

## June 2006 Mineral of the Month: Fluorapophyllite

It is a pleasure to examine this month's mineral, a favorite among collectors because of its beauty, pastel colors, bright luster, well-developed crystal habits, and close association with zeolite minerals.

### PHYSICAL PROPERTIES

Chemistry:  $\text{KCa}_4\text{Si}_8\text{O}_{20}(\text{F},\text{OH})\cdot 8\text{H}_2\text{O}$  Basic Potassium Calcium Silicate Fluoride Hydrate, often containing some sodium and trace amounts of iron and nickel.

Class: Silicates Subclass: Phyllosilicates (Sheet Silicates)

Group: Apophyllite

Crystal System: Tetragonal

Crystal Habits: Usually well-formed, cube-like or tabular crystals with rectangular, striated prisms, square cross sections, and steep, diamond-shaped, pyramidal termination faces; sometimes with flat, rather than pyramidal, terminations in a pseudo-cubic form; also granular, lamellar, and compact.

Color: Usually colorless or white; sometimes gray or pale green, yellow, or red; rarely pale blue or violet.

Luster: Vitreous to pearly on crystal faces, pearly on cleavage surfaces.

Transparency: Transparent to translucent

Streak: White

Cleavage: Perfect in one direction

Fracture: Uneven, very brittle

Hardness: 4.5-5.0

Specific Gravity: 2.3-2.4

Luminescence: Sometimes fluoresces pale yellow-green.

Refractive Index: 1.536

Distinctive Features and Tests: Cube-like crystals with pearly luster on cleavage surfaces; prism striations; and occurrence as a secondary mineral in close association with zeolite minerals. Laboratory analysis is necessary to differentiate fluorapophyllite from closely-related hydroxyapophyllite.

Dana Classification Number: 72.3.1.1

### NAME

By complete coincidence, this makes two months in a row in which the prefix "fluor" has been added to the name of a long-accepted mineral. Ongoing examination with new equipment has allowed scientists to more closely analyze the chemical makeup of minerals, and such examination has resulted in changes to the classification of some minerals. Mineralogists no longer use well-known mineral names such as apatite and apophyllite to describe individual mineral species, but to designate mineral groups consisting of closely related species, with the fluorine-rich members now called "fluorapatite" and "fluorapophyllite." The only hitch is that nontechnical and older mineral guidebooks like John Sinkankas' *Mineralogy* use the original names, as do most casual collectors, dealers, and museums, leading to inevitable confusion.

This month's mineral is pronounced flor-a-PAH-fill-ite or flor-ap-oh-FILL-ite. Its name derives from the Greek *apo*, meaning "detached" or "off," and *phylon*, or "leaf," and translates roughly as "to leaf apart," a reference to its tendency to flake (separate along cleavage planes) into thin sheets when heated. It has been known as "fluorine apophyllite," "fluorapophyllit," "fluorapophyllita," and "fish-eye stone."

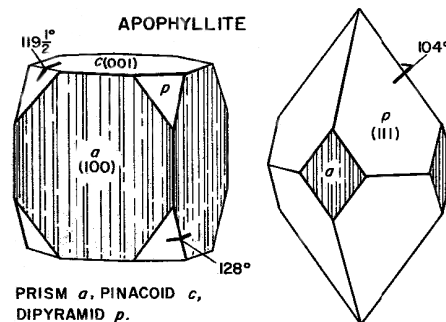


Figure 1. Crystal drawings from John Sinkankas' *Mineralogy*, used by permission. Note name used.

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### *COMPOSITION*

Fluorapophyllite's chemical formula  $\text{KCa}_4\text{Si}_8\text{O}_{20}(\text{F},\text{OH})\cdot 8\text{H}_2\text{O}$  identifies its elemental components as potassium (K), calcium (Ca), silicon (Si), oxygen (O), fluorine (F), and hydrogen (H). The ideal molecular weight of fluorapophyllite is made up of 4.87 percent potassium, 21.08 percent calcium, 29.50 percent silicon, 42.29 percent oxygen, 2.25 percent fluorine, and 0.01 percent hydrogen.

With its double cation and double anion, the latter including both silicate and hydroxyl radicals, along with attached water molecules, fluorapophyllite is a fine example of a complex mineral molecule. But before looking closely at fluorapophyllite itself, remember that a cation is a positively charged ion and an anion is a negatively charged ion. A radical is a group of atoms of different elements that are bound together to act as a single chemical entity. In fluorapophyllite's chemical name, basic hydrous potassium calcium fluorsilicate, the term "basic" indicates the presence of hydroxyl ions  $[(\text{OH})^{-1}]$ , which impart non-acidic chemical properties, while the term "hydrous" indicates the presence of attached water molecules.

The stability of any molecule depends upon an electrical balance between its cations and anions. Fluorapophyllite's two cations include a single ion of potassium ( $\text{K}^{1+}$ ) and four ions of calcium ( $\text{Ca}^{2+}$ ), providing a collective +9 charge. On the anionic side of the formula, the silicate radical  $(\text{Si}_8\text{O}_{20})^{8-}$  includes eight silicon ions ( $\text{Si}^{4+}$ ) and 20 oxygen ions ( $\text{O}^{2-}$ ) and provides a -8 electrical charge. An additional -1 charge is provided by a fluorine ion ( $\text{F}^{-1}$ ) or a hydroxyl ion ( $\text{OH}^{-1}$ ) in what is known as a substitution radical. (The comma within this radical indicates continuous partial or complete substitution.) Thus, the balance of the cationic +9 charge and the anionic -9 charge make the fluorapophyllite molecule electrically stable.

Fluorapophyllite's formula  $\text{KCa}_4\text{Si}_8\text{O}_{20}(\text{F},\text{OH})\cdot 8\text{H}_2\text{O}$  also shows that the molecule has eight attached water molecules ( $\text{H}_2\text{O}$ ) that are known collectively as "water of hydration." Water of hydration is bound to the parent molecule as integral water molecules that retain their characteristic composition and structure. These electrically neutral, attached water molecules function neither as cations nor anions and do not affect the electrical balance of the parent molecule. In water molecules, each oxygen atom is bound covalently by shared electrons to two hydrogen atoms. The two small hydrogen atoms group together on one side of the large oxygen atom. This creates a dipolar effect, with the two grouped hydrogen atoms retaining a small positive charge and the opposite side of the molecule retaining a small negative charge. Water of hydration is attached to the parent molecule by hydrogen, or polar, bonding, in which the water molecules' faintly positive poles are attracted to negative charges.

Fluorapophyllite is a member of the silicates, which are combinations of oxygen and silicon with one or more metals. The basis of the silicate structure is the silica tetrahedron  $(\text{SiO}_4)^{4-}$ , in which four equally spaced oxygen atoms surround a silicon atom. Silicate minerals consist of silica anions bound to metal cations in repeating structural units that form seven general groups: independent tetrahedral silicates (nesosilicates); double tetrahedral silicates (sorosilicates); framework silicates (tectosilicates); single- and double-chain silicates (inosilicates); ring silicates (cyclosilicates); and sheet silicates (phyllosilicates).

The phyllosilicates, which include fluorapophyllite, are also called "sheet silicates," a term that describes their crystal structure. In the sheet silicates, each silica tetrahedron shares three oxygen ions with adjacent tetrahedra. This inter-molecular arrangement forms very thin, sheet-like structures that extend indefinitely in two directions. Within these flat sheets, the silica tetrahedra are arranged in three-, four-, five-, six-, or eight-membered rings. Fluorapophyllite has alternating four- and eight-membered rings. To visualize this arrangement, think of the silica layers as chicken wire with alternating octagonal and square holes. Both octagons and squares have a fourfold symmetry, which is reflected in fluorapophyllite's basic tetragonal or pseudo-cubic crystal form and in the multiples of four in the  $(\text{Si}_8\text{O}_{20})^{4-}$  silicate radical.

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Sheet silicates are subclassified structurally into two-layer and three-layer groups. As a two-layer sheet silicate, fluorapophyllite consists of alternating sheets of silica tetrahedra and sheets of metal cations. Within the silica sheets, the charges of the three shared oxygen ions of each tetrahedra are neutralized, but the remaining, unshared oxygen on the tetrahedral apex still carries a -1 charge. Negatively charged fluorine (or hydroxyl) ions occupy the hexagonal holes between the apices of the sheet's tetrahedra. Adjacent to the silica sheets are sheets of positively charged potassium and calcium ions and attached water molecules. Ionic bonding holds these alternating silica and metal-cation sheets together. The fluorapophyllite lattice thus has three distinct types of atomic bonds: covalent bonds within the silica tetrahedra; ionic bonds between the metal-cation sheets and the silica sheets; and hydrogen bonds that attach the water of hydration.

Strong covalent bonding within the silica tetrahedra gives fluorapophyllite a moderate hardness of 4.5-5.0. But the weak ionic bonding between the sheets creates perfect cleavage in one direction. One of fluorapophyllite's most interesting properties is its tendency to "leaf apart" when heated, a phenomenon caused by the nature of the hydrogen bonding of its attached water molecules. Hydrogen bonds (the bonds that cause water to freeze into ice) are very heat-labile. Application of even moderate heat to fluorapophyllite will break some of the hydrogen bonds. The resulting displacement of water molecules disrupts the atomic spacing between the sheets, thus parting the weak ionic bonds and causing the sheets to separate along their cleavage planes. Higher levels of heat will break all the hydrogen bonds, dehydrating the crystal completely, disrupting all the ionic bonding, and causing it to deteriorate into amorphous particles.

The Dana classification number 72.3.1.1 first identifies fluorapophyllite as a phyllosilicate with a two-dimensional, infinite sheet structure that consists of other than six-membered rings. The subclassification (3) next defines it as a phyllosilicate with sheets consisting only of four- and eight-membered rings. Finally, fluorapophyllite is a member of the apophyllite group (1), of which it is the first (1) of four members. The apophyllite-group minerals, all hydrous silicates with generally similar properties, include:

72.3.1.1 Fluorapophyllite  $\text{KCa}_4\text{Si}_8\text{O}_{20}(\text{F},\text{OH})\cdot 8\text{H}_2\text{O}$

72.3.1.2 Hydroxyapophyllite  $\text{KCa}_4\text{Si}_8\text{O}_{20}(\text{OH},\text{F})\cdot 8\text{H}_2\text{O}$

72.3.1.3 Natroapophyllite  $\text{NaCa}_4\text{Si}_8\text{O}_{20}\text{F}\cdot 8\text{H}_2\text{O}$

72.3.1.4 Carletonite  $\text{KNa}_4\text{Ca}_4\text{Si}_8\text{O}_{20}(\text{CO}_3)_4(\text{OH},\text{F})\cdot \text{H}_2\text{O}$ , a rare silicate that also contains the carbonate radical.

These chemical formulas show the cationic and anionic substitution among the apophyllite-group minerals. Fluorapophyllite is by far the most abundant apophyllite, followed by hydroxyapophyllite. Both natroapophyllite and carletonite are rare. As end members of a complete solid-solution series, fluorapophyllite and hydroxyapophyllite are differentiated by the predominance of either fluorine or hydroxyl ions. Because this can be determined only by laboratory analysis, all apophyllite-group minerals are often referred to simply as "apophyllite." Most specimens labeled "apophyllite" are actually fluorapophyllite.

Fluorapophyllite is a secondary mineral that primarily forms at low temperatures in vesicular (gas-formed) cavities of basaltic rock. When magma is extruded, the reduction in pressure causes gas bubbles to form. As the magma cools and solidifies, these gas bubbles form hollow vugs, or cavities. Over time, the circulation of mineral-rich groundwater through the basalt partially or completely fills these vugs with such minerals as quartz, or silicon dioxide ( $\text{SiO}_2$ ); calcite, or calcium carbonate ( $\text{CaCO}_3$ ); the zeolite minerals, which are hydrous sodium calcium aluminum silicates; prehnite, or basic calcium aluminum silicate [ $\text{Ca}_2\text{Al}_2\text{Si}_3\text{O}_{10}(\text{OH})_2$ ]; and fluorapophyllite. Fluorapophyllite also occurs to a lesser extent in cavities of contact zones where igneous rock has intruded limestone.

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Fluorapophyllite and the other apophyllite-group minerals are often erroneously thought to be zeolites. Zeolites are tectosilicates (framework silicates) that can lose and regain water of hydration without altering their crystal structure. Although fluorapophyllite is a phyllosilicate, it does have certain zeolitic properties, including the ability to lose some water of hydration without altering its crystal structure. However, unlike the zeolites, fluorapophyllite cannot *regain* water of hydration. Further confusion stems from fluorapophyllite's frequent mineralogical association with zeolite minerals in basaltic environments.

Fluorapophyllite is an allochromatic (other-colored) mineral. When pure it is colorless, but trace amounts of impurities such as iron and nickel impart a range of pale colors.

### *COLLECTING LOCALITIES*

The most noted and productive of fluorapophyllite's many collecting localities are the traprock quarries in the Deccan Trap basalt formations at Pashan and Jalgaon near the city of Pune (Poona), Maharashtra, India, as described in *About Our Specimens*.

Fine fluorapophyllite specimens are also found at the Bartolon Quarry at Pocos de Caldas, Minas Gerais, Brazil; the Fengjiashan Mine at Daye, Hubei, China; the Sampo Mine at Takahashi on Japan's Honshu Island; the 1st Sovietski, Bor, and Danbury mines at Dal'negorsk, Primorskiy, in Russia's Far-Eastern Region; in Germany at the Schneeberg and Freiburg districts near Erzegebirge in Saxony and the St. Andreasburg and Bad Harzburg districts in Lower Saxony; and in the Scottstown Quarry, Ballymena, County Antrim, Northern Ireland, and the Seravel Mine, Westgate, County Durham, England.

In Canada, fluorapophyllite is collected at Mt. Sainte-Hillaire, Rouville County, Quebec. Pearly-white crystals in large druses have come from the silver mines near Guanajuato in Mexico's state of Guanajuato.

In the United States, fine fluorapophyllite specimens associated with prehnite have been collected at numerous quarries and road cuts in the basalt formations of Passaic, Bergen, Hudson, Morris, and Essex counties in northern New Jersey. Other notable sites include the Virginia diabase quarries near Centreville in Fairfax County and those near Conklin and Leesburg in Loudon County; the Dyer and Robeson quarries at Birdsboro, Berks County, Pennsylvania; North and South Table Mountain at Golden, Jefferson County, Colorado; and the Christmas Mine in the Banner District of Gila County, Arizona. Large, glassy fluorapophyllite crystals occur in the copper deposits of northern Michigan's Keweenaw Peninsula.

### *JEWELRY & DECORATIVE USES*

Because it is usually colorless or pale in color, only moderately hard (Mohs 4.5-5.0), difficult to cut due to its perfect one-directional cleavage, and sensitive to heat generated by cutting and soldering operations, fluorapophyllite has limited use in jewelry. Small amounts of fluorapophyllite are tumbled and drilled for stringing in necklaces. Some exceptional crystals have been cut into 10- and 20-carat "collector" gems. Fluorapophyllite is also known as "fish-eye stone," a name referring to its gray-blue, mother-of-pearl-like luster on cleavage surfaces and gem facets.

Because of its gem-like, vitreous-to-pearly luster and tendency to occur as well-developed crystals in interesting habits, fluorapophyllite is a very popular mineral collectible as both singular and composite specimens. In composite specimens, fluorapophyllite is usually associated with prehnite and zeolite minerals, combinations that offer fascinating contrasts in color and crystal form. Because of the prevalence of wonderful specimens from India, such pieces remain affordable to all collectors.

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### *HISTORY & LORE*

In the late 1700s, mineralogists considered the apophyllite-group minerals to be a single mineral species. The name “apophyllite” came into use about 1800 when blowpipe tests revealed the mineral’s tendency to exfoliate, or literally “leaf off,” in thin sheets when heated. In 1931, researchers used newly developed x-ray-diffraction techniques to determine the crystal structure of “apophyllite.”

Although advanced chemical-analysis techniques revealed that “apophyllite” was a basic hydrous potassium calcium fluorsilicate with widely varying amounts of fluorine and sodium, the belief that it was a single species persisted until recently. In the 1970s, researchers Pete J. Dunn, Roland C. Rouse, and Julie A. Norberg, working with the Minerals Section of the National Museum of Natural History (Smithsonian Institution) in Washington, D.C., proposed that, because of its close association with fluorine-free zeolite minerals and prehnite, a fluorine-free variety of apophyllite might also exist. After a search of the literature revealed previous apophyllite analyses with low fluorine contents, they began a search for a pure variety in which hydroxyl ions substituted completely for fluorine ions. After analyzing 50 “apophyllite” specimens by scanning electron microscopy and chemical techniques, the team identified a fluorine-free variety and concluded correctly that the fluorine- and hydroxyl-rich varieties are actually independent end members of a solid-solution series.

After further study, Dunn, Rouse, and Norberg published their results in 1978 in the *American Mineralogist* in an article titled “Hydroxyapophyllite, a New Mineral, and a Redefinition of the Apophyllite Group.” That same year, the International Mineralogical Association’s Commission on New Minerals and Mineral Names approved hydroxyapophyllite and fluorapophyllite as new species. Naming these minerals as such has generated lingering controversy. Some mineralogists believe that the proper names should have been “apophyllite-OH” and “apophyllite-F,” respectively. This is because modern mineral-literature glossaries list fluorapophyllite and hydroxyapophyllite alphabetically. Therefore, a search for “apophyllite,” the name used for more than 150 years, often turns up nothing, confusing many collectors and even some earth-science professionals.

Because it was identified by studying specimens from various localities, fluorapophyllite, unlike most minerals, is not assigned a specific type locality. In 1981, mineralogists discovered the newest member of the apophyllite group, the rare mineral natroapophyllite ( $\text{NaCa}_4\text{Si}_8\text{O}_{20}\text{F}\cdot 8\text{H}_2\text{O}$ ). Further research may turn up additional members of the apophyllite group.

According to modern metaphysical beliefs, fluorapophyllite and the other apophyllite-group minerals help reduce fears and feelings of apprehension. By suppressing anxieties, worries, and uncertainties, fluorapophyllite helps the wearer to establish control of emotions and situations. In the past, fluorapophyllite has been medicinally employed to alleviate respiratory and allergenic ailments and to help regenerate skin and mucous membranes. In 1998, the Republic of Mali featured a beautiful composite specimen of fluorapophyllite and stilbite on its 100-franc postage stamp.

### *TECHNOLOGICAL USES*

Fluorapophyllite has no technological uses.

### *ABOUT OUR SPECIMENS*

“Green apophyllite with mesolite from Poona, velvety white okenite puffballs from Bombay, water-clear apophyllite from Jalgaon, pale golden powellite from Nasik, and intensely blue cavansite from Waghonii are

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the paragons of Indian basalt minerals." So starts the January-February issue of the *Mineralogical Record*, devoted entirely to zeolite and related minerals from this area. We have of course, now featured both cavansite and apophyllite twice, and featured okenite in January 1998, and hope to do so again. The high volume of fine specimens coming from this region has made them quite affordable, another wonderful aspect.

Our specimens are labeled as coming from Jalgaon, a specimen-producing district surrounding Jalgaon, a city with a population of nearly 3.7 million people, located about 420 km from Mumbai (formerly Bombay,) in Maharashtra State on the northern part of the Deccan Traps. Jalgaon produces sorghum, cotton,

bananas, and is a center for cotton-textile and vegetable oil mills, and is known for its gold market. As is true of many localities of the Deccan traps, black basalt is quarried for construction purposes. And as is also true of these localities, crystal-lined openings containing wonderful specimens are often found in the basalt. Our wonderful pieces all come from a single large quarry in the village of Paldhi, about 15 miles from Jalgaon. Paldhi is described by an Indian geologist in a kind of stream-of-consciousness fashion as "a small village with the 2000 population the main job is agriculture, when they do not have job in the field they may go to quarry but mostly as in India the people working in quarry are from one community they works at different mines the local people may help them those are known as Dagadi wadar meaning stone worker they are specialist in digging wells for drinking water or agricultural purpose." Quite a mouthful!

Although fluorapophyllite occasionally fluoresces, Jalgaon specimens do not. We have thoroughly enjoyed examining Club member's pieces prior to sending them, noting especially the clarity of many of the crystals; the stalactitic-type formation some pieces show; and the occasional crystals of salmon-colored stilbite that occur with the fluorapophyllite. A few pieces were of fluorapophyllite growing on and almost entirely covering a calcite crystal! Many of the crystals have sharp terminations like the drawing on the right in Figure 1, while others have the sharp crystal with a flat termination. Yes, we have enjoyed every aspect of this month's mineral, which has certainly found its place among our favorites!



**Figure 2.** Quarry source of our specimens, village of Paldhi, about 15 miles from Jalgaon. The minerals are found in vugs in the walls as the quarry is expanded.

References: *Dana's New Mineralogy*, Eighth Edition; *Encyclopedia of Minerals*, Second Edition, Roberts, et al, Van Nostrand Reinhold Co.; *2004 Fleischer's Glossary of Mineral Species*, Joseph Mandarino and Malcolm Back, The Mineralogical Record Company; *Mineralogy*, John Sinkankas, Van Nostrand Reinhold Co.; *Manual of Mineralogy*, Cornelius Hurlbut and Cornelia Klein, Twenty-first Edition, John Wiley & Sons; *Minerals of the World*, Walter Schumann, Sterling Publishing Co.; *Gemstones of the World*, Walter Schumann, Sterling Publishing Co.; *Handbook of Mineralogy: Silica and Silicates*, Vol. 2, J. W. Anthony, et al, Mineral Data Publishing; *Gem and Crystal Treasures*, Peter Bancroft, Western Enterprises, 1984; *Structural Chemistry of Silicates*, F. Liebau, Springer-Verlag Publishing Co.; "Spectacular New Apophyllite from India's Deccan Trap Basalts," Muhammad F. Maki, *Rocks & Minerals*, March-April 2002; "Refinement of the Crystal Structure of Apophyllite," A. Colville, P. Black, and P. Anderson, *American Mineralogist*, Vol. 55, 1971; "Hydroxyapophyllite, a New Mineral and a Redefinition of the Apophyllite Group," Pete J. Dunn, Roland C. Rouse, and Julie A. Norberg, *American Mineralogist*, Vol. 63, 1978; "Nomenclature Revisions in the Apophyllite Group: Hydroxyapophyllite, Apophyllite, and Fluorapophyllite," Pete J. Dunn and Wendell E. Wilson, *The Mineralogical Record*, March-April 1978; "The Nomenclature Debacle," John S. White, *Rocks & Minerals*, May-June 2004; "Connoisseur's Choice: Fluorapophyllite from Jalgaon, Maharashtra, India," Robert B. Cook, *Rocks & Minerals*, May-June 1995; "Minerals of the Deccan Traps, India," Berthold Ottens, Richard V. Gaines, Rustam Z. Kotnavada, Rock H. Currier, and Wendell E. Wilson, *The Mineralogical Record*, January-February 2003.

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