

OCCURRENCE OF WILLEMITE

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ABSTRACT

Willemite (Zn_2SiO_4) is often considered, because of its importance at Franklin, N. J., to be characteristic of deposition under conditions of high temperature. By far the largest majority of occurrences, however, including seven new localities, indicate a formation through secondary alteration of ore minerals, often under arid climatic conditions. A number of alternatives are proposed to explain the formation of willemite as a mineral formed under unusual weathering conditions. An attempt is made to find a factor common to both hydrothermal and epithermal deposition, and to call attention to probable other occurrences which have been overlooked because of the inconspicuous appearance of most of the epithermal deposits.

The outstanding importance of willemite as an ore mineral at Franklin, New Jersey, together with the beautiful and varied appearance of the specimens and crystals found there and displayed in most collections throughout the world, has led to a general misconception of the manner of occurrence of this mineral, and of the conditions under which it forms. Willemite is not an uncommon mineral, it has been found in many places, but few collections display or have specimens from localities other than Franklin, New Jersey; Morésnet, Belgium; Salida, Colorado; and sometimes Musartut, Greenland.

This condition exists despite the fact that Spencer (1927) has described four occurrences, Clark (1916) one, Lindgren (1905–1919) three, Foshag (1934) one, Genth (1887) one, LaCroix (1910–13) six, and Pough (1940) one. Franklin specimens are far more showy than those from any other locality, and willemite is more abundant there than elsewhere, so there is some justification for the emphasis placed on the New Jersey occurrence. Furthermore, Franklin willemite is one of the finest minerals known for its display of fluorescence, and that, in this day of fluorescent lights and the popularization of mineralogy's spectacular aspects, has not detracted from the Franklin fame.

Consequently, the remainder of the world's willemite is blanketed by this competition and goes largely unsung. While many minerals reach their apogee at one particular place, such as Japanese stibnite, and epidote from Untersulzbachtal; we also think of Alaskan epidote and of Nevadan or Hungarian stibnite in an afterthought. No mineral needs to have its other localities pointed out so badly, for generally, no such afterthought follows in willemite's train, and this has led to widespread misunderstanding of willemite's place in a genetic series and its usual manner of formation.

TABLE 1

	Locality	Origin	Habit	a 1120	m 1010	Oth- ers	c 0001	e 0112	p 1011	Oth- ers	Remarks	Authority
1	Morésnet, Belgium	Supergene	Prismatic	✓			✓	✓	✓		Small simple crystals	Lévy 1830
2	Franklin, N. J.	Hypogene	Prismatic and Tabular	✓			✓	✓	✓	✓	Very varied in occurrence	
3	Socorro, N. M.	Supergene!	Prismatic	✓			✓	✓	✓	✓	Associated with barite, wulfenite, mimetite, cerussite and quartz	Genth Penfield Penfield 1887 1894 1894
4	Salida, Colorado	?	Tabular	✓			✓	✓				Lindgren and Hillebrand 1904
5	Morenci, Arizona	Supergene	Prismatic	✓			✓	✓	✓	✓	With hemimorphite, diopside and chrysocholla	Hillebrand 1905
6	Musartut, Greenland	Hypogene!	Prismatic	✓			✓	✓	✓	✓	In quartz dike in sandstone	Bøggilo 1911
7	Oslo, Norway	Hypogene	Prismatic	✓			✓	✓	✓	✓	Contact metamorphic deposit. Skarn minerals, zeolites etc.	Goldschmidt 1911
8	Tchicoumba, French Congo	Supergene!	Prismatic	✓			✓	✓	✓		With wulfenite, quartz, Etc.	Lacroix 1910
9	Pimbi, French Congo	Supergene!	Prismatic	✓			✓	✓	✓		With wulfenite, cerussite, Etc.	Lacroix 1910
10	Djoué Mine, French Congo	Supergene!	Prismatic	✓			✓	✓	✓		With cerussite, diopside, Etc.	Lacroix 1913
11	St. Pierre, Martinique	Hypogene	Prismatic	✓							Formed on zinc objects at time of eruption of Mt. Pelé	Lacroix 1910
12	Constantine, Algeria	Supergene	Prismatic	✓				✓			With hematite, hemimorphite, calcite, and malachite	Lacroix 1910
13	Djebel Ressas, Algeria	?	Prismatic					✓				Lacroix 1910
14	Cho-Dan, French Indo-China	?	Prismatic	✓				✓				Lacroix 1913
15	Beaver Co., Utah	Supergene	Prismatic	✓	✓		✓	✓			With hemimorphite, cerussite, mimetite, quartz, and calcite	Clark 1916
16	Broken Hill, No. Rhodesia	?	Prismatic	✓				✓			Over 100,000 tons massive in bones with hematite, limonite, goethite, and quartz	Buttgenbach Spencer 1927
17	Sable Antelope, No. Rhodesia	?	Prismatic	✓				✓			Hematite and Mn oxides	Spencer 1927
18	Lusaka, No. Rhodesia	?	Prismatic					✓				Spencer 1927
19	Guchab, S.W. Africa	Supergene!	Prismatic	✓				✓			Malachite and chalcocite	Spencer 1927
20	Penoles, Durango, Mexico	Supergene!	Prismatic					✓				Spencer 1927
21	Sinaloa, Mexico	Supergene!	Prismatic					✓				Spencer 1927
22	Lubi, Belgian Congo	Supergene!	Prismatic	✓			✓	✓		✓	With diopside, volborthite, barite and cerussite	Buttgenbach 1927
23	Balmat, New York	Supergene	Tabular	✓	✓		✓	✓	✓	✓	Constant in habit and occurrence	Brown 1936

Just as the Franklin willemite is unusual, so is the Franklin occurrence. Let us summarize the described occurrences of willemite given in Table 1. We see there that Franklin, Oslo, and Musartut are probably all the result of high temperature processes. But all the others seem secondary, formed as the result of the alteration of primary zinc sulphide ores, or secondary hemimorphite. This inevitably forces us to the conclusion that hypogene formation of willemite is not customary and that such occurrences are unusual.

That is only a first step however, for we still cannot say that the formation of willemite by alteration processes is the usual procedure, for clearly it is not. Hemimorphite is the common product of the oxidation of sphalerite when siliceous solutions are available; smithsonite when a carbonate radical is present. We still have the problem of the formation of the anhydrous silicate. As shown by innumerable occurrences, under very varied conditions, hemimorphite is the abundant silicate of zinc. It forms under all types of supergene conditions; at Franklin, at Joplin, and in New Mexico. We have then the problem of why willemite should form at all, and what conditions are necessary for its formation.

TABLE 2

	Locality	Source	Habit and Dimensions	Forms	Paragenesis	Remarks
1	Chihuahua, Mexico	U.S.N.M. #95917	Prismatic 2 mm. X .05-.1	<i>a, c</i>	Calcite and pseudomorph after unknown mineral	Flat, platy casts after—?
2	Los Lamentos, Chih., Mexico	U.S.N.M. #95897	Prismatic 1.5-2 X .2-.3	<i>a, c</i>	Calcite, Descloizite	Brown Inclusions—ends clear
3	Santa Eulalia, Mexico	U.S.N.M. 4 Specimens	Prismatic 1-2 X .1-1	<i>a, c, d, e, S, u, V</i>	Hematite, pseudomorph, willemite, fluorite, calcite	Cavernous limonite. Willemite coating cerussite
4	Mina da Prequiça	Harvard #1463	Prismatic 1.5 X 0.3	<i>a, m, c, e</i>	Limonite, cerussite	Cavernous limonite. Willemite coating cerussite
	Sobral da Adiça, Portugal	Harvard #91751	Prismatic 1 X .4	<i>a, m, I, J, c, e, S, V</i>	Limonite, cerussite	Willemite on limonite-coated cerussite
5	Hillsboro, N. M.	A.M.N.H. #21267	Short Prism. 2 X 2	<i>a, c, e</i>	Limonite, cerussite, descloizite	Crystals in cavities in massive willemite
6	Hilltop, Arizona	A.M.N.H. #21263	Prismatic 2 X 1	<i>a, c, e</i>	Cerussite	In cavernous igneous rocks
7	Evergreen, Colo.	A.M.N.H. #21265	Prismatic 2 X 1	<i>a, c, e</i>	Limonite, malachite, azurite	In cavities in fluorite

A large number of specimens from new willemite localities were examined, and they are listed in Table 2. As will be seen, the morphology of these crystals is simpler than that of the Franklin willemite. This is natural, as the conditions under which it crystallized were undoubtedly more uniform than was the case at Franklin, where the willemite shows several generations and, undoubtedly, was formed under a wide range of conditions.

In general, the crystals of these new localities are more or less elongated first order prisms, often terminated simply by the form c , but sometimes showing additional forms such as e , u , and V . A striated prism zone and I and J are not uncommon, but m is frequently lacking. The crystals from these localities are, without exception, small; in most instances ranging up to 2 mm. in length and half that size in diameter. Fluorescence of the supergene willemite is less marked than that of Franklin; under the quartz mercury-vapor lamp it was observed in only one specimen from Mexico (Mina Sieja) and one specimen from the west (Hilltop, Arizona). Fluorescence is reported by Spencer (1927)¹ as being observed on specimens from Rhodesia and on crystals from Lusaka.

The Mexican material was kindly loaned by the United States National Museum. One occurrence has been described by W. F. Foshag (1934). Several different localities are represented in the collection. Four specimens are from the Mina Sieja, Santa Eulalia, Chihuahua. They vary somewhat in appearance, but all four are marked by a reddish coloration caused by associated and included hematite. All are cavernous and show an earlier generation of compact botryoidal willemite with crystal surfaces, and have later growths of larger free crystals with terminations and well-developed forms. The earlier and more compact willemite tends toward redness and opacity, while the succeeding crystals are colorless and clear. There are later crystals of calcite and small cubes of fluorite on two specimens. Red hematite formed early, either contemporaneous with or preceding the willemite; the fluorite and some of the calcite is later than the last of the willemite crystals. One of these specimens of the early botryoidal willemite shows weak fluorescence under the cold-quartz lamp.

Two more specimens from the same district were studied. A specimen from the 7th level of the Ahumada Mine, Los Lamentos (USNM #95897), is a solid mass of loosely intergrown (1.5 to 2 mm. in length) deep red needles of willemite, slightly barrel-shaped with clear ends terminated by c , and 0.2 to 0.3 mm. in diameter. A small amount of calcite and

¹ L. H. Bauer writes that all of his Balmat specimens, and nearly all the other "foreign" specimens, fluoresce in short ultra violet wavelengths, also under a spark gap. In any case, the broad fluorescence band of Franklin material is unlike that of any other locality.

a few small crystals of descloizite have formed after the willemite, while hematite obviously ceased forming before the end of the willemite stage.

A seventh specimen from the Berta Mine, LaCeja District of Chihuahua (USNM #95917), consists of successive crusts of white, flatly radiating willemite. These rosettes probably crystallized upon some platy mineral which has since been dissolved. At the rosette edges are some well terminated colorless crystals, 1.5 to 2 mm. long, and 0.1 to 0.5 mm. in diameter. The only mineral associated with the willemite on this specimen is some calcite which was deposited later.

Two specimens of similarly supergene willemite were obtained from the Harvard collection through the kindness of Dr. Harry Berman. These came from Portugal, the Mina de Prequiça, Sobral da Adiça. They are very different in appearance, and consist principally of limonite crusts, in the cavities of which cerussite has crystallized in fine small twins. In one of the specimens (Harvard #91751) the cerussite has been coated with a thin limonite layer, before the 1 mm. needles of willemite were deposited. These needles show a rather more complex form development than any of the Mexican specimens, *a*, *I*, *J*, and *m* were seen in the prism zone, and *c*, *e*, *V*, and *S* make up the terminations. The second specimen, on which there has been no limonite coating deposited after the cerussite, shows needles with strongly striated prism zones and less complex terminal forms.

Specimens from a few new localities in the United States were also studied, and very similar crystals were found. Two of the localities, Hillsboro, N. M., and Hilltop, Arizona, were discovered by Edwin Over who, with Arthur Montgomery, kindly contributed the specimens. J. W. Adams and R. V. Gaines furnished specimens from a third locality, the Augusta Mine, near Evergreen, Colorado.

The Hillsboro crystals, reaching 2 mm. in length and about the same in diameter, were the largest seen in this study. They are short prismatic to tabular, colorless to white, and well-developed. The specimens are vuggy masses composed of quartz and compact willemite with drusy pockets lined with willemite crystals. These are covered with later cerussite and descloizite crystals. The crystals all show the same few forms, *c*, *e* and *a*, with no indication of *m*. The more prismatic crystals show a greater tendency toward striated prism zones and short trains of reflections. Only the smallest crystals give good reflections, all of the others seem to represent sub-parallel growths and signals are confused. The cerussite, which followed the willemite, has in turn, been coated with a druse of quartz crystals. There is no iron stain on these crystals, nor on the willemite. In general, the iron seems to have oxidized and migrated further, before the willemite formation of these supergene deposits was concluded.

The Hilltop, Arizona, specimens show a vuggy fine-grained igneous rock, on which innumerable white to rose-colored prisms of willemite have crystallized. Associated with the willemite, and later in its period of formation, are small crystals of cerussite, some in minute but beautiful twins. The willemite is more prismatic but otherwise identical in its forms with that of Hillsboro; a striated prism zone all on and near *a*, with no trace of *m*; and *c* and *e* as terminal forms. A little hematite in one place on the specimen seems to have preceded the willemite. These specimens showed a weak fluorescence under the mercury-vapor light.

The last of the new localities, the Augusta Mine, near Evergreen, Colorado, shows identical crystals of about the same size and habit, but black in color. They occur in pockets in a green fluorite, the pockets are clearly the result of the solution of some other mineral. The willemite is associated with azurite, malachite, cerussite, barite, and red and brown stained earthy material. The ubiquitous *a*, *c*, and *e* are the only forms observed with *a* striated as usual.

The new occurrences and their characteristics are summarized in Table 2. We see there that from the standpoint of associated minerals, the manner of occurrence and the description of the specimens, there can be no question but that all of them are secondary and of supergene origin. Their associates are similar minerals, and possibly most significant of all, the willemite crystals resemble those of any other supergene locality.

All mineralogists are familiar with the formation of certain minerals as being characteristic of certain conditions. The frequent occurrence of wulfenite in the arid areas of the West is so generally recognized that collectors are surprised to find it at Phoenixville, Pennsylvania; or Southampton, Massachusetts. Nor does it occur in notable specimens at those localities; undue interest is attracted by the very rarity of the mineral. Willemite is normally not a showy mineral, and if it were to occur there likewise, as it well might, it may have been overlooked. Certain other minerals also belong to this generally recognized group of minerals characteristic of the alteration process in arid climates. Some are diopside, descloizite, vanadinite, even chrysocolla, and less pronouncedly, pyromorphite, anglesite, cerussite, malachite, azurite, smithsonite, brochantite, cerargyrite, stolzite, etc., and many others of more local occurrence. It may be that there is no difference; that what we are referring to as arid climate weathering is merely deep and intense supergene alteration, resulting solely from a low standing water table and long periods of time. This does not seem to be the whole story, however, for aside from the unassailable chlorides, iodates or bromides, there are other minerals, with more common elements, which can be considered as diagnostic.

Often we do not know why diopside forms instead of chrysocolla, why hematite forms when limonite might, or why phenakite crystallizes in place of bertrandite. Surely some special condition is responsible for the formation of that which we have come by experience to recognize as the unusual. The explanation of that condition is not easily determined, nor can a conclusive answer be given. In all probability, several factors have worked together to create willemite-forming conditions, or any one of several different conditions can result in the crystallization of diopside instead of a solidified formless gel of chrysocolla.

A review of Table 1 shows us that in the great majority of cases willemite has formed as the result of sulphide alteration, and furthermore, that in most of those cases the climate at the locality at the present time is arid. In this table the genesis column indicates the origin ascribed by the original author, or a successor, writing about the locality, when no sign (!) follows the origin given. "Supergene!" or "hypogene!" means that no origin is given, but the description and associated minerals indicate that this is a safe assumption. A question mark indicates that the origin is uncertain. The Northern Rhodesian occurrences were originally described as being hypogene, although Spencer (1927) does not commit himself, but the appearance, associations and relationships all make the supergene origin a strong possibility. The associated minerals of the supergene occurrences are usually those which we have recognized as being typical of arid climates, or often found under similar conditions.

Having shown that willemite characteristically forms as a secondary mineral, and is only rarely and in unusual circumstances the result of hydrothermal reactions, we are confronted with the problem of what those conditions are and where we should look and expect to find additional occurrences of willemite. The primary ore is sphalerite everywhere that primary ore has been found in these willemite occurrences, and it can safely be assumed to be the original mineral elsewhere. Siliceous solutions are also necessary, and an oxidizing environment. There are a number of alternative theories which can be called upon to explain the formation of willemite. The occurrence and associations in many cases indicate that it is a mineral characteristic of secondary alteration in a region where arid climates now prevail. Utah, Mexico, New Mexico, Arizona and Portugal are all such regions. On the other hand, we have notable quantities of willemite at Morésnet and Balmat, neither locality is, at present, an arid region. Consequently, some factor which is frequently active where the climate is dry must sometimes be present elsewhere, to explain these occurrences if we assume oxidation under present climatic conditions. We have several alternatives to discuss.

In an arid region with infrequent precipitation it is natural for the oxidation process to exceed the removal of material in solution; this reaches its climax in the Atacama Desert. In moist climates the soluble sulphates are undoubtedly removed as soon as they are formed, and the concentration of salts in solution is never high. In arid climates, on the other hand, oxidation must continue between rains, and leaching take place only for a brief period as the precipitated water slowly sinks through the rocks. Consequently, concentrations must be high. It is possible that this is the controlling factor. Similar conditions of concentrated solutions could occur, due to peculiarities in water passageways, in places with humid climates, such as Balmat, Evergreen, and Morésnet. An alternative which should not be overlooked is that the alteration to willemite may have taken place under very different climatic conditions than those now prevailing. Alternative one is, then, concentrated solutions. This could also function in the hypogene deposits.

Alternative two deals with the composition of the solutions. The oxidation of the associated minerals of the primary ore may result in very different supergene solutions and some undeposited element, or group of elements, may influence the precipitated minerals. Acidity of the solutions, or lack of it, caused by an abundance or scarcity of pyrite in the original deposit, might well influence the mineral formed. Further, an originally acid solution might be more or less neutralized by solutions rich in carbonate compounds descending from overlying rocks. Alternative two has then two phases: one of relative acidity, or of actual differences in elements present. Manganese is frequently associated with willemite and is shown in many analyses; it might conceivably be a critical factor, though this does not seem probable.

Marcasite is a mineral which might be kept in mind as bearing upon both the first two alternatives. Its great ease of oxidation, together with the excess sulphur which it seems to release could be an important item in both the acidity of the solutions and the concentration due to rapid oxidation. It is not uncommon in the Joplin area, where no willemite has yet been reported; and so may have an inhibitive effect upon the formation of that mineral. In abundance it may even affect the next alternative.

Alternative three is one of temperature. Fairly high temperatures may be attained simply through oxidation in a confined space, as the writer has observed in the Missouri sink-hole iron deposits. Such an increase in temperature might easily take place under conditions of rapid oxidation with a relatively small amount of moisture and a slow circulation in a confined area. A rise in temperature might also be brought about by oxidation taking place at a considerable depth. It can readily be seen

that in neither case would there be a very high temperature, but we have no way of knowing what the critical point is between the formation of hemimorphite and willemite. It may be at a comparatively low temperature.

Another factor which may work in conjunction with those above, or alone, is that of pressure. Oxidation taking place at the depth of the water table in arid regions may well be under considerably more pressure than oxidation taking place only a few feet below the surface. This may not be of significance, for spaces which are not filled with water to the surface should be under little greater pressure than similar open spaces at the surface. Water-filled rocks, however, at depths of one or two thousand feet should be under considerably higher pressures and the reactions would take place under conditions different from those prevailing near the surface. It would be logical to expect different products.

The hematite-limonite relationship is closest to the willemite-hemimorphite parallel and it is undoubtedly of significance in any interpretation. It has been suggested that the explanation which may be valid for Franklin (formation of willemite from the metamorphism of hemimorphite), will also hold for other localities and that a dehydration of supergene ores through mild metamorphism is universally responsible. There is said to be evidence of such dehydration of weathered products at Hillsboro, Tres Hermanas, and Beaver County, in addition to Balmat, and it may be asked, why, if arid climate weathering alone is responsible, are there not still more frequent occurrences of willemite. In this case the function of the arid climate is merely that of providing a low water table, permitting deeper supergene alteration than prevails in more humid regions. Postoxidation igneous activity is not marked at most of these new localities. Lindgren (1905) and Foshag (1934), for instance, both speak of the oxidation as if it post-dated any igneous emanations, though their failure to mention it does not, of course, mean that there is none. The authorities describing the regions may have considered such hypogene solutions unimportant, as they would be from a standpoint of ore introduction.

There are doubtless other factors which may be involved, and certainly the problem is not a simple one. Willemite is frequently made synthetically. Morey and Ingerson (1937) list several successes, and it is now being made for the new fluorescent lights by the General Electric Company, but the methods all involve relatively high temperatures. These successes in synthesis thus in no way contribute to the problem of its slow formation in nature at low temperatures. A few facts about the occurrences stand out which are of interest, worthy of note, and possibly of value in a final interpretation.

Willemite formed under conditions of secondary alteration is uniform in size and crystal habit, in its paragenesis, its distribution and its appearance. This would indicate its formation under essentially similar conditions. We know that it can also form under abnormal conditions of high temperature from hypogene solutions. Then it varies in those qualities previously listed for supergene willemite. Hematite is a more frequent associate of willemite, limonite more common with hemimorphite. These facts we know, the rest we shall have to work out.

Of all of the alternatives proposed, the writer believes that the most important is that of concentration of solutions. Willemite may be the characteristic precipitate from saturated solutions, hemimorphite the product of weak solutions. Undoubtedly, other factors play their part, but in all the phenomena of mineral formation in arid climates we find one thing, a local concentration of elements usually widely disseminated in small quantities. Wulfenite and vanadinite, cerargyrite and desclozite are typical of arid regions and of this statement. All represent a local congestion of normally disseminated elements and it is not improbable that willemite, their frequent associate, is another manifestation of the same processes.

The alternative suggestion is well worth considering, though we still have the problem of the cause of the mild metamorphism. If it is correct, we may expect to find additional occurrences of willemite where extensive pre-Cambrian oxidation of zinc sulphide has been preserved and later metamorphosed, or in regions near centers of volcanic activity in more recent times, giving rise to local metamorphism of the secondary products of oxidation.

But the problem is not a simple one which can be solved by rationalizing at a desk or in a laboratory. Let us examine more occurrences in greater detail and let us learn more about why hematite forms; let us examine zinc deposits, especially in dry climates with our eyes open, and we may find the inconspicuous, but interesting willemite. When we have done that, we may gain a conception, not only of the processes by which willemite forms, but also of the entire problem of supergene alteration and secondary mineral formation. Then, too, we can think about wulfenite and diopase, vanadinite and brochantite, gems of the collectors' cabinet but still mysteries to the mineralogist.

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