# MINERALOGY OF THORIUM

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### ABSTRACT

The known minerals that contain thorium are divided into two groups: those that contain the element as an essential constituent, and those that contain thorium as a vicarious constituent. The first group contains 6 distinct species, of which only the silicates, thorite and thorogummite, and the oxide, thorianite, are of importance, together with 16 minor chemical variants of these species that have been accorded varietal names. The second group, numbering more than 60 minerals, in places contains variable and usually small amounts of thorium in solid solution. Some of these minerals, notably monazite, are of economic interest because of their occurrence in relatively large amounts.

### INTRODUCTION

The average content of the element thorium in the outer crust of the earth is about 12 grams per ton. Thorium is a little less abundant than lead and about three times as abundant as uranium. Thorium in the lithosphere is contained chiefly in minerals in which it is a nonessential constituent. Of the six known thorium minerals, only the thorium silicates—thorite and thorogummite—and the thorium dioxide, thorianite, are of potential economic interest. Most of the earth's thorium is dispersed in small and variable amounts in solid solution in other minerals as an occasional, vicarious constituent. The distribution of thorium in this manner is a selective process. The principal ore mineral of thorium is the cerium phosphate, monazite, which contains variable amounts of thorium in solid solution.

The mineralogical expression and the geochemical distribution of thorium both as an essential and as a vicarious constituent show striking similarities, both genetically and economically, to certain other elements; namely, uranium, cerium, zirconium, and hafnium. The very similar elements hafnium and zirconium will be considered hereafter as a unit with the symbol Zr. In the present paper, the mineralogy of thorium will be characterized in context with these elements, and the crystallochemical and other factors that influence their geochemical descent will be indicated.

That close geochemical relations should exist among these elements is suggested by similarities in their general chemical behavior and, more fundamentally, by similarities in their electronic structures. In the lanthanide series of elements extending from atomic number 58 (Ce) to 71 (Lu), the back filling in the *4f* shell that causes the very close chemical resemblances in this series is paralleled in the actinide series of elements 90 (Th) to 96 (Cm) through back filling in the *5f* (or *6d*) shell. Quadrivalent Ce thus is similar to Th. $Ce^4$ also is analogous to both Hf (72) and Zr (40), through filling of the *5d* and *4d* shells. Quadrivalent uranium shows close analogies to thorium. The similarities between Th, U, Ce, and Zr are illustrated by the isostructural relation of their dioxides and orthosilicates and by the extensive mutual solid solution series in these and other groups of compounds. The mineralogical and geochemical disimilarities between these elements are due primarily to the small size of the $Zr^4$ ion as compared to $Th^4$, $Ce^4$, and $U^4$ as a group, and to the assumption by uranium of the sexivalent state and by cerium of the trivalent state—both stable states under geologic conditions—in addition to the quadrivalent states of these elements.

## THE DESCRIPTIVE MINERALOGY OF THORIUM

The element thorium which was named after Thor, the Scandinavian god of war, was discoverd in 1828 by the Swedish chemist J. J. Berzelius in a mineral from a pegmatite on the island of Lövö in the Langesund fiord, southern Norway. The name thorite was given to the mineral. Thorite and its hydroxyl-containing variant, thorogummite, and thorianite are the most important and widely distributed of the six known thorium minerals (table 1).

Thorite ideally has the formula $ThSiO_4$, and material of this composition can be readily synthesized. The composition of the natural mineral, however, always is greatly modified both by the entrance of other elements into solid solution, notably U, Fe, Ca, rare earths and P, and by secondary alteration. The alteration is of two types, which operate concomitantly. Thorite is radioactive, and alpha particles together with beta and gamma radiation are emitted by it or its decay products in the passage to the stable end element lead 208. The internal emission and absorption of alpha particles disrupts the normal crystalline structure of the mineral and causes it to become partly or completely disordered (metamict). This type of alteration occurs in many other thorium and uranium minerals and in recent years has been the subject of much investigation (Pabst, 1952; Pellas, 1951, 1952). This structural disintegration is accompanied by chemical alteration, involving hydration—commonly to the extent of 8 to 10 percent by weight of $H_2O$—and oxidation of $U^4$ to $U^6$ and of $Fe^2$ to $Fe^3$. A partial leaching of Si also may take place at this stage. These features of thorite, together with difficulties in the chemical analysis of the mineral, have brought much confusion to the description of the species. No less than 14 different minerals described in the older literature as distinct species are now known or thought to be identical with thorite or its variant thorogummite (table 1). The analytical difficulties attending thorite led to the description of three supposed new elements, carolinium, donarium, and berzelium. Zircon also has a bulky nomenclatural halo of ill-defined or synonymous minerals that has arisen for similar reasons (table 6), and the analytical difficulties here resulted in the supposed new elements ostranium, norium, and jargonium. The identity of the orange-yellow isotropic substance called orangite with thorite was established only after controversy in the period 1850–70. The description and discrediting of supposed new minerals in these categories unfortunately is still an active matter.

Thorogummite is a recently established (Frondel, 1953) variant of thorite in which tetrahedral groupings of four (OH) ions substitute vicariously for the $(SiO_4)$ groups of the structure. The formula may be written $Th(SiO_4)_{1-x}(OH)_{4x}$ in which $x$ is as much as 0.25 in natural material. In the uranium analogue, coffinite (Stieff, Stern, and Sherwood, 1955), $U(SiO_4)_{1-x}(OH)_{4x}$, the value of $x$ is about 0.5. Cyrtolite is an analogous hydroxyl-containing variety of zircon (table 4). The mechanism of hydroxyl substitution in these minerals is similar to that operating in the so-called hydrogarnets, in which a partial series in natural material and a complete series in synthetic material extend from $Ca_3Al_2(SiO_4)_3$ by way of $Ca_3Al_2(SiO_4)_{3-x}(OH)_{4x}$ to $Ca_3Al_2(OH)_{12}$. Thorogummite always appears to be a low-temperature secondary mineral in nature, produced by the recrystallization of metamict thorite by the weathering of other minerals, but the question of the entrance of (OH) into high-temperature pegmatitic or vein thorite and zircon remains open. Thorogummite and cyrtolite have been synthesized hydrothermally at temperatures as low as 150° C, and the occurrence of (OH) in both synthetic and natural material has been verified by thermal and infrared analysis.

Thorite and thorogummite are tetragonal in crystallization and belong in the zircon structure type. The structure is based on isolated $SiO_4$ tetrahedra (nesosilicates) with the thorium ions in 8-coordination with oxygen. Owing to its metamict character, thorite affords only faint and diffuse X-ray diffraction effects or gives no X-ray pattern at all. When metamict thorite is heated to 800°–900° C or higher, it recrystallizes exothermally to an aggregate of minute crystals of either tetragonal or monoclinic $ThSiO_4$, or to a mixture of one or the other of these with $ThO_2$. The nature of the recrystallization products varies with the temperature and time of heating, the composition of the original material, and other factors not clearly understood. The recrystallization products give sharp X-ray diffraction effects, and this is a convenient means of identification. Thorogummite gives X-ray patterns in unheated material. When thorogummite is heated, water including (OH) is lost, and a small amount of $ThO_2$ may appear, but the diffraction pattern remains that of tetragonal $ThSiO_4$. Metamict thorite is isotropic with a relatively low and variable index of refraction. Thorogummite is anisotropic, but the particle size is much too small for satisfactory optical study and optical techniques generally are unsatisfactory as a means of identifying these minerals.

Following the discovery of thorite in 1828, the next well-defined mineral containing thorium as an essential constituent to be discovered was thorianite, $ThO_2$, in 1904. This was followed by the description of pilbarite in 1910, huttonite in 1951, cheralite in 1953, and the redefinition of thorogummite in 1953. The crystal structure of thorianite is analogous to that of fluorite, $CaF_2$, with the thorium ions in 8-coordination with oxygen. The crystallochemical and mineralogical relations of $UO_2$, $CeO_2$, and $ZrO_2$ to $ThO_2$ are discussed in the following section. Natural thorianite contains considerable amounts of other elements in solid solution, particularly U and Ce, and like all thorium and uranium minerals accumulates Pb due to radioactive decay. Helium also is present. The uranium is present as both $U^4$ and $U^6$. The content of $U^6$ probably is secondary, owing to oxidation of $U^4$ with a concommitant entrance of oxgyen into the vacant 8-fold position of the structure, as in partly oxidized uraninite, but no study of the effect has been made. The oxidation of synthetic solid solutions of the composition $(Th,U)O_2$ has been studied experimentally by J. S. Anderson and coworkers (1954). Thorianite and uraninite, almost alone among radioactive minerals, do not become metamict, apparently because of the inherent stability of the fluorite-type structure.

Of the remaining thorium minerals, pilbarite is an ill-defined substance found sparingly as a secondary mineral in the weathered outcrop of a pegmatite in Western Australia. It needs further study. Huttonite, the monoclinic polymorph of $ThSiO_4$, is known as detrital grains in the heavy-mineral fraction of beach sands in New Zealand. It may be more widely distributed as an accessory mineral in black sands than is known. Huttonite has not been found in place, but it probably occurs in pegmatites and quartz-segregation veins in metamorphic rocks. The factors that influence the formation of huttonite and thorite are not known. Both phases have been synthesized by sintering of $ThO_2$ and $SiO_2$, the formation of huttonite being favored at higher temperatures, and both phases have been obtained hydrothermally between 400° and 700° C by reaction of coprecipitated gels of $ThO_2$ and $SiO_2$ in the presence of $ThF_4$. Cheralite is monoclinic and isostructural with both monazite and huttonite. It represents an intermediate member of the system $ThSiO_4$-$CePO_4$-$CaTh(PO_4)_2$. In this system a considerable range of solid solution exists by the substitution of $Ce^3$ and $Th^4$ with valence compensation effected by the coupled substitution of $Ca^2$ for $Ce^3$ or of $Si^4$ for $P^5$. Material in this series near monazite in composition is quite common, but huttonite and cheralite are rare.

The first large-scale use for thorium came with the development by Welsbach about 1884 of the incandescent gas-mantle, composed of $ThO_2$ with about 1 percent of $CeO_2$. Thorium now also finds use as a catalyst, in refractories, and in thoriated tungsten filaments. It is of potential importance in the field of atomic energy, since the natural isotope of thorium, thorium-232, can be converted by neutron bombardment to thorium-233, which yields uranium-233, a fissionable isotope. The thorium for gas mantles was obtained from monazite. The radioactive element mesothorium is a byproduct. The occurrence of thorium in monazite was first described by Kersten in 1839. The amount of thorium present varies widely, although roughly constant in the material from a given locality or area, and thorium-free monazite is rare. The usual content is less than 8 to 10 percent by weight of $ThO_2$, but in some specimens the content is as much as 30 percent $ThO_2$. Monazite, it is said, averages about 1 part $U_3O_8$ per 50 parts $ThO_2$. The manner in which the thorium is contained in monazite is discussed extensively in the literature. It was long thought to be present as mechanically admixed thorite or another thorium compound, and a few such occurrences have been observed, but it is now believed that the element is present in solid solution in substitution for Ce.

Other minerals in addition to monazite contain thorium as a vicarious constituent. Table 2 lists about 60 minerals—some are ill defined—that have been found at times to contain more than 0.1 percent by weight of $ThO_2$. A great many minerals, including several in which thorium could be expected to occur for crystallochemical reasons, never have been examined for this element. The minerals that contain vicarious thorium are chiefly silicates, oxides, and niobate-tantalates. Almost all these minerals are primary, and occur chiefly in pegmatites associated with alkalic or granitic igneous rocks. Only five of them, monazite, xenotime, allanite, zircon, and, to a less extent, pyrochlore, have a wide distribution as accessory minerals in igneous or metamorphic rocks. The cations that occur associated with Th in these minerals are typically Ce and other rare earths, Zr, Ca, and U. The main factors involved in this association are geochemical and crystallochemical. The former tend to associate these elements with thorium in the parent solutions or melts from which the minerals have formed, and the latter determine the entrance of thorium into the various mineral phases that are formed.

The most effective host minerals for Th are compounds of U, Zr, or Ce that are isostructural with thorium compounds, such as zircon, uraninite, and monazite, or compounds in which mechanisms of coupled substitution are available for the housing of large, polyvalent ions, such as the niobate-tantalates and monazite.

## COMPARISON OF THE MINERALOGY OF THORIUM, URANIUM, CERIUM, AND ZIRCONIUM

Thorium, uranium, cerium, and zirconium are markedly oxyphile, and the known minerals of these elements all contain oxygen with the exception of a few rare fluorides of cerium. Sulfides, sulfosalts, tellurides, and the native metals are not known as minerals. Thorium and uranium form synthetic sulfides and carbides, and the oxysulfides ThOS and UOS, but the formation of these substances in nature is unlikely on thermochemical grounds.

The common minerals that contain Th, U, and Zr as essential constituents are oxides and silicates. Cerium differs in that it does not occur directly as an oxide, although $CeO_2$ is a well-known synthetic compound, and $Ce_4$ is commonly present, sometimes in large amounts, in solid solution in natural $UO_2$ and $ThO_2$. Cerium is carried in minerals chiefly as phosphates and silicates. As vicarious constituents, Th, U, Ce, and Zr again are found principally in oxides and silicates. Thorium also is an important vicarious constituent in the cerium phosphate, monazite. Further, these four elements are characteristic constituents of the niobate-tantalates.

The principal differences in the mineralogy and geochemistry of Th, U, Ce, and Zr stem from the valence states taken by these elements in nature. Thorium and zirconium always are quadrivalent. Uranium occurs both as quadrivalent and sexivalent ions, and cerium both as quadrivalent and trivalent ions. The diversity of the mineralogy of uranium—about 70 distinct species containing uranium as an essential constituent are known as compared to 6 for thorium and 15 for zirconium—is due primarily to the stability of the sexivalent $U^6$ ion. More properly, $U^6$ should be described as the uranyl ion. In crystal structures, $U^6$ ordinarily is in distorted 6-fold or 8-fold coordination, and the two apical oxygen ions of the coordination group are more closely associated with the $U^6$ to give a linear complex cation, $(UO_2)^{-2}$. In $(UO_2)(CO_3)$ the $U^6$ is in 8-fold coordination with 6 oxygen ions in a plane and 2 apical oxygen ions (Christ, Clark, and Evans, 1955). The $U^6$ in partly oxidized $UO_2$ presumably is in distorted 8-fold coordination. The bivalent uranyl ion is of sufficient stability to preserve its identity in solution. The known uranium minerals are listed in table 7. All the primary uranium minerals, including uraninite and coffinite, which contain U as an essential constituent, and the many minerals that contain U as a vicarious constituent, house the element only in the quadrivalent state, so far as is known. The 68 or so remaining minerals that contain U as an essential constituent are built of the uranyl ion. All are of secondary origin. None of these minerals have analogues containing Th, Ce, or Zr in place of U. This is due to the large size and unusual shape of the uranyl ion, which has no counterpart in the compounds of Th, Ce, or Zr. It is only quadrivalent uranium that forms compounds isostructural with compounds of Th, Ce, and Zr. In these minerals the ions involved all are of the same charge and are of spherical shape with comparable radii (table 3) so that they play the same crystallochemical role. It also may be noted that the uranyl ion, so far as is known, does not occur as an important vicarious constituent in other minerals. This strongly contrasts with the behavior of Th, Zr, quadrivalent U, and both quadrivalent and trivalent Ce. This behavior again is due to the size and shape of the uranyl ion, which does not allow it to proxy readily for the ordinary ions of crystal structures. The entrance of sexivalent uranium into solid solution in the structure of the silicates of igneous rocks and pegmatites also would be influenced by other factors, including the thermal instability of the uranyl ion at high temperatures and the oxygen content of the magma. Some rock silicates show a weak uranyl fluorescence at liquid air temperatures.

Uranium differs from thorium and zirconium in that it occurs in large amounts in hydrothermal deposits formed at relatively low temperatures. In hydrothermal veins, uraninite is typically associated with sulfides, arsenides, and sulfosalts of Cu, Fe, Ni, Co, Zn, Bi, Ag, and other elements. These deposits are generally very low in their content of Th, Ce, and Zr. On exposure to weathering, uraninite readily oxidizes to the uranyl ion, and the other minerals oxidize to relatively soluble sulfates and other oxysalts. This results in the formation in the oxidized zone of such deposits of a large variety of hydrated uranyl oxides, sulfates, arsenates or phosphates, for the most part also containing Cu, Ca, Mg, Ba or other cations in addition to the uranyl ion. The alkaline-earth cations and the phosphate content are in part derived from associated gangue minerals but mostly come from the wall rock or the meteoric circulation. The uranyl oxysalts in general are relatively soluble and occur typically in gossans, as efflorescences on outcrops and the walls of mine workings, and in caliche-type deposits. Large amounts of uranium can be lost during oxidation by solution in the meteoric circulation, particularly in acid waters (Phair and Levine, 1953). This again contrasts with the behavior of Th and Zr. The primary compounds of these elements are chemically stable and quite insoluble, and during the weathering cycle tend to accumulate in the detrital residues. Soluble salts of these elements hydrolyze readily to insoluble hydrous oxides. Zirconium does not form minerals in the zone of weathering, and thorium and cerium only very rarely.

Cerium, like uranium, differs from thorium and zirconium in that it possesses two valences that are stable under geologic conditions, $Ce^4$ and $Ce^3$. Both ions are mineralogically important. The $Ce^4$ ion bears a close crystallochemical resemblance to the $Th^4$ and $U^4$ in particular. It has not yet been found in minerals as an essential constituent although the occurrence of $CeO_2$ and $CeSiO_4$ analogous to $ThO_2$ and $ThSiO_4$ could be expected. $Ce^4$ does occur, however, dispersed in solid solution as a vicarious constituent in uranium, thorium, and other minerals. It may be noted in this relation that cerium apparently often has been reported in mineral analyses in the trivalent state, as $Ce_2O_3$, without knowledge of its actual valence state in the mineral. The $Ce^3$ ion is considerably larger than the $Ce^4$ ion (table 3) and has no mineralogically stable counterpart in Th, U, or Zr. The mineralogy of the $Ce^3$ ion therefore stands alone among these elements. Trivalent cerium occurs in minerals both as an essential and as a vicarious constituent (tables 2 and 5) and it occurs in significant amounts as a vicarious constituent much more widely than does Zr, although Zr is roughly five times more abundant than Ce in the lithosphere. This is due partly to the fact that the $Ce^3$ ion is relatively close in size to certain other elements and enters readily into solid solution in their compounds. These elements include the trivalent rare earths and particularly the common element Ca. Calcium is bivalent, and the substitution of $Ca^2$ by $Ce^3$ requires a concomitant substitution to compensate for the change in valence. In cerian fluorite, the substitution of $Ce^3$ for Ca is compensated by the entrance of additional F into structural vacancies; in apatite and the apatite-type mineral britholite and its numerous analogues the substitution of $Ce^3$ for $Ca^2$ is compensated by a concomitant substitution of $Si^4$ for $P^5$; in sphene the entrance of $Ce^3$ is compensated by the substitution of $Al^3$ for $Ti^4$. The initial content of Ce, and especially of Y, in alkalic igneous rocks retained in the magmatic stage of crystallization is much larger than that of Th. Y is taken into solid solution in accessory sphene, apatite, fluorite, and garnet more readily than in monazite (Sahama and Vähätalo, 1939).

Although Zr is a relatively abundant element, its mineralogy both as an essential and as a vicarious constituent is very much restricted. This is due primarily to the high charge and intermediate size of the $Zr^4$ ion (table 3). Among the geologically important quadrivalent elements, Zr does not substitute appreciably for Si into the framework of silicates or for Ti because of the large difference in size. Zr ordinarily is 8-coordinated with oxygen, whereas Si and Ti are 4- and 6-coordinated, respectively. Among other geologically important elements, where bivalent Fe, Ca, and Mg and the univalent alkalies come into consideration, there is a somewhat closer approximation in ionic size to $Zr^4$, but there is a larger difference in ionic charge, involving 2 or 3 units instead of but 1 as in the pair $Ce^3$–$Ca^2$. This latter circumstance severely limits the extent of a possible coupled substitution through both the demands of local electrostatic (Pauling rule) balance in the crystal structure and the lack of small, low-charged cations that can substitute concomitantly either for the central ion (Si, Al, P, As, S) of the anionic framework or for other cations in order to effect valence compensation. Actually the great bulk of Zr in nature is contained in but a single mineral, zircon, $ZrSiO_4$. Almost all the known minerals containing Zr as an essential or vicarious element are silicates (table 6). None of these minerals, aside from zircon, are known to carry appreciable amounts of Th in solid solution, although minerals containing $Ce^3$ commonly contain small amounts of $Th^4$. $Th^4$ is much closer in size to $Ce^3$ than to $Zr^4$.

The dioxides of $Th^4$, $U^4$, and $Ce^4$ are isostructural, in the $CaF_2$ structure type, and form a complete solid-solution series between all end compositions, $(Th, U, Ce)O_2$ at high temperatures in synthetic material (Magneli and Kihlborg, 1951; Rüdorff and Valet, 1952; Trzebiatowski and Selwood, 1950). In natural material, $CeO_2$ or solid solutions with $Ce > (Th, U)$ is not known, but $Ce^4$ is present in solid solution in considerable amounts in both natural $UO_2$ and $ThO_2$. Zirconium oxide has several polymorphs. The form stable at natural conditions is the monoclinic phase, baddeleyite, with a distorted $CaF_2$-type structure, but one of the high-temperature polymorphs, not known in nature, is isometric with the $CaF_2$ structure. In the synthetic system $UO_2$–$ZrO_2$ a series (Lambertson and Mueller, 1953) extends from $UO_2$ to about 40 percent by weight of $ZrO_2$, with a diphase region extending to about 50 percent by weight of $ZrO_2$ and a tetragonal series extending from that point to $ZrO_2$. There also is evidence of solid solubility of Th in synthetic monoclinic $ZrO_2$. Very much smaller amounts of Zr occur in solid solution in natural $ThO_2$ and $UO_2$. There is question whether the Zr reported in some analyses of pegmatitic uraninite is present in solid solution or as admixed zircon. $CeO_2$ forms only a partial series with $ZrO_2$ in synthetic material (Duwez and Odell, 1950). The solubility relations of the dioxides of Th, U, and Ce on one hand and of Zr on the other clearly show the effect of the relatively small Zr ion. Trivalent rare earths such as La and Y go into limited solid solution in $ThO_2$ in both natural and synthetic material (Brauer and Gradinger, 1951). The C-type polymorphs of $La_2O_3$, $Y_2O_3$, and such, have a structure related to that of $ThO_2$, but with certain cation positions vacant, and the series from $ThO_2$ involves the coupled entrance of vacancies to provide valence compensation.

In natural material the series between $ThO_2$ and $UO_2$ apparently has a large central gap. In general, the uraninite of pegmatites contains relatively large amounts of Th and Ce in solid solution, whereas the uraninite of hydrothermal veins and black-ore sandstone deposits of the Colorado Plateau is notably deficient in these elements. This is a problem of geochemical association rather than of the temperature dependency of the solid solubility.

Besides the dioxides, some of the elements under consideration form other oxides. Ce forms $Ce_2O_3$, not known as a mineral; U forms $U_3O_8$, several polymorphs of $UO_3$, and other anhydrous oxides. None of these are known as minerals, although $U_3O_8$ might be expected to occur in nature. Uranium also forms a number of hydrated uranyl oxides, in part also containing Ba or Pb, that are widespread in small amounts. None of these uranium compounds have counterparts with Th, Ce, or Zr.

The economic importance of the oxides of Th, U, Ce, and Zr varies widely. Uraninite (here referred to as $UO_2$ but actually departing considerably from this composition owing to partial oxidation and solid solution) is the principal ore mineral of uranium. $CeO_2$ does not occur in nature, and cerium is obtained almost wholly from the cerium phosphate, monazite. $ThO_2$ is not an important actual or prospective ore mineral of thorium, although it is probably widely distributed as a very minor accessory mineral in beach and river sands. Small amounts of thorianite, however, have been obtained in Ceylon. Baddeleyite, $ZrO_2$, although an extremely rare mineral as compared to zircon, is an important ore mineral at one locality, in the Poços de Caldas, Brazil (Guimarães, 1948), where it occurs in large amounts with zircon as fibrous crusts in hydrothermal veins derived from and situated in syenitic igneous rocks. The parent rocks contain much Zr as eudialyte and as a vicarious constituent in alkalic pyroxenes and other silicates. The complex Zr silicate eudialyte occurs in large deposits in alkalic rocks in southern Greenland (Bøggild, 1953) and the Kola peninsula and is a potential source of Zr; it contains some Ce but lacks significant amounts of Th. Thorianite, $ThO_2$, does not occur in hydrothermal deposits analogous to baddeleyite. Sodium metasomatism in alkalic rocks may result in the liberation, and hydrothermal concentration, of Zr and certain other polyvalent ions (Luchitski, 1947).

The principal silicates of the elements here considered are those of the thorite and thorogummite groups (table 4). The available evidence indicates a very limited solid solubility between the isostructural phases $ThSiO_4$ and $ZrSiO_4$, extending perhaps to a few mol percent under natural conditions. In the ternary system $USiO_4$-$ThSiO_4$-$ZrSiO_4$ there is extensive solid solubility of U into $ThSiO_4$ and a very limited solid solubility, apparently of the order of a few atomic percent, of U into $ZrSiO_4$. Anhydrous $USiO_4$ does not occur in nature and has not been synthesized in spite of repeated attempts, but the solid solution $(Th,U)SiO_4$ extends from $ThSiO_4$ toward $USiO_4$ at least as far as Th:U=1:1. $CeSiO_4$ also does not occur in nature and has not yet been synthesized. The ill-defined mineral freyalite may represent a Th-containing variety of $CeSiO_4$ or its hydroxyl-containing analogue. Cerium is commonly present in considerable amounts in thorite and thorogummite but is lacking or presen only in small amounts in zircon. The mutual solubilit in the system $ThSiO_4$-$USiO_4$-$ZrSiO_4$ and the entranc of $Ce^4$ into the phases of this system clearly illustrat the role played by ionic size. The extent of soli solubility of U in both natural and synthetic materi appears to be greater in thorogummite and cyrtolit than in anhydrous thorite and zircon. Thorium lacing in the uranium member of the thorogummit group, coffinite, in the Colorado Plateau area but th is due to the lack of thorium in this geochemic province.

Huttonite, a monoclinic polymorph of $ThSiO_4$ is structural with monazite, has no known analogues in Ce⁴, or Zr silicates. Among the eight monoclinic a tetragonal polymorphs possible between $ZrSiO_4$, ThSi $YPO_4$, and $CePO_4$, it may be noted that the tetragon structure is taken by the small ions Zr and Y and t monoclinic structure by the large ion Ce. Only T intermediate in size, develops both polymorphs. Pu $CeSiO_4$ presumably would be monoclinic and isostru tural with huttonite. In synthetic material the sol solubility of U in monoclinic $ThSiO_4$ appears to be l than that in tetragonal $ThSiO_4$.

Thorium forms no natural silicates other than thori thorogummite, huttonite, and the ill-defined mine pilbarite. Uranium forms no silicates that cont solely $U^4$, other than coffinite, but does form a num or uranyl silicates (table 7). All of these are second in origin, and none have analogues with Th, Ce, or in place of U. Cerium forms silicates contain essential $Ce^3$, and $Ce^3$ enters as a vicarious element i still others (tables 2 and 5). Some cerium silicates contain Th and U in solid solution, but others, such as rite, have not yet been observed to carry significant amounts of these elements. Allanite, a common silicate of the epidote group, carries Ce, Th, and U in solid solution. The elements Ca, La, and Y also are close associates of Ce in silicates. In addition to zircon, zirconium forms a considerable number of silicates (table 6). Most of these are complex in constitution, and all are rare. Further, none of them have analogues with Th, Ce, or U. The complex zirconium silicates generally contain alkalies in their composition; this feature is less marked with the cerium silicates and is lacking with uranium silicates.

U and Zr have not been found as phosphates. Th forms a silicate-phosphate, cheralite, that has already been discussed. Small amounts of $(PO_4)$ are commonly found in thorite and thorogummite substituting for $(SiO_4)$. This series, with accompanying valence compensation by substitution of $Ce^3$ for Th, leads to a hypothetical tetragonal polymorph of $CePO_4$ isostructural with xenotime, $YPO_4$, and zircon. Compared to monazite, xenotime carries negligible amounts of Th. Cerium has a marked affinity for P as compared to $U^4$, Th, and Zr. Monazite is a very common mineral, and other cerium minerals contain P as an essential or vicarious constituent (tables 2 and 5). Some of these magmatic or pegmatitic minerals including britholite, abukamalite, belovite, and melanocerite are structural derivatives of apatite. The vicarious Th content of these apatitelike minerals is of interest in connection with the occurrence of $U^4$ in small amounts in the apatite of phosphate rock. The substitution of Th is much larger than that of U, in part probably owing to factors favoring the entrance of compensating Si into magmatic rather than sedimentary apatite. Both the Th and the U substitute into the Ca positions of the apatite structure. In late magmatic and pegmatitic apatite the amount of substitution for Ca is in the order $Ce^3 > Th > U > Zr$. This is not the order of closeness of approach in ionic size to Ca, but it is the order of increasing electronegativity of the ions. In an isostructural group of phases, as in the present instance, increasing electronegativity of the cation is in a general way accompanied by a less ionic and weaker bond with the anion held in common. The entrance of $Ce^3$ into solid solution in apatite is favored both by this factor and by the relative ease of valence compensation. The observed enrichment of Ce and other rare earths in pegmatitic rather than early magmatic Ca minerals also appears to be related to the relative electronegativity of the ions (Ringwood, 1955).

The only natural halogen-containing compounds of Th, U, Ce, and Zr are those of Ce (table 5). Thorite is found with fluorite in pegmatites and in some vein deposits, and the fluocarbonates of cerium are typical associates, but fluorine does not occur as an essential constituent in any thorium mineral.

The many uranyl carbonates include rutherfordine and a considerable number of complex carbonates containing the uranyl ion together with other cations, chiefly Cu, Ca, Mg, and Na. These minerals, together with the carbonate-fluoride-sulfate schroeckingerite and with lanthanite, are relatively soluble secondary minerals that occur chiefly as efflorescences. The numerous fluorocarbonates of cerium, on the other hand, are relatively insoluble and form at much higher temperatures. These minerals appear to contain relatively little thorium in solid solution as compared to the silicates and phosphates of cerium.

The niobate-tantalates are characterized as a group by the presence, often in large amounts, of Th, U, Zr, Ti, $Fe^3$, and rare earths. The relative amounts of these elements vary widely through mutual substitution, and commonly a distinction between essential and vicarious constituents is not significant. The yttrium group of rare earths usually predominates over the cerium group. The diverse and large content of ions of high valence in the niobate-tantalates offers opportunities for mutual substitution that are not available to the same ions as they occur individually in silicates and other compounds dominated by the presence of bivalent and univalent cations. The niobate-tantalates are not of economic importance as sources of Th or U, because of their rarity and refractory chemical nature.

## GEOCHEMISTRY AND OCCURRENCE OF THORIUM

The geochemistry of Th, U, Ce, and Zr, as these elements appear in igneous rocks, is similar and is governed largely by the low concentration and high valence of their ions. The activation energy of migration of their ions (E-value of Wickman, 1943) is relatively high, which would tend to freeze the ions in the main stage of crystallization of the magma, but their concentration is in general too low to permit the appearance of phases in which these elements are essential constituents. In broad terms, they either concentrate in residual solutions or are included in solid solution in the minerals that form the bulk of the rock. In the case of gallium, an element of roughly the same abundance as thorium, the identity in valence and near identity in ionic radius with aluminum causes virtually all of this element to become dispersed in solid solution in aluminosilicates of the magmatic stage. In thorium and uranium, however, the high charge and large size of their quadrivalent ions do not permit entrance into the normal rock minerals, and as crystallization proceeds the residual solutions become enriched in these elements. Zirconium, much commoner than uranium and thorium, in part crystallizes in the magmatic stage as accessory zircon and may then house a certain amount of Th and U, a circumstance on which the age-determination method of E. S. Larsen, Jr., is based. Both monazite and xenotime, however, appear to house Th and U more readily than zircon (Hutton, 1947). Zirconium and the rare earths, notably $Ce^4$, together with other polyvalent elements such as Nb and Ta also tend to concentrate in the residual solutions. Thorium, cerium, and uranium have a marked affinity for alkalic rather than granitic or intermediate igneous rocks in the broad course of magmatic differentiation. The bulk composition of the magma also influences the mineralogical expression of trace elements. The ratio of alkalies to aluminum was emphasized in this regard by A. E. Fersman and V. M. Goldschmidt in their concept of plumasitic and agpaitic magmas. Zirconium appears to be responsive to this factor. In highly aluminous magmas it crystallizes directly as a phase, zircon, but in highly alkalic magmas the excess of strongly electropositive elements complexes the Zr, which is comparable in electronegativity to Al, as a vicarious substitute for Al in silicates. Thorium and uranium, less electronegative and much larger in size than aluminum, are not as markedly dispersed in this way in the silicates of alkalic rocks.

The main types of deposits that carry concentrations of thorium minerals are pegmatites, hydrothermal veins, and detrital deposits. Pegmatites associated with alkalic igneous rocks, particularly nepheline syenites and their variants, are notably rich in thorium. Well-known and important occurrences of this type are in southern Norway (Brőgger, 1890), the Kola peninsula of Russia (Fersman, 1926; Fersman and Bohnstedt, 1937) and southern Greenland (Bøggild, 1953). These pegmatites also are relatively high in content of rare earths, with the cerium earths predominating over the yttrium earths, together with Zr, Nb, Ca, P, and F. Tantalum and uranium are minor constituents. These pegmatites are feldspathic but generally lack quartz and may contain nepheline (often altered to zeolites), together with pyroxenes and a variety of complex silicates containing Zr. Apatite is a characteristic accessory mineral in the pegmatites, and it sometimes occurs separately, as in the Kola peninsula (Antonov, 1934), as very large deposits associated with alkalic igneous rocks. This apatite typically contains Ce and Si and very small amounts of Th. Pegmatites derived from granitic rocks tend to contrast with those from alkalic rocks in containing, on the whole, a smaller amount of Th and in being relatively enriched in Y over Ce, Ta over Nb, and U over Th. The granite pegmatites generally are quartz-rich, with Zr present chiefly as zircon. A small production of thorite has been obtained from alkalic pegmatites in Norway, but these deposits in general are not potential large-scale sources of Th. Thorium may possibly be obtained as a byproduct in the utilization of magmatic apatite, analogous to the production of uranium from sedimentary phosphate rock.

Hydrothermal vein deposits containing thorium have become known only during the past few years. They differ from the base-metal sulfide types of veins, in which thorium is lacking in significant amounts, and they show resemblances to the cerium- and thorium-rich pegmatites associated with alkalic rocks. The Powderhorn district, Gunnison County, Colo., contains veins and mineralized shear zones carrying thorite or thorogummite with calcite, dolomite, quartz, barite, cerian fluorapatite, and bastnaesite, according to J. C. Olson and S. R. Wallace (written communication). The deposits occur in and near alkalic igneous rocks, including the Iron Hill stock, that are notably rich in rare earths, Ti, Nb, Ba, and Sr. Concentrations of perovskite occur in the area. In the Mountain Pass area, San Bernardino County, Calif. (Olson, Shawe, Pray, and Sharp, 1954), large veins and irregular bodies containing bastnaesite with calcite, dolomite, barite, parisite, and sahamalite occur in association with alkalic igneous rocks. The deposit contains Th in small amounts as a vicarious constituent. In the Salmon Bay area on Prince of Wales Island, Alaska (Wedow and others, 1953), veins carrying thorite with carbonates, hematite, and monazite are associated with other veins lacking Th but carrying carbonates with parisite and bastnaesite. Some deposits of bastnaesite, as in the fluorite-barite veins of the Gallinas Mountains, N. Mex. (Glass and Smalley, 1945), and the contact metamorphic deposit at Bastnaes, Sweden, do not carry appreciable amounts of thorium. Vein deposits containing thorite or thorogummite but lacking rare earths are also known. In the Wet Mountains in Custer and Fremont Counties, Colo., a very large area contains veins containing a thoritelike mineral together with hematite, quartz, barite, carbonates, and minor fluorite and sulfides (R. A. Christman, written communication). The deposits appear to be related to albite syenite intrusive bodies. Quartz-hematite veins with a thoritelike mineral and minor amounts of monazite and allanite also occur in districts in east-central

Idaho and southwestern Montana (Trites and Tooker, 1953). These vein deposits of Th and Ce minerals in general are extremely low in U, Zr, Y, Nb, and Ti. They show that the geochemical descent of Th and Ce, like that of U, continues into the hydrothermal stage. The baddeleyite deposits of Brazil are roughly analogues of the Th and U vein deposits, but Zr in general is retained in high-temperature minerals. The separation of U and (Th, Ce) in the hydrothermal stage remains unexplained. Uranium apparently carries further into the lower-intensity hydrothermal zones, together with S, As, and heavy metals.

Thorium also occurs as a very minor constituent in deposits other than veins and pegmatites, such as with Nb in carbonatites and in a few contact metamorphic deposits. In the sedimentary cycle, thorium (Koczy, 1949a, 1949b), unlike uranium, is not a significant constituent of the carbonaceous marine black shales and apparently does not play an important biogeochemical role. Zirconium is like thorium in this respect and is deposited chiefly as detrital zircon in nearshore clastic sediments. In sea water, uranium is enriched relative to thorium.

Alluvial deposits contain the largest known reserves of thorium. The thorium-containing mineral, monazite, occurs widely distributed as an accessory mineral in igneous and metamorphic rocks and in pegmatites. The content in gneiss and schist sometimes runs as high as 0.1 percent by weight of monazite, but efforts to mine and concentrate the mineral directly have been unsuccessful. The high specific gravity, hardness, and general stability of monazite cause it, when freed by weathering, to become mechanically concentrated in alluvial deposits. Important deposits of this kind are found in Travancore, Ceylon, the States of Bahia and Espirito Santo in Brazil, New South Wales, and Queensland. In the United States, alluvial deposits of monazite are found in Florida, the Appalachian region, Idaho, and elsewhere. The monazite is associated with ilmenite, zircon, magnetite, and garnet chiefly, and is separated and concentrated by magnetic methods. Thorite (Hutton, 1950) and thorianite are widespread in trace amounts in alluvial deposits. The known deposits of monazite sands are very large, and the world's supply of thorium is now and probably will continue to be largely drawn from them.

TABLE 1.—*Minerals containing thorium as an essential constituent, with their varieties and synonyms*

| *Name* | *Composition* | *Content of $ThO_2$ (percent)* | *Reference No.*[1] |
|---|---|---|---|
| Cheralite | $(Th,Ca,Ce)\ (PO_4,SiO_4)$ | 30, variable | 1, 4 |
| Huttonite | $Th(SiO_4)$ | 81.5 (ideal [2]) | 1, 5 |
| Pilbarite | $ThO_2 \cdot UO_3 \cdot PbO \cdot 2SiO_2 \cdot 4H_2O$ | 31, variable | 1, 6 |
| Thorianite | $ThO_2$ | Series to $UO_2$ | 1, 2 |
| Thorite | $Th(SiO_4)$ | 81.5 (ideal [2]) | 1, 2 |
| Thorogummite | $Th(SiO_4)_{1-x}(OH)_{4x}$ | 24 to 58 or more | 1, 7 |
| | *Varieties and synonyms* | | |
| Aldanite | Variety of thorianite high in Pb and U content | | 1, 9 |
| Auerlite | Variety of thorite or thorogummite relatively high in $(PO_4)$ content | | 1 |
| Calciothorite | Variety of thorite or thorogummite relatively high in Ca content | | 1, 18 |
| Chlorothorite | Synonym of thorogummite | | 7 |
| Enalite | Variety of thorite or thorogummite relatively high in content of U, rare earths, and $(PO_4)$? | | 1 |
| Eucrasite | Variety of thorite or thorogummite relatively high in content of Ca and rare earths | | 1, 18 |
| Ferrothorite | Variety of thorite or thorogummite relatively high in $Fe^3$ content | | 1 |
| Freyalite | Variety of thorite or thorogummite very high in Ce content | | 1, 18 |
| Hyblite | Synonym of thorogummite; apparently contains some $(SO_4)$ | | 1, 7 |
| Hydrothorite | Synonym of thorogummite | | 1, 7 |
| Mackintoshite | Synonym of thorogummite. Contains $U^4$ | | 1, 7 |
| Maitlandite | Synonym of thorogummite. Relatively high in $U^4$ and Pb content | | 1, 7 |
| Nicolayite | Synonym of thorogummite | | 1, 7 |
| Orangite | Varietal name for yellow to orange metamict thorite | | 1, 18 |
| Uranothorianite | Varietal name for thorianite high in content of uranium [uranoan thorianite, $(Th,U)O_2$] | | 1 |
| Uranothorite | Varietal name for thorite high in content of uranium [uranoan thorite, $(Th,U)(SiO_4)$] | | 1 |
| Wisaksonite | Synonym of thorite | | 10 |

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[1] References cited at end of table 2.

[2] Usually less because of substitution of other elements for thorium.

TABLE 2.—*Minerals containing thorium as a vicarious constituent, with their varieties and synonyms*

| *Name* | *Composition* | *Content of $ThO_2$ (percent)* | *Reference No.* |
|---|---|---|---|
| **Abukamalite** | $(Ca,Y,Th)_5(Si,P,AlO_4)_3(O,F)$ | $<1$ | 23 |
| **Allanite** | $(Ca,Ce,Th)_2(Al,Fe,Mg)_3.Si_3O_{12}(OH)$ | 0 to ≈3 | |
| **Alvite** | Ill-defined substance, near zircon | 15 in one analysis | |
| **Ampangabeite** | $\approx(Y,Er,U,Ca,Th)_2(Nb,Ta,Fe,Ti)_7O_{18}$ | 1 to 2 | 2 |
| **Bastnaesite** | $(Ce,La)(CO_3)F$ | $<1$ | 3 |
| **Betafite** | $\approx(U,Ca)(Nb,Ta,Ti)_3O9.nH_2O$ | 0 to ≈1 | 1, 2 |
| **Blomstrandine** | Synonym of priorite | | |
| **Brannerite** | $\approx(U,Ca,Fe,Th,Y)_3Ti_5O_{16}$ | 0 to 12 | 1 |
| **Bröggerite** | Variety of uraninite relatively high in content of rare earths and thorium. Thorian uraninite. | 14 | |
| **Cappelenite** | $\approx BaY_6B_6(SiO_4)_3O_{12}(OH)_2$ | $<1$ | 18 |
| **Calciosamarskite** | Variety(?) of samarskite relatively rich in Ca | 2 to 3 | 2 |
| **Caryocerite** | $\approx(Ce,La,Th,Ca)_5(Si,B)_3(O,OH,F)_{13}$ | 13.6 in one analysis | 18 |
| **Cerianite** | $(Ce,Th)O_2$ | ≈5 | 24 |
| **Chinglusite** | $\approx(Na,K)_4(Mn,Ca)_5(Ti,Zr)_3Si_{14}O_{41}\cdot 9H_2O$ | $<1$ | 15 |
| **Cordylite** | $(Ce,La)_2Ba(CO_3)_3F$ | $<1$ | 3 |
| **Clarkeite** | $\approx(Na,Ca,Pb)_2U_2(O,H_2O)_7$ | 2.4 in one analysis | 1 |
| **Cyrtolite** | Altered zircon containing $H_2O$ and often small amounts of U and Th. | | |
| **Davidite** | $(Fe,Ce,U)(Ti,Fe,V,Cr)_3(O,OH)_7$ | $<1$ | 1 |
| **Euxenite** | $(Y,Ca,Ce,U,Th)(Nb,Ta,Ti)_2O_5$ | 0 to ≈5 | 2 |
| **Eschynite** | $(Ce,Ca,Fe,Th)(Ti,Nb)_2O_6$ | 0 to 17 | 2 |
| **Fergusonite** | $(Y,Er,Ce,U,Th)(Nb,Ta,Ti)O_4$ | 0 to ≈5 | 2 |
| **Fersmite** | $(Ca,Ce,Na)(Nb,Ti,Fe,Al)_2(O,OH,F)_6$ | $<1$ | |
| **Formanite** | $(Y,Er,U,Th)(Ta,Nb)O_4$ | ≈1 | 2 |
| **Fourmarierite** | $\approx PbO.4UO_3.5H_2O$ | small | 1 |
| **Gummite** | Generic term for orange-red to yellowish alteration products of uraninite; chiefly fourmarierite and vandendriesscheite. | | 1 |
| **Hagatelite** | Variety of zircon containing rare earths, Nb, Ta, and Th | ≈1.5 | 17 |
| **Hokutolite** | Radioactive variety of barite containing Pb and probable also Ra, U, Th. | | 3 |
| **Irinite** | Thorian variety of loparite (perovskite) with composition $(Na,Ce,Th)(Ti,Nb)(O,OH)_3$. | | 13 |
| **Johnstrupite** | Silicate of Ce,Ca,Ti,Zr,Na | $<1$ | 1, 2 |
| **Khlopinite** | $\approx(Y,U,Th)_3(Nb,Ta,Ti,Fe)_7O_{20}$ | 2.2 in only analysis | 2 |
| **Lovchorrite** | $\approx Ce_4Ca_{10}Ti_3Si_{10}O_{39}F_6$ | $<1$ | 16 |
| **Lovozerite** | $\approx(Na,K)_2(Mn,Ca)ZrSi_6O_{16}.3H_2O$ | small amount | 14 |
| **Lyndochite** | Variety of euxenite-polycrase relatively high in Ca and Th. | ≈5 | 2 |
| **Melanocerite** | $\approx(Ce,Y,La,Ca)_5(Si,B,P)_3(O,OH,F)_{13}$ | 1.7 in one analysis | 18 |
| **Microlite** | $(Na,Ca)_2(Ta,Nb)_2O_6(O,OH,F)$ | $<1$ | 1, 2 |
| **Mosandrite** | $\approx NaCa_6Ce_2(Ti,Zr)_2Si_7O_{25}(OH,F)_7$ | $<1$ | 18 |
| **Muromontite** | Apparently a variety of allanite high in Be content | ≈1 | |
| **Monazite** | $(Ce,Y,La,Th)(PO_4)$ | 0 to ≈30, usually 10 to 12 | 3 |
| **Naegite** | Variety of zircon containing rare earths, U, and Th | 2.8 | 20 |
| **Nuolite** | Ill-defined mixture | 2 to 4 | 2 |
| **Orthite** | Synonym of allanite | 0 to ≈3 | |
| **Oyamalite** | Variety of zircon containing rare earths, $(PO_4)$, and Th (≈1 percent) | | 17 |
| **Perrierite** | Perhaps identical with allanite | 4.6 | 21 |
| **Pisekite** | Niobate-tantalate with U, Th, rare earths. Th present, amount not known. | | 1 |
| **Pyrochlore** | $NaCa(Nb,Ta)_2O_6F$ | 0 to ≈5 | 1, 2 |
| **Polycrase** | $(Y,Ca,Ce,U,Th)(Ti,Nb,Ta)_2O_5$ | 1 to 5 | 2 |
| **Polymignyte** | $\approx(Ca,Fe,Y,Zr)(Nb,Ti,Ta)O_4$ | 3.9 in one analysis | 2 |
| **Priorite** | $(Y,Er,Ca,Fe,Th)(Ti,Nb)_2O_6$ | 0 to ≈8 | 2 |
| **Rinkite** | $\approx(Na,Ca)_{12}(Ce,Ti)_5Si_8(O,F)_{36}$ | $<1$ | 16 |
| **Rinkolite** | $\approx(Ca,Na)_{22}Ce_2(Si,Ti)_{10}O_{28}(F,OH)_8$ | small | 16 |
| **Samarskite** | $(Y,Er,Ce,U,Fe,Th)(Nb,Ta)_2O_6$ | 0 to ≈4 | 2 |
| **Steenstrupine** | $\approx(Na,Ca,Ce,La,Th)_3(Mn,Fe,Ta)(Si,Be,P)_3(O,OH,F)_{12}$ | 2 to 7 | 22 |
| **Tanteuxenite** | Variety of euxenite relatively rich in Ta | 2.8 in one analysis | 2 |
| **Thalenite** | $Y_4Si_4O_{13}(OH)_2$ | $<1$ | |
| **Thucholite** | Hydrocarbon mixture containing much U, Th, and rare earths in the ash. | | 11, 12 |
| **Tritomite** | Silicate with B, F, Ce, La, Ca, Th | ≈9 | 18 |
| **Tscheffkinite** | Silicate of Ce, La, Th, Fe, Ti, Ca | 0 to 20, usually ≈1 | 19 |
| **Tengerite** | $\approx CaY_3(CO_3)_4(OH)_3.3H_2O$ | ≈1 | 3 |
| **Uraninite** | $UO_2$ (ideally); usually partly oxidized, with Ce, Y, Pb, Th, etc. | 0 to 14 at least | 1, 2 |
| **Uranophane** | $Ca(UO_2)_2(SiO_3)_2(OH)_2\cdot 5H_2O$ | ≈2 in one analysis | 1 |

TABLE 2.—*Minerals containing thorium as a vicarious constituent, with their varieties and synonyms*—Continued

| *Name* | *Composition* | *Content of ThO₂ (percent)* | *Reference No.* |
|---|---|---|---|
| Vandendriesscheite | $\approx PbO.7UO_3.12H_2O$ | small | |
| Vudyavrite | Altered variety of lovchorrite | $\approx 1$ | 16 |
| Wiikite | Ill-defined mixture | 0 to 3.7 | 2 |
| Xenotime | $Y(PO_4)$ | 0 to 2.5 | 3 |
| Yttrialite | $\approx (Y,Th)_2Si_2O_7$ | $\approx 12$ | 8 |
| Yttrocrasite | $\approx (Y,Th,U,Ca)_2(Ti,Fe,W)_4O_{11}$ | 8.75 in only analysis | 2 |
| Yttrotantalite | $\approx (Fe,Y,U,Ca)(Nb,Ta)O_4$ | $<1$ | 2 |
| Zircon | $Zr(SiO_4)$ | usually $<1$ or none; some varieties higher. | |
| Zirkelite | $\approx (Ca,Fe,Th,U)_2(Ti,Zr)_2O_5$ | 7.3 in only analysis | 2 |

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TABLE 3.—*Ionic radii of certain elements*

[Goldschmidt's values, in kX units]

| *Valence* | *Usual coordination number with oxygen* | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | *4* | | *6* | | *8* | *8 or higher* | | |
| 4 | Si | V | Ti | W | Zr | $Ce^4$ | U | Th |
| | 0.39 | 0.61 | 0.64 | 0.68 | 0.87 | 1.02 | 1.05 | 1.10 |
| 3 | Al | Al | $Fe^3$ | | | Y | La | $Ce^3$ |
| | 0.57 | 0.57 | 0.67 | | | 1.06 | 1.22 | 1.18 |
| 2 | Be | Mg | $Fe^2$ | | | | Ca | |
| | 0.34 | 0.78 | 0.83 | | | | 1.06 | |
| 1 | | | | | | Na | K | |
| | | | | | | 0.98 | 1.33 | |

TABLE 4.—*Tetragonal nesosilicates of Th, U, Ce, and Zr*

| *Thorite group* | *Thorogummite group* |
|---|---|
| $Th(SiO_4)$, thorite | $Th(SiO_4)_{1-x}(OH)_{4x}$, thorogummite. |
| $U(SiO_4)$ [not known] | $U(SiO_4)_{1-x}(OH)_{4x}$, coffinite. |
| $Ce(SiO_4)$ [not known] | $(Ce,Th)(SiO_4)_{1-x}(OH)_{4x}$, freyalite? |
| $Zr(SiO_4)$, zircon | $Zr(SiO_4)_{1-x}(OH)_{4x}$, cyrtolite. |

TABLE 5.—*Minerals containing cerium as an essential or important vicarious constituent (data supplement table 2)*

| *Name* | *Composition* |
|---|---|
| | ESSENTIAL CONSTITUENT |
| Fluocerite | $(Ce,La)F_3$ |
| Tysonite | Synonym of fluocerite. |
| Sahamalite | $(Mg,Fe)(Ce,La)_2(CO_3)_4$ |
| Bastnaesite | $CeFCO_3$ |
| Parisite | $Ce_2Ca(CO_3)_3F_2$ |
| Roentgenite | $Ce_3Ca_2(CO_3)_5F_3$ |
| Synchisite | $CeCa(CO_3)_2F$ |
| Cordylite | $Ce_2Ba(CO_3)_3F_2$ |
| Ancylite | $(Ce,La)_4(Sr,Ca)_3(CO_3)_7(OH)_4\cdot 3H_2O$ |
| Calcio-ancylite | $(Ce,La)_4(Ca,Sr)_3(CO_3)_7(OH)_4\cdot 3H_2O$ |
| Lanthanite | $(La,Ce)_2(CO_3)_3\cdot 8H_2O$ |
| Ambatoarinite | Carbonate of Ce, La, and Sr. |
| Weibyite | Near ancylite in composition. |
| Beiyinite | Ill-defined rare-earth carbonate. |
| Oborite | Ill-defined rare-earth carbonate. |
| Rhabdophane | $(Ce,La,Y)(PO_4)\cdot H_2O$ |
| Florencite | $CeAl_3(PO_4)_2(OH)_6$ |
| Cerite | $(Ce,Ca)_2Si(O,OH)_5$? |
| Lessingite | A calcium-rich variety of cerite. |
| Törnebohmite | Near cerite in composition. |
| Hellandite | $Ca_2(Ce,La,Y,Al,Fe)_6Si_4O_{19}$(?) |
| Cenosite | $Ca_2(Ce,Y)_2Si_4O_{12}(CO_3)\cdot H_2O$ |

TABLE 5.—*Minerals containing cerium as an essential or important vicarious constituent (data supplement table 2)*—Continued

| *Name* | *Composition* |
|---|---|
| | VICARIOUS CONSTITUENT |
| Abukumalite | $(Ca,Y)_5(P,Si)_3O_{12}(OH,F)$ |
| Britholite | $(Na,Ca,Ce)_5(P,Si)_3O_{12}(OH,F)$ |
| Cerorthite | Variety of allanite rich in Ce. |
| Churchite | $Y(PO_4)\cdot 2H_2O$ |
| Codazzite | Cerian variety of dolomite. |
| Erikite | Near abukamalite in composition. |
| Gadolinite | $Be_2FeY_2Si_2O_{10}$ |
| Ishikawaite | $(U,Fe,Y)(Nb,Ta)O_4$ |
| Kalkowskite | $Fe_2Ti_3O_9$ |
| Knopite | Cerian variety of perovskite. |
| Metagadolinite | Ill-defined silicate of Fe and rare earths. |
| Nagatelite | $Ca_2(Ce,La)_2Al_4Fe_2(Si,P)_6O_{25}(OH)$ |
| Weinschenkite | Synonym of churchite. |
| Yttrocerite | Cerian variety of fluorite. |

TABLE 6.—*Minerals containing zirconium as an essential or important vicarious constituent*

| *Name* | *Composition* |
|---|---|
| | ESSENTIAL CONSTITUENT |
| Baddeleyite | $ZrO_2$ |
| Catapleiite | $(Na,Ca)ZrSi_3O_9\cdot 2H_2O$ |
| Dalyite | $K_2ZrSi_6O_{15}$ |
| Elpidite | $Na_2ZrSi_6O_{15}\cdot 3H_2O$ |
| Eucolite | Variety of eudialyte |
| Eudialyte | $Na_4(Ca,Fe)_2ZrSi_6O_{17}(OH,Cl)_2$(?) |
| Guarinite | Na,Ca,Zr fluosilicate (ill-defined) |
| Hiortdahlite | $(Ca,Na)_{13}Zr_3Si_9(O,OH,F)_{38}$(?) |
| Låvenite | $NaCa_2ZrSi_2O_8F$(?) |
| Lovozerite | $(Na,K)_2(Mn,Ca)ZrSi_6O_{16}\cdot 3H_2O$ |
| Loranskite | $(Y,Ce,Ca)_4Zr_3Ta_4O_{22}$(?) |
| Oliveiraite | $Zr_3Ti_2O_{10}\cdot 2H_2O$ (doubtful species) |
| Rosenbuschite | $(Ca,Na)_3(Zr,Ti)Si_2O_8F$(?) |
| Wadeite | $K_2CaZrSi_4O_{12}$ |
| Wöhlerite | $NaCa_2(Zr,Nb)Si_2O_8(O,OH,F)$(?) |
| Zircon | $ZrSiO_4$ |
| Zirkelite | $(Ca,Fe)(Zr,Ti)_2O_5$ |
| Zirfesite | Hydrated silicate of Zr and Fe. |
| | VICARIOUS CONSTITUENT |
| Acmite | Synonym of aergirine |
| Astrophyllite | $(Na,K)_2(Fe,Mn)_4TiSi_4O_{14}(OH)$ |
| Aegirine | $NaFeSi_2O_6$ |
| Betafite | $(U,Ca)(Nb,Ta,Ti)_3O_9\cdot nH_2O$ |
| Beckelite | $Ca_3(Y,Ca,Ce)_4Si_3O_{15}$ |
| Chinglusite | $Na_4Mn_5Ti_3Si_{14}O_{41}\cdot 9H_2O$ |

TABLE 6.—*Minerals containing zirconium as an essential or important vicarious constituent*—Continued

| *Name* | *Composition* |
|---|---|
| VICARIOUS CONSTITUENT—Continued | |
| Fergusonite | (Y,Er,Ce,Fe)(Nb,Ta,Ti)$O_4$ |
| Giannettite | Na,Ca,Mn,Ti,Zr silicate |
| Hainite | Silicate of Na,Ca,Ce,Ti,Zr |
| Lamprophyllite | $Na_2SrTiSi_2O_8$ |
| Murmanite | Na,Ti,Mn,Ca silicate |
| Pennaite | Na,Ca,Mn,Ti,Zr silicate |
| Polymignite | (Ca,Fe,Y)(Nb,Ta,Ti)$O_4$ |
| Pyrochlore | $(Na,Ca)_2(Nb,Ta)_2O_6F$ |
| Samarskite | (Y,Er,Ce,U,Fe,Th)$(Nb,Ta)_2O_6$ |
| Titanocerite | Ti, rare-earth, Zr silicate |
| Yttrotantalite | (Fe,Y,U,Ca)(Nb,Ta,Sn)$O_4$ |

SYNONYMS AND VARIETIES OF ZIRCON AND CYRTOLITE

Alvite. Altered thorian variety of zircon.
Auerbachite. Altered zircon, or cyrtolite.
Adelpholite (part). Altered zircon, or cyrtolite.
Azorite. Synonym of zircon.
Anderbergite. Synonym of cyrtolite?
Beccarite. Synonym of zircon.
Calyptolite. Synonym of zircon.
Cyrtolite. Hydroxyl-substituted variant of zircon.
Engelhardite. Synonym of zircon.
Diocroma. Synonym of zircon.
Hagatelite. Variety of zircon with Nb, Ta, Th, and rare earths.
Jargoon. Synonym of zircon.
Jacinth. Synonym of zircon.
Hyacinth. Synonym of zircon.
Malacon. Altered zircon, or cyrtolite.
Meta-zircon. Altered zircon.
Naegite. Variety of zircon with Th, U, Nb, Ta, and rare earths.
Ostranite. Synonym of zircon.
Polykrasilite. Variety of zircon.
Oyamalite. Variety of zircon with $PO_4$ and rare earths.
Orvillite. Altered zircon, or cyrtolite?
Oerstedtite. Altered zircon, or cyrtolite?
Pseudo-zircon. Altered zircon, or cyrtolite?
Yamagutilite. Variety of zircon with $PO_4$ and rare earths.
Zirconite. Synonym of zircon.
Zirconoid. Altered zircon.
Ribeiraite. Variety of zircon.
Tachyaphaltite. Altered thorian zircon or cyrtolite.

TABLE 7.—*Minerals containing uranium as an essential constituent*

| *Name* | *Composition* |
|---|---|
| OXIDES | |
| Uraninite | $UO_2$ |
| Ianthinite | $2UO_2\cdot 7H_2O$(?) |
| Epi-ianthinite | $\approx UO_3\cdot 2H_2O$ |
| Becquerelite | $7UO_3\cdot H_2O$ |
| Schoepite | $2UO_3\cdot 5H_2O$ |
| Richetite | Hydrated oxide of U and Pb(?) |
| Masuyite | $\approx UO_3\cdot 2H_2O$ |
| Vandendriesscheite | $PbO\cdot 7UO_3\cdot 12H_2O$ |
| Fourmarierite | $PbO\cdot 4UO_3\cdot 7H_2O$ |
| Curite | $3PbO\cdot 8UO_3\cdot 4H_2O$ |
| Billietite | $BaO\cdot 6UO_3\cdot 11H_2O$ |
| Vandenbrandeite | $Cu(UO_2)O_2\cdot 2H_2O$ |
| Uranospherite | $(BiO)(UO_2)(OH)_3$(?) |
| Clarkeite | $(Na,K,Ca,Pb)_2U_2O_7\cdot nH_2O$ |
| CARBONATES | |
| Rutherfordine | $(UO_2)(CO_3)$ |
| Diderichite | Identical with rutherfordine |
| Liebigite | $Ca_2(UO_2)(CO_3)_3\cdot 10\text{–}11H_2O$ |
| Voglite | Ca,Cu, uranyl carbonate(?) |
| Andersonite | $Na_2Ca(UO_2)(CO_3)_3\cdot 6H_2O$ |
| Bayleyite | $Mg_2(UO_2)(CO_3)_3\cdot 18H_2O$ |
| Rabbittite | $Ca_3Mg_3(UO_2)_2(CO_3)_6(OH)_4\cdot 18H_2O$ |
| Swartzite | $CaMg(UO_2)(CO_3)_3\cdot 12H_2O$ |
| Sharpite | $(UO_2)(CO_3)\cdot H_2O$(?) |
| Studtite | Pb, uranyl carbonate(?) |
| Schroeckingerite | $NaCa_3(UO_2)(CO_3)_3(SO_4)F\cdot 10H_2O$ |
| SULFATES | |
| Uranopilite | $(UO_2)_6(SO_4)(OH)_{10}\cdot 12H_2O$ |
| Meta-uranopilite | Doubtful basic uranyl sulfate |
| Zippeite | Near $2UO_3\cdot SO_3\cdot 5H_2O$ |
| Johannite | $Cu(UO_2)_2(SO_4)_2(OH)_2\cdot 6H_2O$ |
| Peligotite | Identical with johannite |
| PHOSPHATES-ARSENATES | |
| Autunite | $Ca(UO_2)_2(PO_4)_2\cdot 8\text{–}12H_2O$ |
| Meta-autunite | $Ca(UO_2)_2(PO_4)_2\cdot 6\text{–}8H_2O$ |
| Torbernite | $Cu(UO_2)_2(PO_4)_2\cdot 12H_2O$ |
| Metatorbernite | $Cu(UO_2)_2(PO_4)_2\cdot 6\text{–}8H_2O$ |
| Zeunerite | $Cu(UO_2)_2(AsO_4)_2\cdot 8\text{–}10H_2O$ |
| Metazeunerite | $Cu(UO_2)_2(AsO_4)_2\cdot 8H_2O$ |
| Uranocircite | $Ba(UO_2)_2(PO_4)_2\cdot 8\text{–}10H_2O$ |
| Meta-uranocircite | $Ba(UO_2)_2(PO_4)_2\cdot 8H_2O$ |
| Saleeite | $Mg(UO_2)_2(PO_4)_2\cdot 8\text{–}10H_2O$ |
| Bassetite | $Fe(UO_2)_2(PO_4)_2\cdot 8H_2O$ |
| Uranospathite | $Cu(UO_2)_2(As,PO_4)_2\cdot 16H_2O$(?) |
| Kahlerite | $Fe(UO_2)_2(AsO_4)_2\cdot 8H_2O$ |
| Novacekite | $Mg(UO_2)_2(AsO_4)_2\cdot 8\text{–}10H_2O$ |
| Metanovacekite | $Mg(UO_2)_2(AsO_4)_2\cdot 8H_2O$ |
| Abernathyite | $K_2(UO_2)_2(AsO_4)_2\cdot 8H_2O$ |
| Troegerite | $H_2(UO_2)_2(AsO_4)_2\cdot 8H_2O$ |
| Uranospinite | $Ca(UO_2)_2(AsO_4)_2\cdot 10H_2O$ |
| Sabugalite | $HAl(UO_2)_4(PO_4)_4\cdot 16H_2O$ |
| Fritzscheite | Doubtful Mn, uranyl phosphate-vanadate |
| Parsonite | $Pb_2(UO_2)(PO_4)_2(OH)_4\cdot 7H_2O$ |
| Renardite | $Pb(UO_2)_4(PO_4)_2(OH)_4\cdot 7H_2O$ |
| Phosphuranylite | $Ca(UO_2)_4(PO_4)_2(OH_4\cdot 7H_2O$ |
| Dewindtite | Doubtful Pb, uranyl phosphate |
| Dumontite | $Pb_2(UO_2)_3(PO_4)_2(OH)_4\cdot 3H_2O$ |
| Walpurgite | $Bi_4(UO_2)(AsO_4)_2\cdot 3H_2O$ |
| VANADATES | |
| Carnotite | $K_2(UO_2)_2(VO_4)_2\cdot 1\text{–}3H_2O$ |
| Tyuyamunite | $Ca(UO_2)_2(VO_4)_2\cdot 7\text{–}11H_2O$ |
| Metatyuyamunite | $Ca(UO_2)(VO_4)_2\cdot 5\text{–}7H_2O$ |
| Sengierite | $Cu(UO_2)(VO_4)(OH)\cdot 4\text{–}5H_2O$ |
| Ferghanite | $U_3(VO_4)_2\cdot 6H_2O$ |
| Rauvite | $CaO\cdot 2UO_3\cdot 2V_2O_5\cdot 16H_2O$(?) |
| Uvanite | $U_2V_6O_{21}\cdot 15H_2O$(?) |
| SILICATES | |
| Uranophane | $Ca(UO_2)_2(SiO_3)_2(OH)_2\cdot 5H_2O$ |
| Sklodowskite | $Mg(UO_2)_2(SiO_3)_2(OH_2\cdot 5H_2O$ |
| Cuprosklodowskite | $Cu(UO_2)_2(SiO_3)_2(OH)_2\cdot 5H_2O$ |
| Beta-uranophane | $Ca(UO_2)_2(SiO_3)_2(OH)_2\cdot 5H_2O$ |
| Soddyite | $(UO_2)_5(SiO_4)_2(OH)_2\cdot 5H_2O$ |
| Kasolite | $Pb(UO_2)(SiO_3)(OH)_2$ |
| Pilbarite | $UO_3\cdot PbO\cdot ThO_2\cdot 2SiO_2\cdot 4H_2O$(?) |
| Coffinite | $U(SiO_4)_{1-x}(OH)_{4x}$ |
| Gastunite | Ca,Pb, uranyl silicate |
| NIOBATE-TANTALATES | |
| Betafite | $(U,Ca)(Nb,Ta,Ti)_3O_9\cdot nH_2O$ |
| Brannerite | $(U,Ca,Fe,Th,Y)_3Ti_5O_{16}$(?) |
| Pisekite | U,Ti,Th, rare-earth niobate-tantalate |
| Delorenzite | $(U,Y,Fe)(Ti,Sn)_3O_8$ |
| MOLYBDATE | |
| Umohoite | $(UO_2)(MoO_4)\cdot 4H_2O$ |

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