

The mineralogy of pegmatites

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The Mineralogical Society of America, in conjunction with the Friends of Mineralogy, held a symposium on February 15-16, 1981 in Tucson, Arizona, on the Mineralogy of Pegmatites. Gordon E. Brown, Jr. of Stanford University served as organizer and technical chairman and Richard A. Bideaux of Bideaux Minerals, Tucson, Arizona, served as local general chairman.

The two day program, attended by over 150 mineralogists, consisted of 17 invited or contributed papers, which described a variety of pegmatite localities in the U.S., Canada, South America, Africa, and the USSR, and six papers on more general subjects ranging from the origin of color in pegmatite minerals to isotopic and experimental studies bearing on the paragenesis of pegmatites. In addition, Clifford Fondel of Harvard University presented an informative plenary lecture on the mineralogy and mining of early American pegmatites.

Of the twenty-three papers presented, five were of a review nature and are listed by title only. Abstracts of the other eighteen are reprinted below.

G.E. Brown, Jr.

MINERALOGY OF THE BADU PEGMATITE, LLANO COUNTY, TEXAS

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Since the discovery of the Badu pegmatite in 1936 by Mrs. Tillie Badu Moss, the pegmatite has been worked intermittently as a source of feldspar and, in recent years, for decorative quartz. The mine is located approximately 4 miles southwest of Buchanan Dam and 0.6 miles south of the Llano-Burnet highway. This location is only 4.5 miles S 35°W from the famous Barringer Hill pegmatite, which is now flooded and well below the surface elevation of Lake Buchanan. The surface exposure of the Badu pegmatite is approximately 700 feet long (east-west) and a maximum of approximately 350 feet wide. The pegmatite chiefly consists of large masses of white, milky quartz and perthitic pink microcline, some of which are more than 30 feet in diameter and most of which are single crystals. Minor minerals in the Badu pegmatite include muscovite, biotite, tourmaline, garnet, iron pyrite, sphene, anatase, fluorite, hematite, limonite, gadolinite and bastnasite. Small, clay-filled pockets in the feldspar masses have produced attractive euhedral colorless and smoky quartz crystals, many of which are deeply etched and contain fluid inclusions with movable bubbles and smoky "phantoms." The southeast wall of the present mine pit contains a discolored area with abundant limonite stains. Detailed examination of this area revealed sphene pseudomorphs, now mostly altered to anatase. The alteration of the sphene apparently was caused by a late stage fluorine-rich fluid which caused the production of the fluorite-anatase-hematite assemblage. This reaction can be described by a simple chemical equation if the sphene is assumed to contain minor amounts of iron and hydroxyl. The major portion of this reaction has been reproduced in the laboratory.

PEGMATITES OF THE WAUSAU PLUTON, WISCONSIN

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The Wausau Pluton, a large, subvolcanic body of Precambrian age, is exposed over an area of approximately 360 square km in Marathon County, Wisconsin. It consists of biotite-bearing quartz syenite and is cut by numerous dikes of pegmatite that are most abundant in an outcrop area of about 8 square km. Some of these dikes are as much as 80 m long and m thick, but corresponding average dimensions for all the dikes probably are less than 20 m and 1 m. About one-third of the pegmatite bodies contain irregular pockets, ranging in maximum dimension from less than 1 cm to at least 4 m, that have yielded an unusual variety of minerals. Nearly 50 species have been identified thus far.

The most abundant and widespread pocket constituents are quartz, albite, microcline, muscovite, and clay minerals. Surprisingly common to many of the pockets are almandine, anatase, bertrandite, brookite, chlorite, columbite-tantalite, goethite, grossular, hematite, ilmenite, lepidocrocite, opal, phenakite, psilomelane, rutile, siderite and xenotime. Less common minerals, a few of them found in only one pocket to date, include anhydrite, ankerite, barite, bavenite, beryl, biotite, boulangerite, calaverite, cookeite, fluorite, galena, gold, jamesonite, manganocalcite, microlite, monazite, parisite-synchesite, sanidine, sphalerite, stibiconite, stilpnomelane, tourmaline, and several other species not yet identified. This remarkable assemblage, which evidently represents a system with unusual concentrations of Be, Nb-Ta, Sb, and Ti, has provided excellent specimens and some superb micro-material. The pegmatite occurrences are not widely known, but they have yielded many noteworthy crystals despite rather difficult collecting conditions.

SOME FAMOUS PEGMATITE LOCALITIES IN THE U.S.S.R.

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There are numerous mineralogically diverse pegmatite deposits in the U.S.S.R., some of which are of economic importance. The localities reviewed below are past and present producers of many of the exceptionally fine mineral specimens that appear in European and American mineralogical museums.

Two interesting pegmatite regions are located within the Fenno-Scandinavian shield area. Those located on the Keivy Ridge, Kola Peninsula contain abundant amazonites of two predominant colors: greenish-blue and grass-green. Pegmatites located in northern Karelia are characterized by abundant muscovite, "ceramic minerals," and red microcline and have a variety of accessory minerals including tourmaline. The Volynsk pegmatites, located within the Ukrainian shield, are associated with the Korosten granitic pluton. They contain miarolitic cavities filled with large crystals of morion (smoky quartz), green beryl, and rosy topaz.

Two famous pegmatite localities occur in the Urals: (1) miarolitic pegmatites of the Mursinka-Aduy belt near Sverdlovsk containing beautiful crystals of blue topaz and heliodor beryl occurring on the surface of a peculiar "comb" albite; and (2) granitic pegmatites in the Ilimensk mountains near Miask which contain amazonite and numerous REE-Nb-containing accessory minerals. The latter pegmatites are located near nepheline pegmatites which contain abundant cancrinite, sodalite, "helvine", etc. Interesting tourmaline-, spodumene-, beryl-, columbite-, cassiterite-bearing pegmatites are located in the Turkestan Ridge area, Uzbekian Republic, Central Asia. In the Baikal region near Sludyanka and in the Olkhon region (Tazheran Steppe on the NW shore of Baikal) there are pegmatites containing amazonite and numerous REE accessory minerals. Miarolite pegmatites occur at Transbaikalia (in the Adun-Chelon mountains and in the Borschovochny Ridge area along the Shilka River) and contain excellent crystals of quartz, beryl, topaz, and various varieties of tourmaline. The Mama pegmatite belt, located in the North Baikal Highland is rich in muscovite and comprises various genetic types including analogues to the Bihar (India) pegmatites which contain "ruby mica veins". Finally, the REE-containing pegmatites of the Aldan Shield contain barium orthoclase and many accessory minerals.

ALPINE TYPE BERYL-EMERALD DEPOSITS NEAR HIDDENITE, NORTH CAROLINA

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Occurrences of beryl and its chromium-bearing variety emerald in the vicinity of Hiddenite, Alexander County, North Carolina have been known since 1875. Several previous attempts to exploit the deposits for mineral specimens and gemstones have met with only limited success. Most descriptions of the deposits class them as pegmatites enclosed in the local biotite-gneiss but field examinations in 1969 showed that they are hydrothermal veins deposited along a series of cross-cutting fractures in the gneiss. In non-saprolitized portions, wall alteration extends to several cm with the production of replacement muscovite and albite with some rutile. Within the veins, many of which are only partly filled, quartz (milky, smoky, late amethystine), muscovite, albite, goethite (pseudomorphous after Fe-bearing carbonate), beryl (aquamarine, late emerald) and rutile are common species. The morphology of the veins as well as the mineralization bear strong resemblances to the Alpine klefts classed as "Fundortgruppe 10f" by H.A. Stalder, and others, in *Die Mineralfunder der Schweiz* (1973). Despite the presence of small pegmatite bodies intercalated in the gneiss, beryl mineralization appears to be confined to the veins mentioned. For these reasons, the veins are classed as Alpine in type and possibly represent a unique mode of emerald occurrence. Although no spodumene-bearing body was examined during the author's visit in 1969, it is also likely that the chromium-bearing spodumene (hiddenite) from this district is unique in its mode of occurrence.

BISMUTHIAN STIBIOCOLUMBITE-TANTALITE AND OTHER MINERALS FROM THE LITTLE THREE MINE, RAMONA, CALIFORNIA

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Bismuthian stibiocolumbite-tantalite and other rare mineral species occur in the pocket zone of the main Little Three pegmatite-aplite dike. The bismuthian stibiocolumbite-tantalite is associated with topaz, lepidolite, tourmaline (elbaite), cleavelandite, quartz, microcline, orthoclase, microlite-uranmicrolite, and several secondary mineral phases: montmorillonite, pucherite, bismutite, and cookeite. Crystals of bismuthian stibiocolumbite-tantalite are euhedral, often doubly terminated, 0.01 mm to 20 mm in maximum dimension, black, elongated along c , sometimes flattened on a , and striated parallel to b (with or without polysynthetic twinning). Electron microprobe analyses of four crystals show existence of strong compositional zoning, ranging from $(\text{Sb}_{0.87}\text{Bi}_{0.14})_{1.01}(\text{Ta}_{0.48}\text{Nb}_{0.23}\text{Ti}_{0.18}\text{W}_{0.11})_{1.00}\text{O}_4$ to $(\text{Sb}_{0.92}\text{Bi}_{0.10})_{1.02}(\text{Nb}_{0.83}\text{Ta}_{0.08}\text{Ti}_{0.06}\text{W}_{0.02})_{0.99}\text{O}_4$. The maximum Bi/Bi+Sb ratio is 0.33 and the minimum is 0.06. Spectrographic analysis of bulk crystals confirms the presence of significant amounts of Ti (0.7%), W (0.7%), and Sn (0.07%). Least squares refinement of powder data (40 reflections) yields: $a = 4.952(7)$, $b = 11.803(17)$, $c = 5.596(8)$, $V = 327.11(6)\text{A}^3$. Measured densities range from 6.60 to 6.78.

Uranmicrolite contains major amounts of Bi and minor amounts of Mn, Ce, La, Pb, Sb, and Sn. Spectrographic analyses of several associated minerals also show trace to major amounts of abnormal elements (As, Sn, Ge, REE).

Most of the pale- to medium-blue topaz shows growth zoning (yellow fluorescence) under SW ultraviolet light. The fluorescence may be related to the Ge and Fe content of the topaz.

The unusual compositions and chemical zoning of several of the coexisting minerals were controlled by the individual geochemical environments within the Little Three pegmatite-aplite dike system. These properties are consistent with a genetic model that involves fractionation of a closed system silicate melt coexisting with an aqueous vapor phase. Final crystallization of pocket minerals takes place from the residual aqueous phase. Physical disruption and fracturing of pocket contents, as well as additional crystal growth (including that on broken surfaces), favors a mechanism such as pocket rupture. Loss of the aqueous fluid also permitted preservation of the pocket contents.

FOOTE MINE MINERALOGY

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The complex mineralogy of the Foote Mineral Company spodumene mine near Kings Mountain, North Carolina, can be divided into four distinct stages. All of the nearly 100 different species found there may be readily assigned to one of these stages, and all of the components which comprise these phases were in the primary pegmatite or the wallrock. Exceptional features are the lack of zoning, the remarkably high (about 20%) spodumene content, and the almost total absence of volatiles in the primary pegmatite.

Stage I required that an already crystallized, dry, spodumene-rich pegmatite was squeezed, toothpaste-like, into amphibolite and schist along and across the foliation to form complex swarms in a belt 25 miles long. The primary minerals include microcline, quartz, spodumene, Mn-apatite, cassiterite and beryl.

Stage II involved fracturing of the primary pegmatite with some brecciation, providing conduits for hydrothermal solutions which dissolved much of the primary pegmatite and reacted with the wallrock to seal it. Li-, Cs-, Rb-bearing phases formed by replacement at the contact, while albite and Li, Be, Sn high temperature phases formed in vuggy leached-out areas and solution conduits.

Stage III produced mainly anhydrous phosphates of Mn, Ca, Fe, Li; precipitated at temperatures lower than those of Stage II minerals, but by similar processes.

Stage IV minerals are the product of hypogene alteration of the Stage III suite, and are confined to the upper levels of the quarry. These are mostly hydrated, low-temperature phosphates. This locality has produced seven new species.

THE HARDING PEGMATITE: A SUMMARY OF RECENT RESEARCH

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The Harding pegmatite, located in the Picuris Range 30 km SW of Taos, New Mexico, has yielded commercial beryl, niobium-tantalum minerals, lepidolite and spodumene over a period of half a century. The pegmatite is being donated by Arthur Montgomery to the University of New Mexico to be preserved for educational purposes.

The Harding pegmatites crop out in a belt 800 m long and 50-150 m wide. The main dike, exposed for a length of 300 m with a maximum thickness of 24 m, displays remarkable internal zonation. Zones of contrasting mineralogy parallel the contacts of the dike. Eight lithologic units are distinguished: (1) border zone of quartz + albite + muscovite + perthite; (2) massive quartz + albite + muscovite; (3) quartz + lath spodumene; (4) quartz + sugary albite + perthite; (5) blocky perthite + quartz + albite; (6) microcline + spodumene + lepidolite + albite + muscovite + quartz ("spotted rock"); (7) cleavelandite + rose muscovite + quartz; and (8) cleavelandite + quartz + muscovite. The latter three are partially or wholly due to sub-olidus replacement.

Ages for 42 whole rock and mineral separates from the individual zones were determined using Rb/Sr. The linear fit of the Rb/Sr data for cleavelandites from the cleavelandite-quartz zone yields an age of $1,396 \pm 172$ m.y.b.p.; for whole rocks of the "spotted rock" unit, $1,336 \pm 73$ m.y.b.p. The average of these two ages, 1,366 m.y.b.p., may be taken as the age of the pegmatite.

Fluid inclusions in 62 beryl and quartz samples from all the lithologic units were examined. Homogenization temperatures (uncorrected for pressure) for primary inclusions from the wall, massive quartz and quartz-lath spodumene zones are in the range of 275°C to 325°C. The estimated fluid inclusion salinity, based on freezing point depression, shows that primary inclusions have a consistent salinity of 14-16 wt.% NaCl. The estimated fluid pressure, based on oxygen isotope data and the NaCl and CO₂ content of the fluids in the inclusions, suggests pressures in the range 2.0-2.5 kbar (6.0-7.5 km). Pressure corrected homogenization temperatures are consistent with the previously determined liquidus and solidus for a composite Harding sample containing 8 wt.% water.

THE TANCO PEGMATITE, SOUTHEASTERN MANITOBA

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Rare-element granitic pegmatites are widespread in the Canadian Shield, particularly in the Archean Superior and Slave Provinces. In the Superior Province, their occurrences are concentrated in greenstone belts, especially so in the English River and Quetico Subprovinces in the southwest part of the Shield. It is in the former subprovince where the giant Tanco deposit is located, known for its leading role in the world Ta production and for its reserves of Li, Rb, Cs, Be, Ga and of industrial minerals. It belongs to the Winnipeg River pegmatite district, in which eight geochemically and mineralogically distinct pegmatite groups were recently defined, and most of their parental granites identified. However, the source of Tanco and its Bernic Lake group is not exposed on the surface.

The Tanco pegmatite is a subhorizontal lenticular body, 100 x 500 x 1 m in size, consisting of seven zones of largely primary crystallization and two replacement units. So far, 73 mineral species have been identified in this pegmatite. The most remarkable of these are 9 minerals of Sn, Nb-Ta, and Ti; 28 native elements, sulfides and sulfosalts; petalite and eucryptite; Cs-rich beryl; hafnian zircon; two members of the analcime-pollucite series, and two new species, tancoite and cerňyite. Compositional characteristics of most minerals indicate extreme levels of fractionation attained by the pegmatite: K/Rb is as low as 4.0 in K-feldspar and 2.1 in lepidolite, and Rb/Cs decreases to 8.0 and 4.5 in these species, respectively; Nb/Ta is 0.05 in wodginite and 0.01 in microlite; Al/Ga reaches 1100 in feldspars and 260 in some micas; Zr/Hf in zircon is close to 5.0, and Zn/Cd ranges from 42 to 0.7.

KINDREDS AND DISTRICTS OF RARE-MINERAL PEGMATITES IN COLORADO

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Rare-mineral pegmatites in Colorado are widely distributed, but Precambrian ones are concentrated in: (1) the Front Range from the Wyoming line to Pikes Peak, (2) the Arkansas River area from Canon City to Salida and Troup Creek Pass and, (3) the Quartz Creek district of Gunnison County. Tertiary rare-mineral pegmatites occur in the Collegiate Range on Mount Antero. Precambrian pegmatites fall into four main groups: (1) those related to Boulder Creek granitoid plutons (~1700 m.y.) (Eight Mile Park district, Fremont Co.); (2) those related to Silver Plume granitoid plutons (~1400 m.y.) (Crystal Mtn. district, Larimer Co.); (3) those related to the Pikes Peak batholith (~1040 m.y.) (South Platte district, Jefferson Co., amazonite pegmatites of the Pikes Peak area); and (4) those related to the Mt. Rosa alkalic granite (~1000 m.y.) (cryolite pegmatites of the Cheyenne Mtn. area).

The rare-element mineralogy of the pegmatites depends on concentrations of the same rare-elements in their parent granitoids. Although the three main generations of granitoids each have their own characteristic assemblage and concentrations of rare elements, there is considerable variation from one pluton to another or even in zoned plutons. The genetically related pegmatites reflect both their kindred descent and this district variation in their rare-mineral assemblages. Thus, for example, Li minerals occur in some Boulder Creek pegmatite districts (Quartz Creek; Eight Mile Park); in Silver Plume districts (Crystal Mtn.); and in one Pikes Peak district (Crystal Park). Similarly RE minerals are found in pegmatites descended from Boulder Creek plutons (Lookout Mtn., Fremont Co.) from Silver Plume plutons (Jamestown) and from the Pikes Peak batholith (South Platte).

THE BLACK CLOUD PEGMATITE, TELLER COUNTY, COLORADO

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The Black Cloud pegmatite which occurs near Divide, Teller County, Colorado, is one of the most unusual rare-earth element mineral pegmatites of North America. The pegmatite, which was worked for feldspar in the 1950's and again for quartz in the 1960's is exposed in two open cuts. The deposit is ellipsoidal in shape with a bulbous nose that tapers southeastward. It is roughly 350 feet in length and is as much as 230 feet wide. Contacts with the surrounding Boulder Creek granite are sharp with no exomorphic effects. The body exhibits a well-defined internal zonal structure consisting of: 1) Wall Zone of graphic granite; 2) Outer Core of irregular masses of microcline perthite and quartz with rare beryl crystals; 3) Inner Core of massive white quartz. Large books of biotite several feet across and up to five inches thick are localized along the contact between the inner and outer core. Fluorite- and albite-rich units replace earlier formed minerals of the primary zonal sequence. These replacement units are the source of an extensive suite of rare-earth minerals. Fluocerite is the most notable of these. This exceedingly rare mineral, which is found at only one other U.S. locality, is present in masses up to eight inches long weighing over one pound. Associated rare-earth minerals include the following groups:

- a) Fluorides: fluorite, fluocerite
- b) Carbonate: bastnaesite
- c) Phosphates: monazite, xenotime
- d) Oxides: samarskite, yttrantalite, fergusonite
- e) Silicates: allanite, gadolinite, zircon, thorite, and several unidentified phases.

Other replacement phases include bertrandite, kaolinite, sericite, chlorites, opal, and iron and manganese oxides. The presence of this distinctive assemblage and the absence of such characteristic accessory pegmatite minerals as tourmaline, garnet, and apatite, and the rarity of muscovite and zircon make the Black Cloud unique among pegmatites of the United States.

RECENT PROGRESS IN EXPERIMENTAL STUDIES OF PEGMATITE GENESIS

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The experimental studies on the genesis of granitic pegmatites pioneered by R.H. Jahns and C.W. Burnham in the late 1950's have been continued and extended. Using the same composite samples of the Spruce Pine, North Carolina and Harding, New Mexico pegmatites, the detailed phase equilibria as functions of temperature and water content have been determined at confining pressures of 2, 5, and 8 kilobars. The nucleation densities and crystal growth rates of the feldspars and quartz have been measured at several isotherms at 5 kilobars as functions of the water content of the initial melt. The growth morphologies and spatial distribution of the various crystalline phases, both as isolated crystals and in interaction with other phases, have also been tabulated. These parameters are useful in developing models to explain many of the textural features which are distinctive of pegmatites.

One of the most exciting results of the present research has been the direct synthesis of graphic intergrowths of quartz in albite or oligoclase. A model has been developed to explain the origin of this texture based on the simultaneous crystallization of quartz and feldspar from the melt under conditions of local supersaturation. The presence of this texture tends to indicate certain relationships between various kinetic factors such as the nucleation and growth rate of feldspar and the diffusive redistribution of components at the melt-crystal interface rather than any state of bulk chemical equilibrium.

It is hoped that in studying these pegmatites, an extreme case in the development of granitic textures, insights into the more normal paths of crystallization will be gained.

LIGHT STABLE ISOTOPE GEOCHEMISTRY OF GRANITIC GEM- AND NON-GEM-BEARING PEGMATITES

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The study of oxygen, carbon, and hydrogen isotope systematics of whole-rocks, mineral separates, and inclusion fluids from granitic pegmatites has elucidated many details regarding the magmatic crystallization history of certain pegmatites. Further, suggestion of the origins of many of the constituents of pegmatites (including gem minerals), comparison of isotopic variations in pegmatites of Precambrian to Late Cretaceous age, comparison of both deep- and shallow-seated environments, and distinction between magmatic and hydrothermal features in pegmatites and associated wall rocks are also possible. Isotopic compositions are generally similar for all granitic pegmatites examined, but differences in mineral fractionations for simple and complex pegmatites are found. Examples from more than ten major pegmatite districts in North America and Scandinavia are discussed.

The whole-rock oxygen isotope composition of pegmatites appears in most cases to reflect pre-crystallization isotopic exchange with wall rocks, while "closed-system" crystallization is indicated for the greater part of a pegmatite's crystallization history. Oxygen isotope fractionations indicate temperatures of formation (540-800°C) compatible with experimental data. Lower solidus temperatures of Li-bearing pegmatites are reflected by larger isotopic fractionations, and subsolidus crystallization of many minerals is indicated by both textural and isotopic evidence.

The hydrogen isotope variations of muscovite, biotite, and water released from fluid inclusions indicate that interaction with meteoric water is not typical of pegmatite emplacement and crystallization. The isotopic composition of mica falls within the range of "magmatic" water ($\delta D = -40$ to -80 ‰). The measured hydrogen isotope fractionations between inclusion fluid and hydrous minerals indicate temperatures of crystallization of magmatic to hydrothermal stages to be ~ 700 to 500°C .

The carbon isotope compositions of CO_2 released from fluid inclusions are typically in the range of -6.0 to -8.0 ‰ ($\delta^{13}\text{C}$). Some variation beyond this range (-2.2 to -15.6 ‰) may be due to late-stage contamination from wall rocks, loss of CO_2 from pegmatites during crystallization, or to analytical difficulties due to the very small sample size involved.

GEM-BEARING PEGMATITES AT KLEIN SPITZKOPPE, NAMIBIA (SOUTH-WEST AFRICA)

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These pegmatites have produced fine specimens of topaz and beryl since about 1900, with production peaking in the late 1930's. Klein Spitzkoppe is an inselberg of the Spitzkoppe granite (± 180 m.y.) intrusive into the Damara schists (± 500 m.y.). The pegmatites are little more than schlieren in the granite, with occasional larger miarolytic cavities. These coarse segregations are usually unzoned, but the adjacent wallrock may be altered to tourmaline, topaz and beryl. Topaz (up to 2%) and fluorite are the most common accessory minerals throughout the Spitzkoppe granite, which is inferred to have cooled under less than 300 meters of cover.

Microcline, often blue, is abundant in the cavities, as is perthite, sometimes with the albite lamellae partially dissolved. The topaz is usually implanted on rusty quartz, as highly-perfect water-clear crystals, usually colorless but rarely blue. The beryl crystals, of several generations, are always elongated on c . They most commonly are pale green or blue, and are sometimes color zoned along c . The locality is noted for fine yellow beryl crystals, always modified by $\{11\bar{2}1\}$. Much rarer associated minerals are green to yellow octahedral fluorite, phenacite, bertrandite and florencite (steipelmannite).

There are marked similarities between these pegmatites and those occurring at Mt. Antero, Colorado.

THE GEOLOGY OF LITHIUM PEGMATITES IN THE WHITE PICACHO DISTRICT,
 MARICOPA AND YAVAPAI COUNTIES, ARIZONA

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The crystallization sequence and alteration history of primary spodumene, montebrasite, and lithiophilite have been studied in nine zoned pegmatites of the White Picacho district (WPD), Arizona. A general (and simplified) crystallization sequence for these pegmatites appears to be:

plagioclase-quartz-microcline -border
 microcline-quartz-plagioclase-(muscovite) -wall
 microcline-quartz-(spodumene)
 quartz-spodumene -intermediate zones
 quartz-montebrasite
 quartz-(lithiophilite) -core

Lithiophilite also occurs with columbite-tantalite, Mn-apatite, and Mn-tourmaline (verdelite) in relatively coarse-grained cleavelandite complexes that appear to have replaced pre-existing pegmatite. X-ray powder patterns, refined cell parameters, microprobe analyses, and some optical properties have been determined for most of the following WPD phases:

<u>Primary Li-minerals</u>		
1. spodumene	10. Ca-hureaulite	21. augelite
2. montebrasite (4-6 wt%F)	11. robertsite	22. scorzalite
3. lithiophilite	12. fillowite	23. kulanite
<u>Secondary Minerals</u>	13. dickinsonite	24. carbonate-apatite
4. eucryptite	14. fairfieldite	<u>Other primary phases</u>
5. albite	15. Mn-chloro-apatite	25. micas
6. micas	16. rhodochrosite	26. Mn-tourmaline
7. hureaulite	17. montebrasite (2-3 wt%F)	27. zircon
8. triploidite	18. crandallite	28. beryl
9. eosphorite	19. hydroxy-apatite	29. manganocolumbite
	20. brazilianite	30. columbite

THE OCCURRENCE OF PHOSPHATE MINERALS IN GRANITIC PEGMATITES: AN EXAMPLE OF LITHIOPHILITE FROM THE STEWART MINE NEAR PALA

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A comparison of thirteen well-studied, Li-rich granitic pegmatites which contain primary phosphate minerals has shown some significant similarities and differences in their features. Localities surveyed include Branchville, Connecticut; Palermo, New Hampshire; Varutrask, Sweden; Bernic Lake, Manitoba; Hagedorf, Germany; Newry, Maine; White Picacho, Arizona; Keystone, South Dakota; Viitaniemi, Finland; Sapucaia, Brazil; Bikita, Zimbabwe; Harding, New Mexico; and Pala, California. These pegmatites vary in geologic age (2.6 to 0.15 b.y.), in host rock type, in apparent depth of emplacement, and in size and shape. They each exhibit an internal zoning structure, with the primary phosphates usually occurring in the intermediate zones closely associated with the core zone. In addition to quartz, feldspars and micas which comprise the bulk of these pegmatites, phosphates are also associated with a variety of Li-silicates such as spodumene, lepidolite, petalite and eucryptite. The number of primary phosphates in granitic pegmatites is limited, with the most abundant phases being lithiophilite-triptychite $\text{Li}(\text{Mn,Fe})\text{PO}_4$, amblygonite-montebrasite $(\text{Li,Na})\text{Al}(\text{PO}_4)(\text{F,OH})$, and apatite $\text{Ca}_5(\text{PO}_4)_3(\text{F,OH})$. Current evidence suggests these minerals crystallized directly from a pegmatitic melt. These primary phosphates can occur either as crude crystals or massive nodules which range from several centimeters to several meters in size. The pegmatites surveyed show a diversity of secondary phosphates which formed by a combination of hydration, oxidation, alkali-leaching, and metasomatism of the primary phosphates under both hydrothermal and superegne conditions. These secondary phases often contain additional elements such as Na, Mg, Zn, Sr, Ba, Be and K. Hydrothermal alteration is thought to be a result of late-stage reaction between the residual pegmatite fluid or other hydrothermal solutions and the earlier-formed minerals. The variable extent of hydrothermal alteration observed in different pegmatites appears to be a function of the period of time that the pegmatite was subjected to high temperature and pressure magmatic conditions. The Stewart pegmatite in the Pala District of San Diego County is a typical example of a complex, internally-zoned granitic pegmatite which contains both Li-silicate and Li-phosphate minerals. Amblygonite, which exhibits no evidence of hydrothermal alteration, occurs in massive bodies in the core of the dike. Euhedral crystals of lithiophilite up to 30 cm in size are found in the quartz-microcline intermediate zone associated with a small number of secondary phosphates which appear to be mainly of superegne origin. The apparent shallow depth of emplacement (< 3 km) of the Stewart pegmatite suggests that there was little opportunity for the formation of secondary phosphates by hydrothermal alteration, in contrast to the extensive alteration found in pegmatites emplaced at greater depths.

THE PHOSPHATE MINERALOGY OF THE PALERMO PEGMATITE

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The Palermo No. 1 pegmatite near North Groton, New Hampshire has yielded a greater variety of phosphate mineral species than any other pegmatite in the world. A total of 56 distinct species (excluding uranylphosphates) have been verified including 11 which were first discovered here.

The phosphates are concentrated near the boundary between the intermediate zone and core of the pegmatite where primary phosphate phases crystallized during the sequential crystallization of the pegmatite. Four Fe-Mn bearing primary phosphates: triphylite, wolfeite, sarcopside, and graftonite, and two Al bearing primary phosphates: augelite and montebrasite, have been recognized. Triphylite is volumetrically most important.

Attack on the primary phosphates, principally triphylite, by late-stage, core-derived solutions was pronounced and resulted in a variety of replacements depending upon the chemistry of the solutions. Simple aqueous solutions produced ludlamite and vivianite replacements; CO₂-rich solutions yielded siderite; calcium-rich solutions produced carbonate-apatite; whitlockite, sodium and calcium-rich solutions produced arrojadite and alluaudite; and simple oxidation yielded ferrisicklerite and heterosite. Usually some combination of these replacements is observed.

Table 1. The primary pegmatite phosphate minerals*

Name	Formula	Number End-member Species
<u>Glaserite-derived structures</u>		
**Beusite-graftonite	Ca(Mn,Fe) ₂ (PO ₄) ₂	2
**Wyllieite-ferrowyllieite	(Na,Ca) ₂ (Mn,Fe,Mg) ₂ Al(PO ₄) ₃	9
Fallowite-johnsomervilleite	Na ₂ Ca(Mn,Fe) ₇ (PO ₄) ₆	2
**Apatite group	Ca ₅ (F,OH,Cl)(PO ₄) ₃	4
**Dickinsonite-arrojadite	KNa ₅ (Mn,Fe) ₁₄ AlF(PO ₄) ₁₂	2
Griphite	(Mn,Na) ₆ CaFeAl ₂ F ₂ (PO ₄) ₆	2
**Triplite-zwieselite	(Mn,Fe) ₂ F(PO ₄)	2
**Tripliodite-wolfeite	(Mn,Fe) ₂ (OH)(PO ₄)	2
Whitlockite	Ca ₉ (Mg,Fe)H(PO ₄) ₇	2
<u>Hexagonal close-packed structures</u>		
**Lithiophilite-triphylite	(Li,Na)(Mn,Fe)(PO ₄)	3
Sarcopside	(Fe,Mn,Mg) ₃ (PO ₄) ₂	1
Lithiophosphate	Li ₃ (PO ₄)	1
<u>Other structures</u>		
Berlinite	Al(PO ₄)	1
Hurlbutite	CaBe ₂ (PO ₄) ₂	1
Beryllonite	NaBe(PO ₄)	1
Trolleite	Al ₄ (OH) ₃ (PO ₄) ₃	1
**Amblygonite-montebrasite	(Li,Na)Al(F,OH)(PO ₄)	3
Augelite	Al ₂ (OH) ₃ (PO ₄)	1

*The minerals in each category are arranged in approximate decreasing temperature of crystallization.

**Most common species.

Within the replacement units, and in hydrothermal veins which have traversed them, are found the myriad of well crystallized secondary phosphates. The secondary phosphates are observed to occur in several basic assemblages which are most readily related to certain prominent cations. Principle assemblages noted are as follows:

1. Calcium: carbonate-apatite, whitlockite.
2. Iron-manganese:
 - a. reduced (Fe^{2+}): arrojadite, alluaudite, phosphoferrite, ludlamite, vivianite, metavivianite, messelite, anapaite, fairfieldite, hureaulite.
 - b. intermediate (Fe^{2+} and Fe^{3+}): rockbridgeite, dufrenite, barbosalite, strunzite, whitmoreite, beraunite.
 - c. oxidized (Fe^{3+}): laueite, pseudolaueite, stewartite, xanthoxenite, mitridatite, jahnsite, strengite, phosphosiderite, kryshanovskite, leucophosphate, tavorite, bermanite, ferrisicklerite, heterosite.
3. Aluminum: scorzalite, samuelsonite, bjarebyite, palermoite, goedkenite, goyazite, childrenite, foggite, paravauxite, gordonite, montebbrasite, brazilianite, wardite.
4. Beryllium: moraesite, roscherite, hydroxyl-herderite
5. Zinc: phosphophyllite, hopeite, schoonerite

No order of crystallization is implied by this list. However, two general trends in the crystallization sequences are observed: later phases usually exhibit greater degrees of hydration and, when possible, greater degrees of oxidation.

THE PRIMARY PEGMATITE PHOSPHATE MINERALS

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Over 180 phosphate mineral species are known from granitic pegmatites. Of these, at least 40 are primary phases; that is they crystallized early, directly from the pegmatitic magma or liquid. The earliest formed minerals are anhydrous and some (such as triphylite) are the starting materials for a whole chain of secondary hydrothermally reworked products.

The primary phosphates have been divided into three categories based on their crystal structures (Table 1). Their structures are often exceedingly complex and two of them--dickinsonite and fillowite--are among the most complex in the entire mineral kingdom. The dickinsonite-arrojadite series is literally a "garbage basket," with over twenty components occurring in the structure.

Giant crystals are known for the primary phosphates. Examples include crystals over one m in dimension for graftonite, apatite, arrojadite and triphylite. Such phases may be important models for nuclear waste containers since the phases are refractory, densely-packed and their metamict examples show no sign of leakage or exchange of ions. The most frequently occurring primary phosphates are: triphylite, apatite, graftonite, arrojadite and amblygonite.

THE GEOLOGY OF LITHIUM PEGMATITES IN THE WHITE PICACHO DISTRICT, MARICOPA AND YAVAPAI COUNTIES, ARIZONA

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The crystallization sequence and alteration history of primary spodumene, montebbrasite, and lithiophilite have been studied in nine zoned pegmatites of the White Picacho district (WPD), Arizona. A general (and simplified) crystallization sequence for these pegmatites appears to be:

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plagioclase-quartz-microcline _____]border
microcline-quartz-plagioclase-(muscovite) _____]wall
microcline-quartz-(spodumene) _____]
quartz-spodumene _____]intermediate zones
quartz-montebbrasite _____]
quartz-(lithiophilite) _____]core
  
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Lithiophilite also occurs with columbite-tantalite, Mn-apatite, and Mn-tourmaline (verdelite) in relatively coarse-grained cleavelandite complexes that appear to have replaced pre-existing pegmatite. X-ray powder patterns, refined cell parameters, microprobe analyses, and some optical properties have been determined for most of the following WPD phases:

Primary Li-minerals

1. spodumene
2. montebrasite (4-6 wt%F)
3. lithiophilite

Secondary Minerals

4. eucryptite
5. albite
6. micas
7. hureaulite
8. triploidite
9. eosphorite

10. Ca-hureaulite
11. robertsita
12. fillowite
13. dickinsonite
14. fairfieldite
15. Mn-chloro-apatite
16. rhodochrosite
17. montebrasite (2-3 wt%F)
18. crandallite
19. hydroxy-apatite
20. brazilianite

21. augelite
 22. scorzalite
 23. kulanite
 24. carbonate-apatite
- Other primary phases
25. micas
 26. Mn-tourmaline
 27. zircon
 28. beryl
 29. manganocolumbite
 30. columbite

Most secondary minerals occur as fine-grained replacement assemblages in spodumene, montebrasite, and lithiophilite. Fracture-controlled pseudomorphic alteration of the primary Li-minerals is widespread and apparently resulted from subsolidus reactions with residual pegmatitic fluids. In the WPD pegmatites, the following general sequences of secondary mineral parageneses have been recognized:

- 1 → 4 + 5 → 5 + 6 → 6
- 2 → 17 → 18 + 19 → 6 + 19 + 20 + 21 + 22 + 23 → 6 → 24
- 3 → 7 + 8 → 9 + 10 + 12 + 13 → 11 + 14 → 15 + 16

Primary crystallization of the WPD pegmatites reflects the usual increase in activities of Li species (or increasing tendency for Li to replace Na and K in alkali aluminosilicates), as well as an increase in the activities of the acidic volatiles P and F (as montebrasite succeeds spodumene as the stable primary Li-phase). Late-stage development of cleavelandite complexes may have continued as subsolidus Na-metasomatism (albitization of spodumene). Ca-metasomatism of montebrasite and lithiophilite probably began concurrently with albitization. Na- and Ca-metasomatism were followed by relatively acidic (K+H)-metasomatism (weak greisenization) that produced much secondary mica. Late-stage fluids in the WPD pegmatites were not F-rich, as evidenced by low F contents of secondary montebrasite, by formation of OH- and Cl-apatites, and by the virtual absence of lepidolite, topaz, fluorite, and triplite.

ORIGIN OF COLOR IN PEGMATITE MINERALS

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The colors of many attractive minerals in gem pegmatites appear to be due, in part, to a history of exposure to ionizing radiation. Tourmaline is green from Fe^{2+} , pink from Mn^{2+} , and much deeper red from Mn^{3+} . Radiation in the laboratory can transform Mn^{2+} to Mn^{3+} suggesting the possibility that the same process happens in nature. Manganese and radiation are also involved in the coloration of kunzite. Lavender kunzite arises from Mn^{3+} . Exposure to intense radiation turns the crystal green. This change is believed to involve the formation of Mn^{4+} . Kunzites which are blue when mined but which fade upon exposure to light demonstrate the occurrence of radiation induced oxidation in the pegmatite veins.

Beryl is blue from Fe^{2+} but turns golden yellow with radiation due to the formation of Fe^{3+} . Green beryl contains both Fe^{2+} and Fe^{3+} . It is not known whether yellow beryl forms directly from Fe^{3+} or first incorporates Fe^{2+} which is subsequently oxidized by radiation.

Blue and brown topaz are produced by radiation. No chemical cause of the colors is known, although a correlation with hydrogen ion concentration and color zones has been observed. A correlation with the concentration and structural site of hydrogen ion has also been observed with amethyst. Amethyst color involves both iron and radiation. It is yet to be determined whether smoky quartz, having radiation induced color involving aluminum ions, also shows color correlations with hydrogen ion. Amazonite feldspar color is generated by irradiation of feldspar containing lead, but factors other than just lead concentration are involved.

Garnets in the pyrope-almandine-spessartite series owe their color to Mn^{2+} and Fe^{2+} . They are among the few pegmatite minerals for which radiation has not been associated with color.

REVIEW PAPERS (LISTED BY TITLE ONLY):

1. Gordon E. Brown, Jr.: On the Nature of Pegmatitic Fluids
2. Richard V. Gwines: The Brazilian Pegmatite Province
3. Richard H. Jahns: Roles of Aqueous Vapor in Pegmatite Genesis
4. Vandall T. King: Crystallization History of a Newry, Maine Pegmatite
5. Willard L. Roberts: Secondary Phosphate Minerals from Pegmatites of the Black Hills, South Dakota