

Carlinite, Tl_2S , a New Mineral from Nevada

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Abstract

Carlinite, Tl_2S , occurs as small subhedral to anhedral grains with fine-grained quartz and hydrocarbon compounds in carbonaceous limestones exposed in the east pit of the Carlin mine, Nevada. Fresh carlinite is dark gray with a bright metallic luster and a dark gray to black streak. Carlinite rapidly oxidizes in air, causing exposed surfaces to become dull and darker in color. The crystallographic parameters of carlinite are: rhombohedral, space group $R\bar{3}$, $a = 12.12 \pm 0.01 \text{ \AA}$, $c = 18.175 \pm 0.005 \text{ \AA}$, $Z = 27$, cell volume = $2310.5 \pm 0.6 \text{ \AA}^3$. Carlinite displays perfect (0001) cleavage, an imperfect prismatic cleavage, and hackly fracture. The strongest X-ray diffraction peaks and relative intensities are: 3.030 \AA , 1000; 2.290 \AA , 33; 1.749 \AA , 30; 2.020 \AA , 28. Powdered carlinite tends to show extreme preferred orientation because of the perfect basal cleavage. Vickers hardness is $23.5 \pm 2.0 \text{ kg/mm}^2$; Mohs hardness, 1; density, $8.1 \pm 0.2 \text{ g/cm}^3$ (meas), 8.55 g/cm^3 (calc). In reflected light, carlinite is "galena white" with a faint bluish cast, weakly birefractant, and moderately anisotropic, with polarization colors that range from brownish-gray to bluish-gray. Reflectances in air are: $R_{650nm} = 40.5 - 42.0$; $R_{689nm} = 39.2 - 40.6$; $R_{646nm} = 38.8 - 40.1$ and $R_{470nm} = 39.6 - 41.3$. Carlinite is essentially pure Tl_2S ; microprobe analysis yielded $Tl = 92.93$ and $S = 7.17$, in wt percent. The most abundant trace element, by emission spectrographic analysis, is As at levels less than 100 ppm. The new mineral is named after the Carlin gold deposit in which it was discovered.

Introduction

A new mineral, carlinite, with composition Tl_2S , was discovered at the Carlin deposit, northern Eureka County, Nevada, as an outgrowth of investigations of the genesis of epithermal gold deposits in Nevada and Utah (Radtke and Dickson, 1975). Some Carlin-type gold deposits previously had been reported to contain abnormally large amounts of dispersed thallium in unknown forms (Weissberg, 1969; Radtke *et al.*, 1972; Radtke, Dickson, and Rytuba, 1974); however, only at the Carlin mine have thallium-bearing minerals been described. These include lorandite, $TlAs_2S_2$ (Radtke *et al.*, 1973), and orpiment, As_2S_3 , that contains 0.1 to 0.35 wt percent thallium in solid solution (Radtke *et al.*, 1974).

Crystalline Tl_2S , synthesized during studies of the system Tl_2S - As_2S_3 in the Hydrothermal Laboratory at Stanford University, has properties consistent with its possible occurrence as a mineral in thallium-

bearing rocks which contain reducing substances, such as hydrocarbons, and which also have a low arsenic content. Subsequent X-ray and microprobe examination of heavy mineral concentrates from carbonaceous, high-thallium, low-arsenic, silicified limestone from the Carlin deposit demonstrated Tl_2S to be present.

The Carlin gold deposit is the largest and best known of the disseminated, replacement-type gold deposits. The mineral Tl_2S is named "carlinite" after the deposit in which it was found. The mineral name and designation as a new mineral have been approved by the Commission of New Minerals and New Mineral Names of the International Mineralogical Association.

Type material is deposited in the Smithsonian Institution (U.S. National Museum), Washington, D. C., and in the Epithermal Minerals Collection, Department of Geology, Stanford University, Stanford, California.

Occurrence

Carlinite occurs as small grains randomly scattered through black brecciated fragments of carbonaceous limestone in shear zones exposed in the east pit of the Carlin gold deposit. The geology of the mine and the sample location are shown on the geologic map of the Carlin deposit by Radtke (1973). The samples that contain carlinite come from the 6440 bench, at an approximate elevation of 6445 feet, at mine coordinates 23,050 N., 19,900 E.

Gold mineralization at the Carlin deposit affected at least the upper 300 meters of siliceous, argillaceous, dolomitic limestones of the Silurian Roberts Mountains Formation. Hydrothermal solutions dissolved calcite and lesser amounts of dolomite, and deposited silica, barium, gold, mercury, arsenic, antimony, and other metals (Radtke and Scheiner, 1970). The presence of thallium at Carlin was first reported by Radtke *et al* (1972). However, Weissberg (1969), who found thallium, gold, arsenic, antimony, and mercury in precipitates from hot springs in New Zealand, had originally pointed to the tendency for these elements to occur in disseminated gold deposits of the western United States.

The limestones of the Roberts Mountains Formation at the Carlin deposit normally contain about 0.3 wt percent organic carbon, but the rocks which contain carlinite have as much as 6 percent organic carbon (A. S. Radtke, unpublished data). B. J. Scheiner of the U.S. Bureau of Mines (oral communication, 1974) reported that organic compounds in such high carbon material were mostly mixtures of hydrocarbons and "humic acid." These petroleum-

like carbon compounds apparently moved into the mineralized rocks along permeable zones during the hydrothermal episode (Radtke and Dickson, 1975).

No other hydrothermally deposited sulfide minerals were found with carlinite in the samples studied. Hand specimens with carlinite contained very small amounts of gold, arsenic, antimony, and mercury. The original limestone host-rock contained calcite, dolomite, illite, quartz, and pyrite. The highly concentrated organic materials and some of the quartz appear to be genetically associated with carlinite.

Physical and Optical Properties

Carlinite occurs as small subhedral to anhedral grains ranging from about 0.005 mm to 0.5 mm in diameter. Most grains are irregular and slightly rounded with a few grains showing poorly defined rhombohedral and tabular forms (Fig. 1).

The new mineral has perfect (0001) cleavage and an imperfect prismatic cleavage or fracture. Vickers hardness of carlinite, determined with a Leitz hardness indenter using a 50-gram load, ranged from 20.1 to 23.8 and averaged 23.5 kg/mm² (5 determinations); the hardness of synthetic Tl₂S, measured similarly, ranged from 22.6 to 24.8 and averaged 24.5 kg/mm² (5 determinations). The average hardness of both carlinite and synthetic Tl₂S is about 1 on the Mohs hardness scale. The mineral is soft and deforms easily. This, together with the perfect basal cleavage, introduces difficulties in grinding; we were not able to prevent strong preferred orientation in the powdered material. The density of carlinite, measured with a Berman balance, is 8.1 ± 0.2 g cm⁻³; the calculated density is 8.55 g cm⁻³. Ketelaar and Gorter (1939) reported a density for synthetic Tl₂S of 8.39 g cm⁻³.

Differential thermal analysis of carlinite showed that no phase changes take place with increasing temperature until melting occurs at 449 ± 5°C and 1 bar. The melting temperature of carlinite agrees well with the melting temperature for Tl₂S of 448°C (Pélabon, 1907) and a temperature of 448.5°C reported by later workers (Trotman-Dickenson, 1973). Physical properties of synthetic Tl₂S, summarized by Trotman-Dickenson (1973), are given in Table 1.

Carlinite is dark gray with metallic luster, but fresh surfaces become darker gray to black, even dull or earthy, because of surface oxidation. The streak of fresh carlinite is dark gray to black. In reflected light in air, carlinite is galena white with a faint bluish cast. Bireflectance is generally weak but is slightly stronger along grain boundaries. Carlinite is moderately to distinctly anisotropic with polarization colors vary-

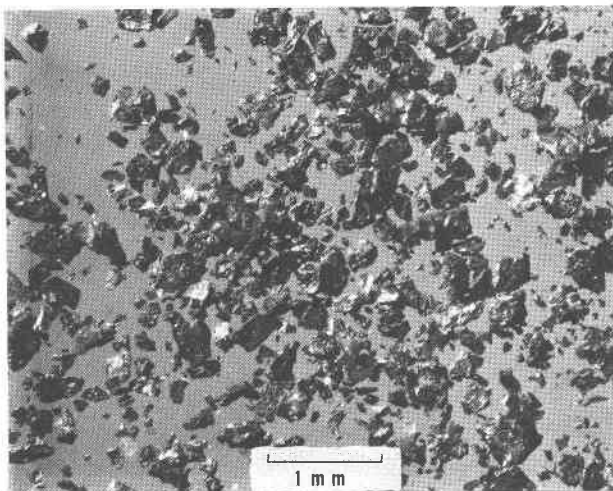


FIG. 1. Carlinite from the Carlin gold deposit (opaque grains), with some quartz (transparent grains).

TABLE 1. Physical Properties of Synthetic Tl₂S
(Compiled by Trotman-Dickenson, 1973)

Property	Value
Density	8.39 g/ml
Melting Point	448.5°C
Heat of Fusion	3 kcal/mole
ΔH _f ^o (25°C)	-23.2 kcal/mole
ΔG _f ^o (25°C)	-22.39 (±.12) kcal/mole
Electrical Conductivity	
At melting point	1.75 ohm ⁻¹ cm ⁻¹
At 771°C	33.8 ohm ⁻¹ cm ⁻¹
Solubility, 20°C	
In water	0.215g Tl ₂ S/liter
In 0.02 M H ₂ SO ₄	1.451g Tl ₂ S/liter

TABLE 3. X-ray Powder Diffraction Data for Carlinite and Synthetic Tl₂S

hkl	Carlinite			Synthetic Tl ₂ S	
	Calculated* d _{hkl} ^o Å	Observed** d _{hkl} ^o Å	I***	Observed** d _{hkl} ^o Å	I***
101	9.087	---	---	---	---
012	6.869	---	---	---	---
003	6.058	---	---	---	---
110	6.058	---	---	---	---
021	5.040	---	---	---	---
202	4.543	---	---	---	---
113	4.283	---	---	---	---
104	4.170	---	---	---	---
211	3.875	---	---	---	---
122	3.635	---	---	---	---
300	3.497	3.497	9	3.503	3
015	3.435}	3.431	9	3.433	9
024	3.435}				
006	3.029}				
303	3.029}	3.030	1000	3.030	1000
220	3.029}				
205	2.988	---	---	---	---
214	2.988	---	---	---	---
131	2.873	---	---	---	---
312	2.771	---	---	---	---
116	2.709}	2.712	12	2.710	6
223	2.709}				
125	2.680	---	---	---	---
401	2.596	---	---	---	---
107	2.520}	---	---	2.520	3
042	2.520}				
134	2.451	---	---	---	---
321	2.386	---	---	---	---
027	2.327}	---	---	2.323	4
232	2.327}				
306	2.290}	2.290	33	2.292	25
410	2.290}				
315	2.272	---	---	---	---
404	2.272	---	---	---	---
018	2.220	---	---	---	---
217	2.172	---	---	---	---
226	2.142	---	---	---	---
413	2.142	---	---	---	---
045	2.127	---	---	---	---
324	2.127	---	---	---	---
208	2.084	---	---	---	---
051	2.084	---	---	---	---
502	2.045	---	---	---	---
009	2.019}	2.020	28	2.019	39
330	2.019}				
235	2.007	---	---	---	---
128	1.971}	---	---	1.973	3
241	1.971}				
137	1.937	---	---	---	---
422	1.937	---	---	---	---
119	1.916}	1.918	3	1.919	4
333	1.916}				
054	1.905	---	---	---	---
511	1.874	---	---	---	---
407	1.845	---	---	---	---
152	1.845	---	---	---	---
416	1.827	---	---	1.831	2
505	1.817}	---	---	1.819	2
244	1.817}				
1.0.10	1.791}	1.792	7	1.792	12
318	1.791}				
327	1.765	---	---	---	---
309	1.749}	1.749	30	1.749	33
600	1.749}				
229	1.680}	1.681	16	1.683	6
336	1.680}				
603	1.680}				
520	1.680}				
2.1.10	1.652}	1.654	6	1.652	2
238	1.652}				

ing from brownish-gray to bluish-gray. In oil, the bireflectance is very faint to very weak, and polarization colors are similar to those in air. Optical properties of synthetic Tl₂S, including color, bireflectance, anisotropism, and polarization colors correspond with those of carlinite.

Reflectivity data for carlinite and synthetic Tl₂S in air are given in Table 2.

Crystallography

X-ray powder diffraction data for carlinite and synthetic Tl₂S, using quartz as an internal standard, are given in Table 3; except for the strong intensity of the combined 006, 303, 200 reflections caused by preferred orientation effects, our data agree well with published data for synthetic Tl₂S (XPDF Card #6-0378: Reuter and Goebel, 1953).

To prevent the rapid oxidation and the formation of coatings of crystalline alteration products on the surfaces of powdered Tl₂S grains, the samples of both carlinite and synthetic Tl₂S were ground under acetone in an agate mortar. The suspension was then sedimented on glass plates under acetone that contained a small amount of acetone-soluble "Duco" cement.

X-ray powder diffraction data for carlinite were refined and the unit cell dimensions were determined

TABLE 2. Reflectivities of Carlinite and Synthetic Tl₂S

Phase	Wavelength (nm)			
	470	546	589	650
Carlinite	39.6-41.3	38.8-40.1	39.2-40.6	40.5-42.0
Synthetic Tl ₂ S	39.8-41.1	38.6-40.0	39.3-40.8	39.8-41.7

TABLE 3, Continued

<i>hkl</i>	Carlinite		<i>I</i> ***	Synthetic Tl ₂ S	
	Calculated*	Observed**		Observed**	<i>I</i> ***
	$d_{hkl}^{\text{Å}}$	$d_{hkl}^{\text{Å}}$		$d_{hkl}^{\text{Å}}$	
0.1.11 057	1.632 1.632	1.634	8	1.633	13
0.0.12 419 606 440	1.515 1.515 1.514 1.514				
4.0.10 428 351	1.494 1.494 1.494	1.491	3	1.490	1
3.0.12 710	1.390 1.390				
0.5.10 348 529 633	1.374 1.374 1.292 1.292	1.372	2	1.374	3
4.1.12 716	1.263 1.263				
0.0.15 3.3.12 449 636 550	1.212 1.212 1.212 1.212 1.212	1.212	8	1.212	13
1.6.10 461	1.201 1.201				
3.4.11 547 642	1.193 1.193 1.193	1.194	5	---	---
3.0.15 6.0.12 719 903 820	1.145 1.145 1.145 1.145 1.145				
5.4.10 188 561	1.080 1.080 0.9156	1.081	8	1.079	17
1.3.19 3.4.17 10.0.10 488 491	0.9087 0.9087 0.9087 0.9087 0.9087				
0.1.20 3.5.16 1.11.2 4.0.19 9.2.10	0.9054 0.9053 0.9053 0.8987 0.8986	0.9053	5	0.9053	13
3.2.19 6.1.17 10.3.1	0.8890 0.8890 0.8889				

*All calculated *hkl*'s listed for $d_{hkl} \geq 1.745 \text{ Å}$. All observed *hkl*'s $d_{hkl} > 0.8889 \text{ Å}$ are indexed, Indices and *d*(Calc) from the least-squares analysis of X-ray powder data using the digital computer program of Appleman and Evans (1973); and a hexagonal cell with $a = 12.12$ and $c = 18.175 \text{ Å}$.

**X-ray diffractometer conditions are: Ni-filtered Cu radiation; $\text{CuK}\alpha = 1.54178 \text{ Å}$; SiO_2 used as internal standard; scanned at $1^\circ 2\theta$ per minute.

***Strongest line is assigned intensity of 1000 to permit assignment of larger numbers as the relative intensities of other lines.

by the use of the cell parameter least-squares program of Appleman and Evans (1973). The rhombohedral unit cell dimensions for carlinite are $a = 12.12 \pm 0.01 \text{ Å}$; $c = 18.175 \pm 0.005 \text{ Å}$; and $V = 2310.5 \pm 0.6 \text{ Å}^3$. The cell parameters compare well with the values of $a = 12.20 \pm 0.07 \text{ Å}$ and $c = 18.17 \pm 0.06 \text{ Å}$ for synthetic Tl₂S reported by Ketelaar and Gorter (1939).

Ketelaar and Gorter (1939) established that synthetic Tl₂S is rhombohedral with space group *R*3, and $Z = 27$. Because of the close agreement between the crystallographic parameters of carlinite and those of synthetic Tl₂S reported by Ketelaar and Gorter (1939), carlinite is assumed to be isostructural with synthetic Tl₂S. The structure may be regarded as a distorted hexagonal closest-packed arrangement of thallium atoms, in which the octahedral interstices between pairs of planes of thallium atoms are occupied by sulfur atoms.

Chemical Composition

Carlinite is Tl₂S with only low levels of other constituents. Results of quantitative chemical analyses of three grains done with the electron microprobe, together with the analytical conditions used, are shown in Table 4. Semiquantitative emission spectrographic analysis of a composite sample of several large hand-picked grains of carlinite show that the mineral may contain small amounts of arsenic but at an abundance of <100 ppm As (Table 5).

Synthesis

Synthetic carlinite can be made by reacting elemental thallium and sulfur in stoichiometric proportions in sealed evacuated pyrex tubes at temperatures ranging from 200°C to 400°C (Carstansen, 1867). Others have prepared Tl₂S by reacting Tl and S in an atmosphere of H₂ (Ketelaar and Gorter, 1939). Generally, the product of the reaction below the melting temperature is fine-grained, porous, and dark-gray, without visible individual crystals. Small droplets of melted Tl₂S, cooled slowly through the freezing temperature at about 10°C per hour, tend to

TABLE 4. Microprobe Chemical Analyses of Carlinite

Grain No.	Weight Percent		
	Tl	S	Total
1	92.95	7.24	100.19
2	93.14	7.19	100.33
3	92.69	7.07	99.76
Ave.	92.93	7.17	100.09
Tl ₂ S	92.73	7.27	100.00

Analytical conditions: (1) Thallium, Mg characteristic line, ADP crystal, 18 kV, synthetic Tl₂S standard; (2) sulfur, K α characteristic line, ADP crystal, 18 kV, synthetic Tl₂S standard.

develop metallic hemispherical single crystals of a lighter dark-gray color with one set of parallel faces, creating the appearance of round domes with circular steps (Fig. 2).

Carlinites precipitates from alkaline solutions of thallos salts on reaction with H_2S (Reuter and Goebel, 1953; Pascal, 1961), but care must be taken to exclude oxygen. Fine, reactive, oxygen-free Tl_2S can be made by reacting thallium ethoxide in ethanol solution, with dry H_2S (Brauer, 1963).

Commercially available " Tl_2S " that we have examined has been poorly crystalline and impure. Some of the impurities are capable of developing high gas pressures in closed tubes at high temperatures. Caution should be exercised in using commercial material for experimental purposes.

Chemical Behavior

Carlinites has the highest reported solubility in aqueous solution of any naturally occurring heavy metal sulfide, 215 ppm Tl_2S at 20°C and 1 bar (Kivalo and Kurkela, 1959; Trotman-Dickenson, 1973). Carlinites reacts with water, as follows: $Tl_2S + H_2O = 2 Tl^+ + HS^- + OH^-$. Therefore, a solution saturated with Tl_2S is basic, with a calculated pH of about 10.7. The alkaline reaction of Tl_2S with water resembles the reactions of Na_2S or K_2S , in keeping with the tendency of thallos compounds to resemble alkali metal compounds in chemical behavior (Songina, 1964). Tl_2S solubility is higher in acid solutions; for example, a solution 0.02 M/l in H_2SO_4 contains 1,451 ppm Tl_2S (Table 1). The reaction with H^+ is: $Tl_2S + 2 H^+ = H_2S + 2 Tl^+$.

Phase relations between Tl_2S and other crystalline

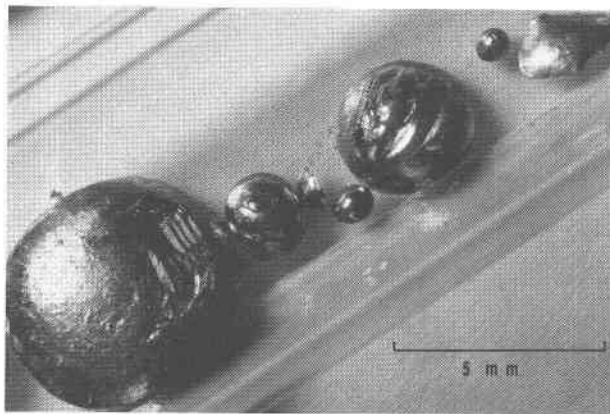


FIG. 2. Crystalline synthetic Tl_2S produced by slow cooling of melt. Note that some small droplets formed as single crystals bounded incompletely by crystal faces.

solids in the system $Tl-S$ have not been studied systematically. Compounds with various Tl/S ratios have been characterized: Tl_2S , Tl_4S_3 , TlS , TlS_2 , and Tl_2S_5 (Hahn and Klinger, 1949; Lee, 1971). Carstangen (1867), who first studied Tl_2S , prepared an amorphous solid with the composition Tl_2S_3 that decomposed on heating *in vacuo* to Tl_2S plus S .

Carlinites oxidizes rapidly on exposure to the atmosphere; oxidation on polished surfaces takes place during microscopic examination over a matter of 30 minutes. Carlinites grains up to 0.2 mm in diameter, separated from the host rock and kept in a glass vial at room conditions for 3 months, oxidized completely.

The oxidation of Tl_2S has been studied by several workers (Fentress and Selwood, 1948; Reuter and Goebel, 1953). Lee (1971) summarized the reactions involved. The reaction with O_2 at room temperature proceeds stepwise, and various oxides and sulfur-oxygen compounds are produced. According to Lee (1971) the reactions that are most likely in the oxidation of carlinites under natural conditions are: first, $2 Tl_2S + 2 O_2 \rightarrow Tl_2S_2O_3 + Tl_2O$; and second, $Tl_2O + O_2 \rightarrow Tl_2O_3$. Given sufficient time, under dry conditions the overall reaction would be: $2 Tl_2S + 3 O_2 \rightarrow Tl_2S_2O_3 + Tl_2O_3$. With water present, thallos thiosulfate degrades according to the reaction: $Tl_2S_2O_3 + H_2O \rightarrow Tl_2SO_4 + H_2S$. Thallos sulfate, thiosulfate, and oxide are more soluble than Tl_2O_3 ; therefore in nature, where oxidation takes place in contact with aqueous solutions, they would tend to be leached, leaving Tl_2O_3 . A rare mineral, avicennite, with a composition of $7 Tl_2O_3 \cdot Fe_2O_3$ has been reported from Russia (Karpova *et al.*, 1958); it is not

TABLE 5. Semiquantitative Emission Spectrographic Analyses of Carlinites

Element	Weight percent
Fe	0.001
Ag	0.0001
As	<0.01
Co	0.0002
Cu	0.0003
Ni	0.0001
Tl	0.0002
Tl	>50.

Analysis of several large grains of carlinites. [Sought but not found: Al, Au, B, Ba, Be, Bi, Ca, Cd, Ce, Cr, Eu, Ga, Ge, Hf, In, K, La, Li, Mg, Mo, Mn, Na, Nb, P, Pb, Pd, Pt, Re, Sb, Sc, Si, Sn, Sr, Ta, Te, Th, U, V, W, Y, Yb, Zn, Zr.]

known whether avicennite is primary or whether it formed by oxidation of primary Tl_2S .

Thallium compounds are highly poisonous (Songina, 1964, p. 319); the potential hazard of carlinite is greater than with other metallic sulfides because of the rapidity with which Tl_2S oxidizes to relatively highly soluble compounds such as Tl_2O . Grains of carlinite will develop poisonous coatings on exposure to air, and we caution that they should be handled carefully, keeping this hazard in mind.

Geochemical Implications

Thallium commonly accompanies arsenic, antimony, and mercury in Carlin-type epithermal gold deposits (Weissberg, 1969; Radtke and Dickson, 1974). Only at the Carlin deposit is thallium known to occur in discrete minerals with high thallium content (Radtke *et al*, 1973; Radtke *et al*, 1974). In other deposits as well as at Carlin, thallium is in dispersed forms in carbonate rocks in and close to gold ores.

Thallium readily forms complex sulfides with other metals such as As, Sb, Ag, Pb, and Cu. Examples of Tl-containing complex sulfide mineral assemblages are the classic Lengenbach deposit in the Binnatal district, Switzerland, described in a special volume published by the Natural History Museum, Bern, by Stalder *et al* (1966–1968), Nowacki and Bahezre (1963), Nowacki *et al* (1967) and Graeser (1967), and the Jas Roux deposit, France (Johan *et al*, 1974).

For carlinite to form and to persist requires an unusual set of conditions: Tl must be in solution in unusually high concentration; other elements with which Tl readily combines must be essentially absent; a mechanism must exist that prevents dissolution; and the environment must be reducing during and after deposition.

Most solutions of natural systems are probably low in thallium. Because carlinite is so soluble in aqueous solutions, special mechanisms are required to create supersaturation and to retain carlinite once it has formed. Nothing is known of the solubility of carlinite in aqueous solutions as a function of temperature and pressure, and the extent to which deep-seated solutions might become supersaturated with carlinite on approaching the surface cannot be evaluated. However, some evidence supports the conclusion that at one stage in the formation of the Carlin deposit the hydrothermal solution boiled (Radtke and Dickson, 1974). Loss of water by boiling solutions initially undersaturated with carlinite would be an effective mechanism for increasing Tl_2S concentration to saturation. Alternatively, the possibility that sulfur-

poor, Tl-bearing solutions encountered S-bearing solutions cannot be ruled out. After carlinite was deposited, it could not have been exposed very long to the action of flowing water without dissolving. Therefore, the existence of carlinite at Carlin attests to the effectiveness with which carlinite grains were protected from aqueous solutions, or possibly, to the short time that has lapsed since formation.

As mentioned earlier, carlinite rapidly oxidizes when removed from the host rocks and exposed to air. In the host rocks the mineral is completely enclosed in a matrix containing as much as 6 percent organic carbon compounds, which act as reducing agents and which prevent the oxidation of Tl_2S .

Acknowledgments

We thank Dr. Charles M. Taylor, Mr. John Slack, and Miss Jo Peterson, of Stanford University, for their valuable aid in some aspects of this study. The manuscript was greatly improved by critical reviews by Professors Gordon E. Brown and Konrad K. Krauskopf of Stanford University, and Wayne E. Hall and Richard C. Erd of the U.S. Geological Survey. The senior author wishes to give special thanks to Mr. Dan Higley of Carlin Gold Mining Company for his assistance and help in obtaining samples from inaccessible areas in the east pit of the mine.

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Manuscript received, January 17, 1975; accepted for publication, March 5, 1975.