to cross- cutting	Yellowknife field, NWT (Meintzer, 1987); Black Hills, South Dakota (Shearer et al., 1987); Cat Lake-Winnipeg Rive field, Manitoba (Černý et al., 1981)			
	NYF	Y,REE,Ti,U,Th,Zr, Nb>Ta,F; poor to abundant mineralization; ceramic minerals	variable	interior to marginal	interior pods, conformable to cross- cutting exterior bodies	Llano Co., Texas (Landes, 1932); South Platte district, Colorado (Simmons <i>et al.</i> , 1987); Western Keivy, Kola, USSR (Beus, 1960)			
Miarolitic	NYF	Be,Y,REE,Ti,U,Th, Zr,Nb>Ta,F; poor mineralization; gemstock	shallow to sub- volcanic; ~1-2 kb	interior to marginal	interior pods and cross- cutting dykes	Pikes Peak, Colorado (Foord, 1982); Idaho (Boggs, 1986); Korosten pluton, Ukraine (Lazarenko <i>et al.</i> , 1973)			

between the muscovite and rare-element classes proper

	Table 3	Classification	n of pegmatite	s of the rare-element class.		
PEGMATITE TYPE [feldspar + mica content]	Pegmatite subtype, geochemical signature	Typical minerals	Economic potential	Typical examples		
RARE- EARTH [Kf>plg to ab; bi≥msc]	allanite-monazite (L)REE,U,Th (P,Be,Nb>Ta)	allanite monazite	(REE)	Upper Tura River, Ural Mtns. (Fersman, 1940) West Portland, Quebec (Spence and Muench, 1935) Kobe, Japan (Tatekawa, 1955)		
	gadolinite Y,(H)REE,Be,Nb>Ta F(U,Th,Ti,Zr)	gadolinite fergusonite euxenite (topaz) (beryl)	Y,REE,U (Be,Nb-Ta)	Shatford Lake group, Manitoba (Černý et al., 1981) Ytterby, Sweden (Nordenskjöld, 1910) Evje-Iveland field, Norway (Bjørlykke, 1935) Barringer Hill, Texas (Landes, 1932) Pyörönmaa, Finland (Vorma et al., 1966)		
<i>BERYL</i> [Kf>ab; msc>bi]	<i>beryl-columbite</i> Be,Nb≷Ta (±Sn,B)	beryl columbite- tantalite	Be	Meyers Ranch, Colorado (Hanley et al., 1950) Greer Lake group, Manitoba (Černý et al., 1981) Donkerhoek, Namibia (Schneiderhöhn, 1961) Ural Mtns., USSR (Kuzmenko, 1976)		
	beryl-columbite- phosphate Be,Nb≷Ta,P (Li,F,±Sn,B)	beryl, colum- bite-tantalite triplite triphylite	(Nb-Ta)	Hagendorf-Süd, Germany (Strunz et al., 1975) Dan Patch, South Dakota (Norton et al., 1964) Connecticut localities (Cameron and Shainin, 1947) Crystal Mtn. field, Colorado (Thurston, 1955)		
COMPLEX [Kf≷ab; msc≷lep]	spodumene Li,Rb,Cs,Be,Ta≷Nb (Sn,P,F,±B)	spodumene beryl tantalite (amblygonite) (lepidolite) (pollucite)	Li,Rb, Cs,Be	Harding, New Mexico (Jahns and Ewing, 1976) Hugo, South Dakota (Norton et al., 1962) Mongolian Altai #3 (Wang et al., 1981) Etta, South Dakota (Norton et al., 1964) White Picacho, Arizona (London and Burt, 1982a) Manono, Zaire (Thoreau, 1950)		
	petalite Li,Rb,Cs,Be,Ta>Nb (Sn,Ga,P,F,±B)	petalite beryl tantalite (amblygonite) (lepidolite)	Ta, (Sn,Ga,Hf)	Tanco, Manitoba (Černý, 1982c) Bikita, Zimbabwe (Cooper, 1964) Varuträsk, Sweden (Quensel, 1956) Luolamäki, Finland (Neuvonen and Vesasalo, 1960) Londonderry, Australia (McMath <i>et al.</i> , 1953) Hirvikallio, Finland (Vesasalo, 1959)		
	lepidolite F,Li,Rb,Cs,Be Ta>Nb (Sn,P,±B)	lepidolite topaz beryl microlite (pollucite)	Li,Rb, Cs,Ta Be (Sn,Ga)	Brown Derby, Colorado (Heinrich, 1967) Pidlite, New Mexico (Jahns, 1953b) Himalaya district, California (Foord, 1976) Khukh-Del-Ula, Mongolia (Vladykin <i>et al.</i> , 1974) Wodgina, Australia (Blockley, 1980)		
	amblygonite P,F,Li,Rb,Cs Be,Ta>Nb (Sn,±B)	amblygonite beryl tantalite (lepidolite) (pollucite)	Li,Rb Cs,Ta Be (Sn,Ga)	Viitaniemi, Finland (Lahti, 1981) Malakialina, Madagascar (Varlamoff, 1972) Peerless, South Dakota (Sheridan et al., 1957) Finnis River, Australia (Jutz, 1986)		
ALBITE-SPOD [ab>Kf; ((msc))]	DUMENE Li (Sn,Be,Ta≷Nb, ±B)	spodumene (cassiterite) (beryl) (tantalite)	Li,Sn (Be,Ta)	Kings Mountain, North Carolina (Kesler, 1976) Preissac-Lacorne, Quebec (Mulligan, 1965) Peg Claims, Maine (Sundelius, 1963) Volta Grande, Brazil (Heinrich, 1964)		
ALBITE [ab>>Kf; (ms,lep)]	Ta≷Nb,Be (Li,±Sn,B)	tantalite beryl (cassiterite)	Ta (Sn)	Hengshan, China (Černý, 1989a) USSR (Solodov, 1969) Tin Dyke, Manitoba (Chackowsky, 1987)		

Further information on the diverse classification systems and their categories is available in Solodov (1971), Kuzmenko (1976), Ginsburg et al. (1979) and Černý (1982a, 1989a,b, 1991a,b).

#### GENERAL CHARACTERISTICS OF RARE-ELEMENT PEGMATITES Shape, Attitude and Size

The forms of rare-element granitic pegmatites are greatly variable, and are controlled mainly by the competency of the enclosing rocks, the depth of emplacement, and the tectonic and metamorphic regime at the time of emplacement.

Schists, gneisses and early intrusions are the predominant hosts of rare-element pegmatites of the LCT family. These pegmatites occur rarely in granitoid intrusions, as networks of fracture-filling dykes hosted by contraction fractures or structures generated by post-consolidation stresses. The shapes of these pegmatites are controlled by the ductility of their host rocks. They range from lenticular, ellipsoidal, turnip- or mushroomshaped forms in plastic-behaving lithologies to fracture-filling dykes and stocks in brittle

host rocks. In contrast, members of the NYF family commonly reside within consanguineous plutons, as facial pods, turnip-like balloons or fracture-filling dykes.

Attitudes range from horizontal to vertical, and they seem to be fairly typical for specific geochemical types; e.g., tabular pegmatites of the most fractionated complex subtypes tend to be subhorizontal. Shape and attitude have considerable control over internal structure and distribution of mineralization.

Lenticular and tabular rare-element pegmatites may be only several metres long and less than 1 m thick, and most of the economically interesting bodies average hundreds of metres in length and a few tens of metres across; stock- or turnip-shaped bodies rarely reach over 100 m. Among the well-zoned giants, the Greenbushes deposit in Australia has been tracked for 3300 m along strike, 400 m in outcrop width and >500 m downdip. The Li- and Sn-rich Manono pegmatite in Zaire has a collective length of subparallel, closely spaced veins of 12 km, with variable widths of 200-700 m. The complex Tanco pegmatite in Manitoba is 1650×800×125 m in size.

#### Internal Structure

Rare-element pegmatites commonly exhibit heterogeneous, complex internal structure. In the broad spectrum of structural patterns, three principal types can be distinguished, typical components of which can usually be recognized even in the most complicated cases: homogeneous, zoned and layered structures.

Homogeneous coarse-grained pegmatites with more or less uniform distribution of most components are exceptional, although even the most fractionated, petalite- and pollucite-bearing subtypes are found among them. The only variety which consistently exhibits a quasi-homogeneous structure is the albite-spodumene type. Except for subtle compositional and textural zoning along contacts, occasional layered aplitic assemblages, and rare pods of randomly oriented blocky texture in their central parts, these pegmatites consist mainly of elongate megacrysts of spodumene and quartz, imbedded in and somewhat corroded by a fine-grained albite + quartz matrix. The megacrysts have semi-random to subparallel orientation, subnormal to oblique to the wall-rock contacts

	Table 4 The three petrogenetic families of rare-element pegmatites.						
Family	Pegmatite types	Geochemical signature	Pegmatite bulk composition	Associated granites	Granite bulk composition	Source lithologies	Examples
LCT	beryl complex albite- spodumene albite	Li,Rb, Cs,Be,Sn,Ga, Ta>Nb(B,P,F)	peraluminous**	(synorogenic to) late orogenic (to anorogenic); largely heterogeneous	peraluminous S,I or mixed S+I types	undepleted upper- to middle-crust supracrustals and basement gneisses	Bikita field, Zimbabwe (Martin, 1964); Utö- Mysingen field, Sweden (Smeds and Černý, 1989); White Picacho field, New Mexico (London and Burt, 1982a)
NYF •	rare-earth	Nb>Ta,Ti, Y,Sc,REE, Zr,U,Th, F	subaluminous to metaluminous (to subalkaline)	(syn-, late, post- to) mainly anorogenic; largely homogeneous	(peraluminous to) subalumin- ous to metaluminous (rarely peralkaline); A and (I) types	depleted middle to lower crustal granulites, or undepleted juvenile granitoids	Shatford Lake group, Manitoba (Černý et al., 1981); Bancroft- Renfrew field, Ontario-Quebec (Černý, 1990; Lentz, 1991); Stockholm area, Sweden (Nordenskjöld, 1910; Smeds, 1990)
Mixed	"cross- bred" LCT and NYF	mixed	(metaluminous to) moderately peraluminous	(postorogenic to) anorogenic; moderately heterogeneous	subaluminous to slightly peraluminous; mixed geochemical signature	mixed protoliths, or assimilation of supracrustals by NYF granites	Tørdal district, S. Norway (Juve and Bergstøl, 1988); Kimito, Finland (Pehrman, 1945); Evje-Iveland field, S. Norway (Bjørlykke, 1935; Frigstad, 1968).

#### Notes

- \* This family also includes pegmatites of the miarolitic class (cf. Table 2)
- \*\* Definitions: Peraluminous, A/CNK>1; subaluminous, A/CNK<1; metaluminous, A/CNK<1 at A/NK>1; subalkaline, A/NK<1; peralkaline, A/NK<1, where  $A = molecular Al_2O_3$ , CNK = CaO + Na<sub>2</sub>O + K<sub>2</sub>O, and NK = Na<sub>2</sub>O + K<sub>2</sub>O (Černý, 1991a)

(Figure 2). The preferred orientation is, in some cases, generated by recrystallization under stress and is superimposed on the original fabric (Kings Mountain deposits, Kesler, 1976; INCO pegmatite, Chackowsky, 1987), but, even at these localities, it represents just a minor modification of the primary texture, which also must have crystallized in a stress field (Peg claims, Sundelius, 1963; Georgia Lake, Pye, 1965; Violet Claim, Černý et al., 1981).

Zoned pegmatites are the most common ones, and the most diversified. They consist of up to nine different units with variable textural characteristics and mineral modes (Figure 3). The most evolved zonal patterns are encountered in highly fractionated intrusions with complex mineralization such as Varuträsk (Quensel, 1956), Hugo (Figure 4; Norton et al., 1962), Bikita (Cooper, 1964) and Tanco (Figure 5; Černý, 1982b).

According to Heinrich (1948) and Cameron et al. (1949), we may distinguish: (1) zones of primary crystallization, more or less complete concentric shells, termed from the margins inward as the border, wall, intermediate (outer, middle, inner, core-margin) zones, and the core; (2) replacement bodies formed at the expense of pre-existing units under lithologic or structural control; and (3) fracture fillings in dilated dislocations. Zones of primary crystallization differ in mineral assemblages, modes and textural styles. With progressive crystallization from the margins inward, zones usually show increasing grain size, decreasing number of rock-forming minerals, and textural changes from granitic or aplitic through graphic or heterogeneous in intermediate zones to blocky and coarsegrained monomineralic in the core.

Replacement units are not as easily identified and defined as are the primary zones. Metasomatic effects range widely in extent and intensity, from selective replacement of individual mineral species (e.g., micas after beryl or topaz), through pervasive, but diffuse, assemblages replacing primary minerals throughout entire zones (e.g., albite + lithian mica after K-feldspar), to massive metasomatic units that replace the bulk of the primary assemblage in pre-existing unit(s). It is this third case which represents mappable pegmatite units, but which possesses only restricted textural evidence of metasomatic origin. Among the mineral assemblages commonly interpreted as metasomatic are massive lepidolite units, and units consisting mainly of saccharoidal or platy albite (Figure 4). They also belong to the most controversial phenomena, as discussed later; some authors consider them primary (e.g., Norton, 1983; Černý et al., 1981; London, 1986a, 1990; Thomas and Spooner, 1988b).

In contrast to the metasomatic assemblages, fracture-filling units are easy to recognize (Figure 3). They are rather scarce, and volumetrically insignificant. The most common case is that of quartz fracture-

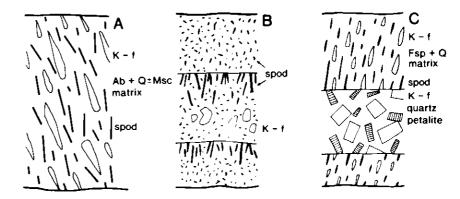


Figure 2 Internal structure of homogeneous pegmatites in cross-sections. (A) Preferred orientation of spodumene and K-feldspar in albite-spodumene type, Kings Mountain, North Carolina (e.g., Kunasz, 1982); (B) Cockscomb texture of spodumene in multiple intrusions of parallel pegmatite dykes, Volta Grande, Brazil (after Heinrich, 1964); (C) Early spodumene-bearing pegmatite (as in A) penetrated by a parallel dyke with random orientation of K-feldspar and petalite, Järkvissle, Sweden.

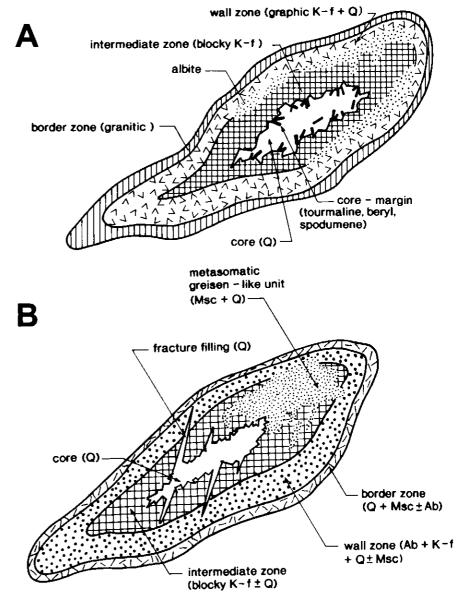


Figure 3 Internal structure of zoned pegmatites in schematic horizontal sections. (A) Concentric pattern of primary zones, with zonal control of the distribution of albite units, and core-margin mineralization; (B) Concentric pattern of primary zones cross-cut by fracture fillings, with a lithology-controlled, but, in part, fracture-related, metasomatic unit.

fillings, rooted in the core and transecting intermediate zones.

Many attempts have been made to generalize a zonal sequence which would encompass all units typical of zoned pegmatites. The idealized 11-member sequence of Cam-

eron et al. (1949) was reduced to 9 units by Norton (1983): (1) plagioclase, quartz, muscovite; (2) plagioclase, quartz; (3) quartz, perthite, plagioclase; (4) perthite, quartz; (5) quartz, plagioclase, perthite, spodumene (or petalite), amblygonite; (6) quartz, spodu-

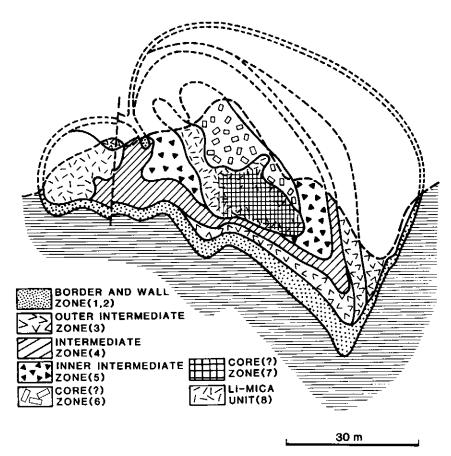


Figure 4 Vertical section through the Hugo pegmatite near Keystone, Black Hills, South Dakota (modified from Norton et al., 1962). The extrapolation above the erosion and mining surface is realistic, based on analogies with closely related adjacent pegmatites. Zone 5 carries abundant spodumene, zones 6 and 7 contain mainly K-feldspar and quartz; lithian mica constitutes the bulk of the partially metasomatic unit 8; cassiterite and columbite are concentrated in zones 3, 4 and 8.

mene (or petalite); (7) quartz, microcline or perthite; (8) quartz; and (9) lepidolite or lithian mica, plagioclase, quartz, microcline. This sequence may serve as a guide in field work and description, but it is overgeneralized and lacks textural attributes. It is rather meaningless in petrological research; some of the important features of complex pegmatites (such as saccharoidal albite units) remain obscure.

Distribution of the internal units in zoned pegmatites is influenced by the shape and attitude of individual intrusions, and by the unit lithology (e.g., Beus, 1960; Solodov, 1971). The most prominent bulges usually have the most evolved internal structure, whereas constrictions and pinching-out margins may consist of only the outermost zone of the concentric pattern. Vertical asymmetry is commonly expressed by updip displacement of inner zones and replacement units, e.g., "hood"-forming K-feldspar accumulations and mica-rich units. Vertical asymmetry is locally evident even in shallowdipping zoned pegmatites, usually expressed in widely different thicknesses of footwall and hanging wall parts of concentric zones.

Lavered pegmatites may be considered extreme cases of the geometric and compositional asymmetry of zoned intrusions. They are usually thin and subhorizontal, with sodic aplitic units in their footwall portions and K-feldspar + quartz-rich pegmatitic units close to the top (Figure 6). Highly fractionated layered pegmatites commonly have numerous, and occasionally large, crystallined vugs. Classic examples of the lavered structure are the lepidolite-rich tantalum deposits of the Quartz Creek field (Staatz and Trites, 1955; Heinrich, 1967), and the gembearing pegmatites of the Rincon, Mesa Grande and Pala districts (Foord, 1976; Stern et al., 1986) and Nilau-Kulam field (Rossovskyi, 1981a,b).

Two aspects of internal structure must be emphasized in conclusion: its diversity and

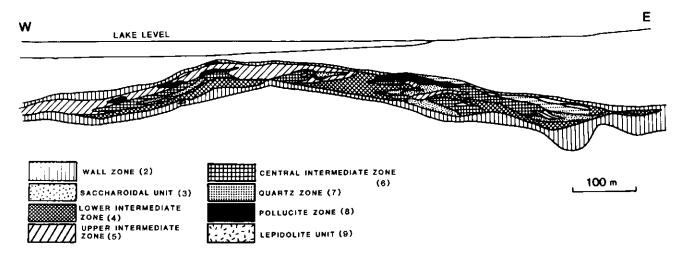


Figure 5 Vertical longitudinal section through the Tanco pegmatite, Bernic Lake, Manitoba (modified from Černý, 1989a). Intermittent border unit 1 is not mappable at this scale. Zones 4 and 5 carry the bulk of spodumene and petalite, whereas 3, 6 and 9 contain the main Nb, Ta, Sn and Be mineralization.

its modifications. The homogeneous, zoned and layered cases are a kind of end-member, each showing extensive variability and, locally, patterns of intermediate character. Also, the primary structure and textures of individual units are occasionally found deformed by post-consolidation tectonic activity. In some cases, thorough metamorphism (Grew, 1981; Franz and Morteani, 1984) or mylonitization (Grip, 1940; Eby, 1986) is observed. Rare-metal mineralization may suffer significant adjustment of mineral assemblage and chemistry of individual phases (Franz and Morteani, 1984; Černý et al., 1991).

#### Mineralogy

The fundamental rock-forming assemblage of rare-element pegmatites is fairly simple: potassic feldspar, albitic plagioclase, quartz and subordinate, if any, muscovite and/or biotite. Most barren pegmatites of the rareelement class do not carry any additional species. However, the paragenesis of a single highly fractionated pegmatite of one of the complex subtypes may exceed a hundred minerals. The diversity is enhanced by phases with rare alkalis and volatile components (Li, Rb, Cs; H<sub>2</sub>O, F<sub>2</sub>, B<sub>2</sub>O<sub>3</sub>, P<sub>2</sub>O<sub>5</sub>) and by a multitude of accessory minerals of lithophile rare elements. Comprehensive lists and characteristics of phases representative of the whole spectrum of pegmatite mineralogy can be found in Černý (1982c), with updates in Černý and Ercit (1985, 1989).

In terms of systematic mineral classes, the silicates, phosphates and oxides dominate over others. Silicates compose the bulk of rare-element pegmatites, and are also the exclusive ore minerals of Li, Rb, Cs, Be, Zr and Hf. Peraluminous silicates, such as muscovite, garnet, tourmaline or aluminosilicates, are typical of pegmatites with bulk Al<sub>2</sub>O<sub>3</sub>/(CaO+Na<sub>2</sub>O+K<sub>2</sub>O)>1, but are scarce to missing in pegmatites of metaluminous chemistry. The number of primary phosphates of Ca, Na, Li, Fe, Mn and Mg in LCT pegmatites is rather restricted (although their abundance may be spectacular), but

generates scores of secondary phases. In contrast, NYF pegmatites are extremely poor in phosphates. The high-valency cations of rare metals (Nb, Ta, Ti, Sn, U, Th) typically form oxide minerals which also include representatives of the hematite, spinel and chrysoberyl groups.

In general, most of the ore minerals of Li, Rb and Cs are found in the primary and, particularly, in the intermediate and central zones. Early generations of Be, Nb, Ta and Ti minerals also occur there, but are mainly concentrated in late units, together with Sn, Zr, Hf, Sc, U, Th, Y, and REE-bearing phases. Albitic, muscovitic and lepidolite units are their main carriers.

For some rare elements such as Nb and Ta, the diversity of their mineralogy increases in more fractionated pegmatites (Černý and Ercit, 1985, 1989), whereas others precipitate in a single primary species, e.g., Be and Cs. Another typical feature of pegmatite mineralogy is the recurrence of many species at different stages of pegmatite crystallization. Multigeneration species such as beryl, columbite-tantalite, tourmaline or microlite display striking changes in physical properties and chemistry, which, in turn, facilitates exploration and evaluation, and aids in petrologic study, but also requires close control in ore processing.

#### Geochemistry

Abundances of many chemical elements are perceptibly to significantly different in the LCT and NYF families. They are grouped below, in the approximate order of decreasing maximum concentrations, in the sequence of the four major categories, as well as within each of them. The following element concentrations are typical of the LCT permatites:

- (1) substantial: O, Si, Al, K, Na, Li;
- (2) subordinate: Rb, Cs, Ca, Fe, Mn, Mg, F. B. P:
- (3) minor: Be, Sn, Ba, Sr, Nb, Ta, Ti, Zr, Hf;
- (4) trace: Y, REE, Ga, W, Sc, C, S, Zn, As, Cu, Pb, Cd, Tl, Ge, Mo, Sb, Bi.

In the NYF pegmatites, the abundances of rare alkali metals decrease, but the roles of rare earth elements (REE) and high field strength elements (HFSE) increase:

- (1) substantial: O, Si, Al, K, Na;
- (2) subordinate: Ca, Fe, Mg, F, Rb, Ba, Li;
- (3) minor: Sr, Mn, Y, REE, Cl, Be, Ba, Ti, Th, U, Zr, Nb, Ta, B, P, Cs, Hf;
- (4) trace: Sc, W, Zn, Ga, C, S, Mo, As, Bi.

The overall predominance of lithophile elements is evident. Semimetals and chalcophile transition metals are relegated to a negligible role. Because of extensive depletion of sulphur, some strongly chalcophile elements are largely restricted to oxidic compounds or native state (e.g., Zn, Tl, Ge, Sb. Bi). Among the lithophile elements, both "fluorophile" (Li, Rb, Cs, Tl, Sn, Be, Mn, Nb, Ta, Y, W, Th, U, HREE (heavy REE)) and "chlorophile" elements (Nb, Y, REE, Fe, Ti, Zn, Zr, Hf) are represented, as classified by Hildreth (1981), Christiansen et al. (1984) and others. Most members of the first group correspond to the LCT elements, whereas the second is in good agreement with the NYF signature.

The bulk composition of rare-element pegmatites is close to that of granite. The main deviations are low total Fe, MgO and CaO, variable  $\rm K_2O/Na_2O$ , and high  $\rm Al_2O_3$  in the peraluminous types (Table 5). In the LCT family, gradual build-up of Li, Rb, Cs, Tl, Ta, Hf, Ga, Ge, B, F and P is typical from the beryl type to the most fractionated representatives of the complex subtypes. Li\_2O may attain 0.7-2.0 wt.% in spodumene- or petaliterich bodies; Rb\_2O, Cs\_2O, B\_2O\_3, P\_2O\_5 and F\_2 are the only other constituents that may be found in concentrations above 0.1 wt.%.

Although the subaluminous to metaluminous character of the NYF-family, rareearth-type pegmatites can be safely estimated from their mineralogy, quantitative analyses of these bodies are not yet available.

Within individual bodies of highly evolved pegmatites, gradual changes in the composition of subsequent zones and replacement bodies are best expressed by the se-

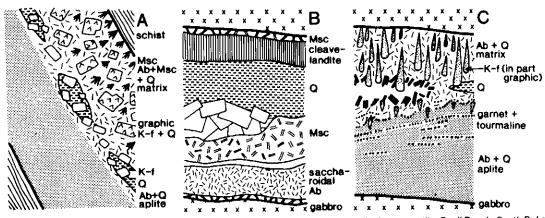


Figure 6 Vertical sections across three representatives of layered pegmatites. (A) A barren steeply dipping pegmatite, Buell Ranch, South Dakota (after Orville, 1960); (B) An unnamed, subhorizontal beryl-bearing pegmatite (after Solodov, 1971); (C) A vug-bearing, subhorizontal pegmatite of the complex lepidolite subtype, Himalaya district, California (after Foord, 1976 and Jahns, 1982).

quence of main alkali metals. The primary zonal crystallization goes from Na(K,Ca) in border and wall zones, through K,Na in intermediate zones, to K, Li(Na,Cs) in core-margins and core. This zonal pattern is interspersed with, and terminated by, Na-rich albitic units, K-bearing and F-enriched greisen-like assemblages and, rarely developed, late K-feldspar, and by Li(Rb,Cs)-rich lepidolite units (modified from Ginsburg, 1960).

In general, fractionation throughout rare-element pegmatite sequences extends the classic trends observed in plutonic granites (Table 6). It reaches extremely low values in element ratios, e.g., K/Rb (1.9 in K-feldspar, 1.4 in micas), K/Tl (320 in lepidolite), Rb/Tl (60 in pollucite), Ba/Rb (<0.000X in K-feldspar), Sr/Rb (0.000X in K-feldspar), Rb/Cs (6.5 in K-feldspar, 4.4 in micas), Nb/Ta (<0.005 in microlite and simpsonite), Th/U (0.02 in microlite), Zr/Hf (0.014 in hafnon), Al/Ga (239 in micas), Si/Ge (540 in topaz) and Zn/Cd (0.7 in hawleyite and černýite) (Černý et al., 1985a,b; Makagon and Shmakin, 1988).

Extensive fractionation is also typical within individual pegmatite bodies. For ex-

ample, K/Rb in K-feldspar is known to vary from 380 to 51 between border zone and core-margin in relatively simple pegmatites, and from 14 to 3.6 in complex bodies. Changes from about 90 to 20 are observed even in single K-feldspar crystals (Jahns, 1982). Nb/Ta in columbite-tantalite may range from 6-5 in intermediate zones to 0.3-0.2 in albitic units.

Hybridization of rare-element pegmatites by reaction with wall rocks of contrasting composition (metapelites, marbles, amphibolites, serpentinites) is very limited. It is usually confined to only the outermost units, which show increased contents of Ca, Mg, Fe. Al or Ti. In contrast, pegmatite exomorphism is commonly extensive in such environments. Pegmatites intruding serpentinites are commonly bordered by anthophyllite, talc, phlogopite and actinolite, indicative of redistribution of alkalis, Ca, Mg, Fe, Si, F and H<sub>2</sub>O along the contacts. In amphibolites, outflow of K, Na, Li, Rb, Cs, B, F and H<sub>2</sub>O is recorded in haloes of albite, rare-alkali biotite to phiogopite, muscovite, tourmaline and holmquistite (Hawthorne and Černý, 1982; Shearer et al., 1984; London, 1986c; Morgan and London, 1987).

	Table 5	Table 5 Bulk composition of rare-element pegmatites.				
	1 "Southern USSR"	2 Siberia USSR	3 Harding NM	4 Tanco MB	5 Pidlite NM	6 INCO MB
SiO <sub>2</sub>	72.82	70.62	75.24	69.74	74.5	73.70
TiO <sub>2</sub>	0.02	0.04	0.05	0.01	-	0.01
$Al_2O_3$	14.00	17.69	14.42	16.50	14.8	16.53
Fe <sub>2</sub> O <sub>3</sub> FeO	0.14 1.00	0.60 0.20	0.65*	0.18*	_	0.18 0.08
MnO	0.06	0.03	0.18	0.21	-	0.16
MgO	0.30	0.28	0.01	_	-	0.05
CaO	0.40	0.65	0.20	0.89	0.2	0.13
Li <sub>2</sub> O	0.10	1.05	0.65	1.18	0.7	1.41
Na₂O	2.30	4.84	4.23	2.69	3.3	3.78
K₂Ō	7.40	1.95	2.74	4.42	5.4	1.73
Rb₂O	0.18	0.21	0.19	1.10	_	0.36
Cs <sub>2</sub> O	0.005	1.31	0.05	0.42	_	0.03
$P_2\bar{O}_5$	0.28	0.90	0.13	1.18	-	< 0.01
$B_2O_3$	0.002	-	_	0.24	_	
F	0.13	0.65	0.64	0.20	0.9	-
H <sub>2</sub> O+	0.58	0.75	_	_	0.6	0.10
total	99.72	101.77	99.38	98.96	100.4	98.33
F=O <sub>2</sub>	-0.05	-0.28	-0.27	0.08	-0.4	_
Σ	99.67	101.49	99.11	98.88	100.00	98.33

#### Notes

- \* total Fe as FeC
- 1: Beryl-type pegmatite with minor Li-micas (Melentyev et al., 1971)
- 2: Spodumene-subtype of complex pegmatite with pollucite (Filippova, 1971)
- 3: Spodumene-subtype of complex pegmatite (Burnham and Jahns, 1962)
- Petalite-subtype of complex pegmatite with abundant lepidolite, amblygonite and pollucite (London, 1990)
- 5: Lepidolite-subtype of complex pegmatite (Jahns, 1953b)
- 6: Albite-spodumene type of pegmatite (Chackowsky, 1987)

#### **REE Abundances**

The geochemistry of rare-earth elements in granitic pegmatites has not been studied to an extent permitting generalization, and most of the few studies available concern individual minerals rather than bulk compositions. It seems certain, however, that, in LCT pegmatites, the REE content of most primary zones is very low, equal to or less than chondritic, and stays at this level even in late, almost secondary units. In contrast, dramatic increase in Y and REEs is shown in late units, which carry most of the accessory minerals in the NYF pegmatites (Simmons et al., 1987). The pioneering studies of REE abundances in rock-forming and accessory minerals of the Black Hills pegmatites seem to suggest that these elements may become useful as indicators of parent environments, sequence of crystallization, and rare-element migration during consolidation of pegmatite intrusions (Walker et al., 1986b; Jolliff et al., 1987). However, much more work is required on diverse pegmatite types, before the behavior of REEs is understood (cf. Register and Brookins, 1978; Register, 1979). isotopic Data.

Radiogenic isotope systems in granitic pegmatites are notorious for being disturbed, particularly at geologically old localities. K-Ar data are rarely significant, except for some of the most recent intrusions (such as the gem-bearing localities in California; Taylor et al., 1979). The few Rb-Sr studies performed to date invariably show a loss of radiogenic 87Sr. This indicates post-consolidation mobility of 87Sr, which creates problems in the dating and interpretation of initial 87Sr/86Sr ratios, particularly for Archean and Early Proterozoic pegmatites. Abnormally high initial ratios and unrealistically low ages are the rule rather than exception (as reviewed by Clark, 1982; cf. also Walker et al., 1986a,b; Clark and Černý, 1987). Disturbances of crystal/melt fractionation patterns also plague Sm-Nd isotopic ratios (Walker, 1984; Walker et al., 1986a,b).

Stable isotope studies on pegmatites and their minerals are also limited, especially for carbon, sulphur, silicon and boron. Only D/H and 18O/16O data have been systematically collected at several localities. Taylor (1968) indicated a range in δ18O for a number of pegmatites between +5.5% and +11.0%; Taylor et al. (1979) and Taylor and Friedrichsen. (1983a) found similar broad variations in δ18O for separate minerals from another series of localities. Open-system behaviour and exchange with host rocks are indicated in many cases, as well as low-temperature exchange among pegmatite minerals; both hinder petrogenetic interpretations and geothermometry (cf. review by Longstaffe, 1982). A complex pattern of re-equilibration was demonstrated by some studies, e.g., Taylor and Friedrichsen (1983b). Nevertheless, some pegmatites were apparently

shielded from external contamination and/or did not suffer substantial re-equilibration: the Tanco and Harding deposits are hosted by gabbroic rocks, but have  $\delta^{18}O$  of quartz from +9.0% to +9.7% and +9.9% to +11.0%, respectively (Taylor and Friedrichsen, 1983a), and the Tin Mountain pegmatite shows trustworthy estimates of temperature from  $\Delta^{18}O$  Qtz-Msc versus Qtz-Fsp (Walker et al., 1986b). In all three of these pegmatites, fractionation progresses from the border zones inward, and temperature estimates decrease in this direction.

#### Fluid Inclusions

Pegmatite minerals have been rewarding subjects of fluid inclusion research, especially during the last two decades when experimental techniques, sample selection and interpretations reached reliable maturity (e.g., Roedder, 1972, 1981a,b, 1984; London, 1986a,b). Extensive diversity of chemical and phase compositions of fluid inclusions is the earmark of granitic pegmatites, not only among different pegmatite types, but also in different units of individual bodies.

Inclusions of what originally was a hydrous silicate melt are common in simple pegmatites and in the outer zones of complex deposits. In the NYF pegmatites, inclusions of a "hydrosaline melt" are found in the internal zones, albitic units and crystals from miarolitic cavities, with up to 42 wt.% dissolved solids in the originally homogeneous fluids

(Roedder, 1981a,b). In contrast, LCT pegmatites contain fluid inclusions with much more dilute solutions (4-6, and locally up to 20, wt.% of dissolved solids being common). The content of  $\rm CO_2$  tends to increase in inclusions in minerals formed during late replacement processes. Daughter crystals found in inclusions within these metasomatic and vug-filling minerals commonly consist of water-soluble salts of Li, Na, K, Cs, B, F and Cl.

So far, only two research efforts are generating data that contribute to understanding the course of pegmatite consolidation. Thus, no generalization is possible at present, all the more since both programs are aimed at a single locality (the Tanco pegmatite), and have produced diverse results and divergent interpretations. Thomas and co-workers established early mixing of pegmatitic and metamorphic fluids along the contacts of the Tanco deposit with host amphibolite, and worked out the thermal course of Tanco crystallization from 720°C to a surprisingly low 260°C (Thomas and Spooner, 1988a,b; Thomas et al., 1988, 1990). They claim that co-existing melt and fluid(s) were present throughout this temperature range. In contrast, London and associates tracked the Tanco consolidation path from the beginning of petalite crystallization at about 620°C/3.0kb to exhaustion of melt at 400°C/2.5kb, and subsequent crystallization plus replacements from fluids of progressively increasing salinity, in part mixed with metamorphic fluids (London, 1986a, 1990; London et al., 1987). London's claims of crystallization from homogeneous melt through most of the consolidation of complex pegmatites is supported by his experimental work, which is discussed later. London (1985) also cautions about the problems encountered in distinguishing between primary and secondary inclusions in quartz.

It is evident that, valuable as the present and potential future insight from fluid inclusions is, this line of research is still in its infancy. Examination of a spectrum of different pegmatite types is required to broaden the scope of the present work, which is aimed at highly fractionated pegmatites (London, 1986b; Shigley et al., 1986; Taylor et al., 1979).

#### MODELS OF INTERNAL PEGMATITE CONSOLIDATION Diversity of Hypotheses

Historically, a multitude of hypotheses have been offered to explain the nature of the phase(s) from which granitic pegmatites crystallize, and to unravel the course of their crystallization. This is not surprising, considering the variety of rock types commonly designated as granitic pegmatites and the diversity of their environments, as illustrated in Figure 1 and Tables 2 to 4. According to the

	Granites		Rare-element Pegmatites <sup>3</sup>					
	high-Ca granites <sup>1</sup>	fine-grained peraluminous leuco- granites <sup>2</sup>	rare-earth type	beryl type	complex type <sup>4</sup>	lepidolite subtype	albite- spodumene type	Red Cross Lake <sup>5</sup>
Li	24	81	19-209	19-622	37-8400	93-4640	5110-10310	1540-9760
Rb	110	305	92-183	101-1065	183-9970	274-1865	1737-5490	6220-14150
Cs	2	8	-12	4-132	9-9400	28-236	104-793	1040-26980
Be	2	4	6-101	4-494	3-605	65-440	97-180	113-294
Ga	17	38		-	45-97	_	-177	120-190
Sn .	1.5	9	70-800	13-536	12-3170	63-1000	89-894	25-100
Nb	20	24	53-1280	8-260	8-213	25-155	44-150	17-36
Та	3.6	4.5	9-710	2-204	12-4620	9-346	37-108	244-802
K/Rb	229	159	130-28	120-40	40-5	24-6	80-3.4	2.6-1.6
K/Cs	12600	11000	10500-4650	1800-900	360-6	186-12	360-22	16-0.7
Rb/Cs	55	57.9	32-12	~18	8.5-1.2	13-7	24-6.5	6-0.6
Al/Ga	4820	2080	_	_	1050-600	_	1300-305	750-500
Nb/Ta	5.6	5.0	6-1.7	3.3-1.4	1.1-0.3	2.5-0.10	2-0.4	0.074-0.024

#### Notes

- from Turekian and Wedepohl (1961)
- 2 from Černý and Meintzer (1988)
- modified from Kuzmenko (1976, table 124), Solodov (1971, tables 40 and 41), Stewart (1978), Chackowsky (1987), Göd (1989) and unpublished data of the author
- 4 spodumene, petalite and amblygonite subtypes
- ranges of data from channel samples across diverse dykes of a swarm rich in lepidolite and spodumene + quartz pseudomorphs after petalite (Jambor and Potter, 1967; Černý et al., 1985a; Wang et al., 1988)

meticulously documented review by Jahns (1955), proposals for the parent medium have included: (1) silicate melts, (2) silicate melts enriched in volatile components, (3) pneumatolytic agents, better defined today as supercritical fluids, and (4) hydrothermal solutions ± gas phases.

As to the emplacement and consolidation process, four principal ideas have been forwarded: (1) crystallization in an essentially closed chamber in a closed or restricted system, (2) deposition from fluids flowing through open-system channelways, (3) recrystallization and/or metasomatism of non-pegmatitic precursors, and (4) combinations of one or another of the above mechanisms with metasomatic replacement by fluids generated within the pegmatite or introduced from an external source.

The study of rare-element pegmatites has contributed extensively to this spectrum of fluids, processes and hypotheses because of the complexity of these deposits, which is unmatched in other pegmatite classes. However, improved mapping and field descriptions, advances in experimental petrology, and detailed paragenetic and geochemical data today constrain the choices much more tightly than even two to three decades ago. Thus, it is not surprising that a general convergence of ideas is in progress, favouring and improving one of the classic views of pegmatite consolidation: that the crystallization proceeds, under decreasing temperature (and variable pressure) and conditions of restricted exchange with wall rock in a volatile-rich, highly hydrous granitic melt, complemented by (or grading into) precipitation from an aqueous fluid. Extensive interaction among the solids, melt and fluid(s) at different stages of consolidation, and particularly toward its termination, is characteristic. Observations supporting such a modified igneous model are reviewed in the following paragraphs.

#### The Igneous Model

**Bulk composition.** Geochemically primitive rare-element pegmatites of the rare-earth and beryl types are remarkably constant in bulk composition (Table 5). They are very close to the thermal minima and eutectics in the Ab-Or-Qtz-H<sub>2</sub>O system (Tuttle and Bowen, 1958; Luth *et al.*, 1964). This relationship holds for homogeneous as well as zoned and layered pegmatites (Jahns and Tuttle, 1963) (Figure 7).

Many albite-spodumene pegmatites and some quasi-homogeneous petalite-bearing bodies contain Li<sub>2</sub>O between 1.5 and 2.0 wt.%; these contents are also typical of internal lithium-rich units of zoned complex pegmatites. These compositions coincide with the thermal valley established experimentally for the Ab-Ecr-Qtz system (Stewart, 1963, 1978) (Figure 7). Note that the compositions are virtually identical for both petalite- and spodumene-bearing pegmatites (Table 5), indicating that the striking difference in lithium mineralogy is a consequence of different P-T conditions rather than magma chemistry (London, 1984) (Figure 8).

Meaningful statistics about the bulk compositions of the lepidolite subtype of complex pegmatites are not available. However, the available compositions and the albite-rich, K-feldspar- and quartz-poor modes of many others indicate close correspondence with the Na-enriched, K-depleted thermal valley in F- and B-bearing haplogranite systems (Manning and Henderson, 1981; Manning and Pichavant, 1985) (Figure 7).

The remarkable difference in lithium mineralogy between the spodumene- or petalite-bearing and lepidolite-rich pegmatites is caused by a (potentially subtle) shift to higher  $\mu$ LIF and  $\mu$ KF, at relatively low acidity. A lower  $\mu$ KF/ $\mu$ HF stabilizes topaz, which is found in significant quantities in some lepidolite pegmatites (Figure 8; London, 1982).

Chemical modelling also indicates that increased  $\mu(PFO_2)$  generates amblygonite + quartz at the expense of Li-aluminosilicates, or amblygonite + albite if  $\mu(NaLi_{,1})$  increases as well (Burt and London, 1982; London and Burt, 1982b) (Figure 8). This explains the relationship between spodumene- or petalite-bearing pegmatites and those of the amblygonite subtype.

The albite type could not be reasonably correlated with any melt composition until recently. As discussed in more detail under "Zonal structure and experimental studies", London et al. (1989) generated residual sodic melts in complex Li, B, F, P, H<sub>2</sub>O-rich systems: the albite pegmatites, generally characterized by low volumes, may possibly crystallize from such melts that escaped from a parent pegmatitic reservoir and solid-ified separately.

Thus, the above experimental work and chemical models satisfactorily explain rareelement pegmatites as products of crystallization from residual granitic melts, with a degree of uncertainty remaining with the albite type.

Temperature and pressure of consolidation. Temperature ranges, bracketed by fluid inclusions, two-feldspar thermometry and other indicators, are very broad, about 750°C to 150°C. Temperatures generally decrease from the margins to the internal zones and secondary units, in accordance

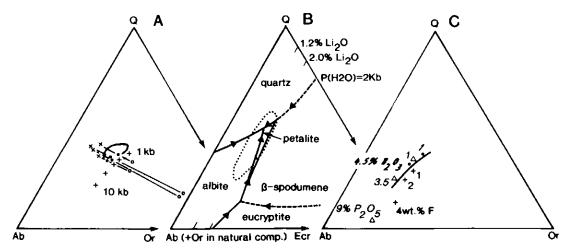


Figure 7 (A) Composition of 21 zoned peraluminous pegmatites (elliptical field) and 4 peraluminous layered pegmatites ( $\mathbf{x}$ , aplite;  $\mathbf{o}$ , pegmatite;  $\mathbf{v}$ , bulk) in the Ab-Or-Qtz diagram; minima and eutectics at 1, 2, 3, 5 and 10 kb  $P(H_2O)$  are indicated (for metaluminous haplogranite, after Luth et al., 1964 and Jahns and Tuttle, 1963); ( $\mathbf{B}$ ) Liquidus surface of the Ab-Qtz-Ecr- $H_2O$  system at  $P(H_2O) = 2$  kb, and the compositional field of pegmatites (or their zones) rich in spodumene or petalite (dotted outline; after Stewart, 1978); ( $\mathbf{C}$ ) Shift of ternary, metaluminous Ab-Or-Qtz minima with increasing content of  $\mathbf{F}$  and  $\mathbf{B}$  (at  $P_{H_2O}$  of 1 kb) and P (at  $P_{H_2O}$  of 2 kb); the fluorine-free 1 to 10 kb trend of Figure 7A is shown for comparison as a solid line. The effect of  $\mathbf{F}$  (crosses; Manning and Henderson, 1981) is realistic for metaluminous NYF-pegmatite magmas, whereas the effects of  $\mathbf{B}$  (dots; Manning and Pichavant, 1985) and P (triangles; London et al., 1990 and  $\mathbf{D}$ . London, pers. comm. 1991) would be much more meaningful if tested in peraluminous bulk compositions.

with textural evidence for sequence of crystallization. Even the temperatures obtained from primary assemblages alone, 750°C to about 450°-400°C (e.g., London, 1986a; Stern et al., 1986; Walker et al., 1986b; Chakoumakos and Lumpkin, 1990) extend below the range of experimental solidification of haplogranite. A better match is found with experimental results on hydrous haplogranite in the presence of Li, Cs, B, P and/or F, all of which depress the liquidus and solidus (Chorlton and Martin, 1978; Wyllie, 1979; Pichavant, 1981; Henderson and Martin, 1985). Confirmation was found in experiments with a composite sample of the Li-rich Harding pegmatite (Burnham and Jahns, 1962; Jahns, 1982) and with the highly fractionated Macusani glass (London et al., 1988, 1989).

Pressures of crystallization are not easy to gauge quantitatively. Fluid inclusion studies suggest a range of 4 to 1.5 kb, and occurrences of andalusite and cordierite also indicate that pressures must have been moderate, generally equivalent to the pressure regime of the enclosing schists in (uppergreenschist to) lower-amphibolite facies of Abukuma-type series.

The P-T ranges of most Li-rich pegmatites are constrained by experimental refinement of the Li-aluminosilicate stabilities (London, 1984) (Figures 8 and 9). The high-P/low-T character of spodumene and the low-P/high-T character of petalite correspond to the geological environments of their occurrences, and the common occurrence of "squi" (spodumene + quartz intergrowths) pseudomorphic after petalite is also explained by the phase diagram.

In accord with the above lines of evidence, compositional relationships in feldspar crystallization also favour precipitation from cooling melts, from border zones inward, but data are available at present only for geochemically primitive pegmatites (Černý et al., 1984). Documentation is virtually non-existent for more fractionated categories.

Cooling rates are of considerable interest, particularly for the most evolved pegmatites emplaced far away from their sources, in medium- to low-grade rocks that passed through their peak metamorphic conditions. Differences between the temperatures of a pegmatite melt and its host rock may be as high as 100°-300°C, and cooling plus crystallization of the melt may be as short as

100-200 years (e.g., London, 1986a, 1991b; Chakoumakos and Lumpkin, 1990).

Zonal structure in experimental studies. Internal compositional and textural heterogeneity of zoned and layered pegmatites was one of the pillars of arguments against a magmatic nature of pegmatite crystallization. Cotectic crystallization of two feldspars and quartz was considered an inevitable termination in residual granitic melts; this is obviously not the case in most pegmatite varieties. However, it was suspected since the early studies of Niggli and Fersman that most pegmatite melts are much more complex in composition and, consequently, in their style of consolidation, than simple haplogranite systems. Two schools of experiment and interpretation have developed since the 1950s that warrant consideration: the classic concept of Jahns and Burnham claiming largely volatile (H2O)saturated conditions for most of pegmatite crystallization, and the recent model of London, arguing for volatile-rich, but undersaturated, conditions until the very late stages of consolidation.

Burnham and Jahns (1962), Jahns and Burnham (1969), Jahns (1982) and, in a recent extension by equilibrium thermodynamic modelling, Burnham and Nekvasil (1986) concluded from experiments on Spruce Pine and Harding pegmatite compositions that:

(1) most of pegmatite crystallization proceeds from volatile-saturated melts, in the presence of co-existing aqueous fluid phase;

(2) the first appearance of coarse-grained, truly pegmatitic texture marks the onset of

(3) the fluid phase enhances mobility of many melt components, facilitates separation of K from Na if substantial CI is present, and may also be responsible for much precipitation of core quartz:

exsolution of the aqueous fluid;

(4) segregation of individual minerals in the innermost parts of pegmatites is responsible

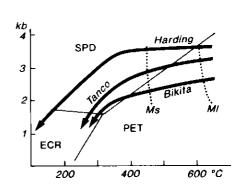


Figure 9 P-T paths of crystallization of Li-rich pegmatites: Harding, New Mexico with dominant spodumene (after Chakoumakos and Lumpkin, 1990); Tanco, Manitoba with substantial proportions of petalite and primary spodumene (after London, 1986a); and Bikita, Zimbabwe with petalite dominant over spodumene (after London's, 1986a, 1990 estimate).

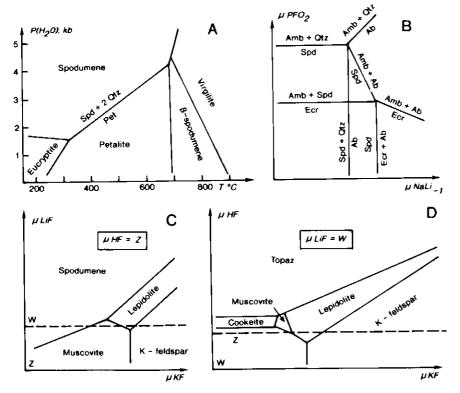


Figure 8 (A) Phase diagram for the bulk composition 20 Ecr 80 Qtz (mole %) in the system LiAlSiO<sub>4</sub>-SiO<sub>2</sub>  $H_2O$  (modified from London, 1984); (B) Schematic isobaric-isothermal  $\mu$ PFO<sub>2</sub> $\mu$ NaLi<sub>-1</sub> phase diagram in the system LiAlO<sub>2</sub>-SiO<sub>2</sub>-PFO<sub>2</sub>-NaLi<sub>-1</sub>, under quartz- and  $H_2O$ -saturated conditions within the stability field of spodumene and all other associated phases (modified from Burt and London, 1982; London and Burt, 1982b); note amblygonite is the only stable Li-phase at high  $\mu$ FO<sub>2</sub>, typical of pegmatites of the amblygonite subtype. (C,D) Schematic isobaric-isothermal  $\mu$ HF- $\mu$ LiF- $\mu$ LiF-phase diagram in the system LiAlO<sub>2</sub>-SiO<sub>2</sub>- $H_2O$ -HF-LiF-KF, under the same conditions as B; dashed lines mark the two sections. W and Z, through the  $\mu$ HF- $\mu$ KF- $\mu$ LiF space (modified from London, 1982), note the effect of increasing chemical potentials of LiF and HF, responsible for generating complex pegmatites of the lepidolite subtype, locally with topaz.

for the apparent deviations of the last crystallization products from the expected magmatic minima and/or eutectics (Figure 10).

The above model was generally accepted during the 1970s and 1980s, but some of its aspects were increasingly questioned. It did not conclusively deal with units considered metasomatic by most researchers; it did not account for volume relationships required by extensive crystallization from aqueous fluid; the extremely low CI contents of fluid inclusions in primary zones argued against any significant role of this halogenide in distribution of cations; and interpretation of the only significant experiment which successfully simulated most attributes of pegmatite crystallization was doubted as to the co-existence of melt and fluid.

The synthetic simulation of pegmatite crystallization performed by London et al. (1988, 1989; London, 1990) evolved from London's studies of fluid inclusions (London, 1986a,b, 1987; London et al., 1987). These studies indicated that melts from which highly fractionated pegmatites crystallized, such as Harding or Tanco, must have been more complex and particularly volatile-enriched than the compositions of solidified pegmatites + H<sub>2</sub>O used by Jahns and Burnham. London's experiments were performed on the Macusani glass, which comes much closer to a parent melt than any averaged holocrystalline pegmatite, with or without additional percentages of some of the volatiles.

Experiments conducted on H<sub>2</sub>O-oversaturated melts gave results inconsistent with features of natural pegmatites:

- the products were homogeneous in grain size, phase distribution, and grain chemistry and contained interstitial vapour spaces;
- (2) vapour/melt partition coefficients for minor elements varied with temperature, but there was no particular fractionation and accumulation observed in the course of crystallization;
- (3) a total of 22 elements typical of rareelement pegmatites preferred melt to vapour. Boron was the only exception, and was separated from metals with which it is associated in natural assemblages;
- (4) crystallization of melts occurred only in experiments with  $\leq 3.5$  wt.% added  $H_2O$ ; above this limit, melt phase persisted metastably even in subliquidus experiments of as much as 3,200 h duration; crystallization proceeded only by condensation from vapour.

In contrast to the above, close correspondence between natural pegmatites and experimental products occurred in largely to totally H<sub>2</sub>O-undersaturated runs:

- (1) combined effects of Li, F, B and P on solubility of water boost the H<sub>2</sub>O saturation of original bulk melt to 11.5 wt.%, and to 15-20 wt.% after 75-85% crystallization from H<sub>2</sub>O-undersaturated starting condition;
- (2) crystal size increases by two orders of magnitude from margin to centre of charges, and fabrics progress from graphic inter-

growths and radial clusters to large, euhedral crystals:

- (3) mineralogy grades from graphic Na-feldspar + quartz at the margin of the charges, through increasingly K-feldspar-enriched fsp+qtz assemblages, into blocky K-feldspar at residual melt pools containing Li, Alstuffed quartz or virgilite and lithium micas in the core:
- (4) solubilities of lithophile rare-element cations in aqueous fluids, exsolved in late stages of some runs, are low relative to those in the melt phase;
- (5) these cations plus B, F and P are increasingly concentrated into residual sodic SiO<sub>2</sub>-depleted melts, which tend to crystallize as fine-grained albite + mica aggregates (± water-soluble quenched glass) in the central parts of experimental charges (Figures 10 and 11).

Thus, the vapour-undersaturated crystallization of macusanite generates micro-scale replicas of zoned pegmatites in terms of textures and mineral assemblages, including K-feldspar + quartz core-margins and quartz + Li-aluminosilicate cores, through a purely magmatic crystallization. At its conclusion, this process produces fine-grained albite and/or mica aggregates with considerably increased contents of incompatible rare elements. The underlying process is one of disequilibrium growth of feldspars and quartz through supersaturation, caused by increasing concentrations of  $\rm H_2O$ , Li, B, F and P in melt. Supersaturation stems from an

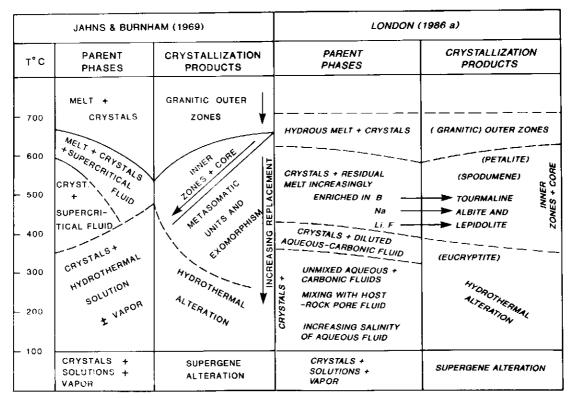


Figure 10 Parent phases and crystallization products in rare-element pegmatites. The general model of Jahns and Burnham (1969) in approximate correlation with temperature, compared with London's (1986a, 1990) model for complex pegmatites, derived from fluid inclusion study and experimental work.

increasing disruption of the alkali-aluminosilicate framework of melt by the above fluxing components, rather than by significant liquidus undercooling. These components also reduce the density of crystal nucleation, increasingly so with their gradual accumulation during crystallization of the melt. Inhibited nucleation promotes the growth of giant crystals, particularly in the intermediate and central zones.

Components such as B, P and F are not requisite, but augment zoning by expanding the liquidus field for quartz. The activity of albite appears to be more depressed than that of K-bearing phases, so that addition of B or F expands the liquidus field of K-feldspar (core-margin blocky microcline) and eventually muscovite (book mica commonly found in a similar position). Consequently, increasing H2O, B, F and P drives the composition of the residual melt to silica-depleted, Na-rich alkaline compositions enriched in incompatible elements that eventually crystallize as late-stage albite ± mica or Li-mica units with prominent rare-metal mineralization. Consolidation of these units is triggered by H<sub>2</sub>O oversaturation and exsolution of aqueous fluid from residual melts, combined with stabilization of tourmaline. Removal of the fluxing components, B, H<sub>2</sub>O and, to a lesser extent, Li, F and P, increases the solidus temperature and leads to dumping of albite, micas and rare-element minerals in fine-grained aggregates.

London's model explains phenomena that are not accommodated in the Jahns-Burnham concept. "Metasomatic" units are documented as primary magmatic; the role of exsolved aqueous fluids is found to be very restricted in volume and time, with no space problem associated; no CI is required for alkali separation in space and time, which is achieved by the effects of B, F and P disequilibrium crystallization and deviation from haplogranite liquidus relationships. It is interesting to note that London's model is in general agreement with the largely intuitive ideas of Jahns (1953a, p. 581, 596-597), from which this author was later deflected by early experimental work based on the mistaken assumption of chemical equilibrium.

Although originally developed from the studies of some of the most fractionated LCT pegmatites, highly enriched in liquidus- and solidus-depressing cations and volatile components, London's (1990, 1991a,b) presentday model is general enough to encompass all categories of pegmatites within the muscovite, rare-element and miarolitic classes. Any individual component, or combination of H<sub>2</sub>O, B, P and F (with or without Li, Cs, Be) produces essentially the same range of qualitative effects on the style of consolidation of pegmatite melts. Increasing the number and concentration of the above components enhances the intensity of these effects.

Nevertheless, some aspects of pegmatite crystallization still defy attempts at petrologic interpretation. Among the complex pegmatites, those with layered internal structure are not totally reconcilable with the above model, as are other varieties with early sodic aplite units. Continued study of these pegmatite types is evidently warranted to complete the model.

Metasomatic and "metasomatic" assemblages. As shown above, sizeable albitic (saccharoidal or cleavelandite) and micaceous (muscovite to lepidolite) units can be explained by late, but primary, crystallization from residual melt. Pollucite units, also considered metasomatic by some investigators, were shown to be near solidus phases by Henderson and Manning (1984) and Henderson and Martin (1985). Vigorous as the metasomatic replacements commonly are along boundaries of the albitic and micaceous units with earlier zones, the bulk of these units could have crystallized from residual melts in situ, alleviating the fluid, space and material transfer problems inherent to entirely metasomatic concepts. However, London (1990) does not exclude the possibility that some extensive units of lithian micas may be metasomatic after assemblages with abundant K-feldspar and/or lithium aluminosilicates.

Most unequivocally established replacements are observed only at very localized scales, and generally reflect widespread disequilibrium between solid phases and late fluids (London and Burt, 1982b; London, 1991a). Metasomatic alteration of primary silicates, phosphates and oxide minerals commonly produces complex assemblages involving hundreds of secondary phases (e.g., Černý and Ercit, 1985, 1989). Early reactions are triggered by alkaline fluids that evolve into neutral and weakly acidic conditions, generating mica + quartz assemblages. Overall, we may conclude that most

metasomatic phenomena result in local modifications of pre-existing mineral assemblages, rather than extensive secondary units. So far, no detailed modern studies have been conducted on units suspected to be of such an origin, and there is no solid evidence available even at the level of textural observations of mineral reactions.

Fractionation and enrichment in rare elements. Steep fractionation gradients in rock-forming and accessory minerals, and extreme enrichment of lithophile rare elements in pegmatites in general and in their late albitic plus micaceous units in particular, have always been a problem. Whereas the spectacular accumulation of rare elements can be explained by extending the mechanism provided by London's model (preferential partition of these elements into increasingly smaller volumes of residual melts from granitic intrusions all the way to "terminal" sodic melt pockets in pegmatites), fractionation among closely related elements is more difficult to interpret.

In the study of highly evolved granites and rhyolites, this problem was explained by dramatic changes in partition coefficients at the most silicic (>73-74 wt.% SiO<sub>2</sub>) end of granitic fractionation (e.g., Mahood and Hildreth, 1983; Michael, 1983; Miller and Mittlefehldt, 1984). This explanation may extend to the crystallization of pegmatite magmas, combined with mass-controlled differences in diffusion rates (e.g., Černý et al., 1985a). Stabilities of complexes, different between related elements such as Nb-Ta, Zr-Hf, Al-Ga or REEs and among diverse complexes of a single cation such as Be, Sn or Ti, may also contribute to the observed trends (e.g., Beus 1960; Černý et al., 1985a, 1986).

Complexing was particularly favoured by investigators claiming that rare-element mineralization is a hydrothermal and metasomatic event; however, speciation of rare metals in albite- or lepidolite-precipitating melts is

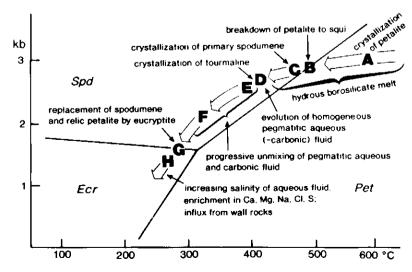


Figure 11 Internal evolution of the Tanco pegmatite, southeastern Manitoba, based on lithium aluminosilicate stabilities and fluid inclusion studies (after London, 1986a, 1990).

still unknown (e.g., London, 1987). Another mechanism, widely advocated as a potent redistributor of rare elements in pegmatite systems, was transport in supercritical fluids co-existing with magma (Jahns, 1982; Černý et al., 1985a). This possibility was effectively eliminated by London's experiments that simulate pegmatite crystallization only in water-undersaturated conditions, and show strong partitioning of rare elements into melt when aqueous fluid does evolve.

Hydrothermal effects. Both Thomas et al. (1988) and London et al. (1989; London, 1990) agree that mixed hydrous + carbonic fluids, unmixing later into separate aqueous and carbonic phases, are active at the last stages of pegmatite consolidation. London's fluid inclusion studies of the Tanco pegmatite place the appearance of this fluid at about 420°C/2.6kb, and its unmixing at only slightly lower P-T values (Figures 9 and 10). In London's model, the action of these fluids is rather restricted, and is expressed mainly in the formation of miarolitic cavities, exomorphic alteration haloes, and internal alteration.

Fluids evolved from residual albitic melts on stabilization of tourmaline (and the loss of other fluxing components) segregate into miarolitic pockets that become lined with crystals of rock-forming and accessory minerals, coated with phases of progressively lower-temperature character (such as cookeite, adularia, zeolites). Many pockets rupture because of the increasing pressure of the evolving vapour, shattering the crystal lining (Foord, 1976; Jahns, 1982). The lower solubility of the main volatile component, H<sub>2</sub>O, in F,B,P,Li-poor pegmatite melts explains the restricted frequency of miarolitic cavities in the geochemically primitive pegmatites of the rare-earth and beryl types, rare as they are in these categories.

Alteration aureoles surrounding rare-element pegmatites are best developed in wall rocks of contrasting chemistry, such as amphibolites. They were documented as essentially greenschist-facies retrogression, generated by gradual release of aqueous fluids and vapours from the pegmatites, by Morgan and London (1987). Černý et al. (1984) and Thomas and Spooner (1988a) documented, among others, exomorphic effects connected with early magmatic stages of pegmatite consolidation. Zagorskyi et al. (1974), Zagorskyi and Khaltuyeva (1976) and Shearer et al. (1984, 1986) stressed the multiplicity of exomorphic stages, spanning a substantial range of pegmatite consolidation.

Understandably enough, the most detailed studies devoted to exomorphic alteration were performed on the geochemically most evolved pegmatites. In the Tanco deposit, Morgan and London (1987) found mineralogical and fluid inclusion evidence that pegmatite fluids, initially rich in Li and B liberated on destabilization of late sodic borosilicate melts, infiltrated host-rock

amphibolites at near-solidus temperatures (≤500°C). Alkali-rich, F-bearing fluids were nearly contemporaneous, and were generated by an F-rich vapour reacting with feldspars. At lower temperatures, the fluids were more dilute and caused an essentially propylitic alteration. After unmixing, the less dense CO<sub>2</sub> fluid tended to rise and generate carbonates mainly above the hanging-wall contacts. Morgan and London (1987) calculated that large quantities of B, F and alkalis were lost by exomorphism. This increased the ASI (aluminum saturation index) of the pegmatite over that of its parent magma. However, the zone of alteration is confined to only a few metres from the contacts, is mainly fracture controlled and consequently is sporadic in distribution and intensity. The process involved only relatively small volumes of solute-rich fluid from the pegmatite.

Near-contact fluid flow is, however, a bidirectional affair. Early inflow of wall-rock components is indicated by tourmaline radiating from contacts inwards (documenting Fe, Mg introduction from outside the pegmatite; London, 1990) and by stable isotope exchange (Taylor et al., 1979).

Internal alteration within pegmatite bodies is a common, but usually thinly dispersed, activity. Reaction of solids with hydrothermal fluids and vapours generates leaching cavities and open fissures coated with low-temperature varieties of feldspars, zeolites and carbonates (Ginsburg's 1960 "alpine-vein" stage). These hydrothermal assemblages commonly originate after thermal equilibration of the pegmatite with its host rocks, which facilitates introduction of Ca, Fe, Mg, Ba, CI and S from surrounding lithologies. These and other elements are documented from fluid inclusions and from mineral chemistries (carbonates, Fe-Mg chlorites, epidote, Ca-rich secondary minerals of Be, and others).

#### Concluding Remarks

The preceding section presented persuasive arguments that rare-element pegmatites are essentially magmatic phenomena. The melts from which they consolidate are complex and not directly comparable to haplogranite compositions, which are normally considered to represent the last stages of granitic differentiation. Consequently, the pegmatite melts take dramatically different courses of consolidation, controlled by the qualitative spectrum and quantitative accumulation of volatiles. As mentioned above, London's model is not yet a cure-all answer to the whole problem, but it has already developed into an all-encompassing concept which covers granitic pegmatites of all geochemical varieties. Still, "reconciliation" with contrary results of Thomas et al. will be necessary, as well as interpretation of the aplitic units, of the primary fabric of albitespodumene pegmatites, and of the nature and derivation of albite pegmatites.

Nevertheless, the late-magmatic nature of pegmatite crystallization in a restricted system is confirmed today. Considerations of multiphase injection ("Nachschub"; e.g., Nikitin, 1957; Norton, 1981; Gaupp et al., 1984; Möller, 1989), metasomatic origin of extensive albitic and micaceous units (e.g., Ginsburg, 1960; Solodov, 1962, 1971; Norton et al., 1962; Stewart, 1978; Černý, 1982a,b; Foord and Cook, 1989; Thomas and Spooner, 1988a,b), recrystallization plus metasomatism of granitoid rocks (e.g., Schaller, 1925; Landes, 1933; Nikitin, 1957), opensystem syndilational crystallization from streaming vapour (Kretz et al., 1989), or exsolution of immiscible liquid (e.g., Melentyev and Delitsyn, 1969) may be either relegated to marginal, quantitatively insignificant phenomena or entirely abandoned.

#### **ACKNOWLEDGEMENTS**

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