Ellisite, T1₃AsS₃, a new mineral from the Carlin gold deposit, Nevada, and associated sulfide and sulfosalt minerals

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Abstract

Ellisite, Tl₃AsS₃, occurs with lorandite, getchellite, christite, realgar, and native arsenic as dispersed grains, and with realgar, lorandite, christite, and getchellite in small discontinuous patches, in mineralized laminated carbonaceous dolomite beds in the Carlin gold deposit, Nevada. The mineral is named for Dr. A. J. Ellis of the Chemistry Division, D.S.I.R., New Zealand. Ellisite is opaque, and the color is dark gray, streak is light brown with a tinge of orange, and luster on fresh surfaces is metallic but less bright than that of stibnite. The mineral is trigonal, hexagonal parameters a = 12.324(2), c = 9.647(2)A; cell volume = 1269.0(4)A³, Z = 7. The d values (A) of the strongest X-ray diffraction lines and their relative intensities are: 2.669 (100), 3.214 (53), 5.333 (37), 2.327 (28), and 3.559 (20). Ellisite shows excellent to good rhombohedral cleavage and hackly fracture. Vickers hardness measured with a 50-g load varied from 36.4-44.0 and averaged 39.3 kg mm⁻², and with a 25-g load varied from 41.0-51.5 and averaged 45.9 kg mm⁻² (Mohs hardness, about 2). Density of synthetic Tl₃AsS₃ is 7.10(5) (meas) and 7.18 g cm⁻³ (calc). In reflected light the mineral is light gray with a distinct purple tint (light purplish-gray), very weakly bireflectant, ranging from light purplish-gray to light pinkish-gray, and anisotropic, with polarization colors blue-purple, red-purple, and brownish-orange; the mineral has a deep red to deep orange-red internal reflection. Reflectances (R) in air are: 650nm = 28.3, 589nm = 28.9, 546nm = 29.7, 470nm = 31.7. Electron microprobe analyses gave T1 78.2, As 9.6, S 12.3, sum 100.1 weight percent. Most abundant trace elements, determined by emission spectrographic analysis, are Sb (500 ppm) and Fe (200 ppm).

Disseminated assemblages include: the gold-pyrite-quartz in the ore bodies; the thalliumarsenic assemblage of ellisite-christite-lorandite-getchellite-realgar-native arsenic-quartz; the mercury assemblage of galkhaite-cinnabar-quartz; and the carlinite-pyrite-hydrocarbon-quartz assemblage. Vein mineral assemblages which crosscut rocks, ores, and disseminated zones include christite-lorandite-orpiment-realgar-barite-quartz, weissbergite-stibnite-quartz, and frankdicksonite-quartz. Veins are distinct and separate from each other and postdate the ore and zones of disseminated sulfides. Gold ore bodies formed from solutions at temperatures between 160–180°C, but many minerals in late veins formed at temperatures above 200°C. Most of the disseminated sulfide and sulfosalt minerals probably formed during the late stages of the more moderate temperature event and the vein minerals were deposited from hotter fluids (200–300°C) in response to supersaturation caused by falling temperatures and pressures of the fluids during ascent.

Introduction

The new mineral ellisite, of composition Tl_3AsS_3 , belongs to a suite of minerals containing thallium, arsenic, and sulfur occurring in the Carlin gold deposit, Eureka County, Nevada. Ellisite is named for Dr. A. J. Ellis of the Chemistry Division, D.S.I.R., New Zealand, in recognition of his many contributions in the field of hydrothermal geochemistry. The mineral

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Fig. 1. Scanning electron microscope photograph of ellisite grains showing rhombohedral cleavage fragments.

name and designation as a new mineral have been approved by the Commission on New Minerals and Mineral Names of the International Mineralogical Association.

Type material in very limited amount is deposited in the Epithermal Minerals Collection, Department of Geology, Stanford University, Stanford, California.

Occurrence

Ellisite occurs in mineralized, laminated, argillaceous, carbonaceous dolomite beds of the Roberts Mountains Formation in the East ore zone of the Carlin gold deposit. Samples containing ellisite have been found in two places. The first is in an area at the southwest end of the East ore zone near mine coordinates 22,900 N, 19,800 E, benches 6380 to 6400. The second is in the central part of the East ore zone between mine coordinates 23,100 N, 20,300 E, to 23,500 N, 20,700 E, between benches 6340 and 6350 (Radtke, 1973).

In both areas dispersed grains of ellisite are spatially associated with lorandite, getchellite, christite, and realgar. Prior to this last stage of mineralization, hydrothermal quartz and pyrite replaced calcite, and

Table I. Reflectances of ellisite, in percent

	Wavelength (nm)			
	470	546	589	650
In air	31.7	29.7	28.9	28.3
In oil	17.5	16.2	15.3	14.9

kaolinite and sericite formed in the matrix of the carbonate host rock. The rocks also contain up to 6 weight percent organic carbon which was introduced by hydrothermal fluids.

Both areas also contain veinlets, seams, and irregular patches, normal and parallel to bedding planes in the laminated host rock, made up of ellisite, abundant realgar, small amounts of lorandite, christite, and getchellite. The veinlets and patches cut the mineralized rocks, indicating that they were later than the main hydrothermal stage of gold mineralization (Radtke and Dickson, 1974; Dickson *et al.*, 1978).

Physical and optical properties

Most of the grains of ellisite were anhedral to irregular, and ranged from about 0.5 to 1.3 mm in length; however, several broken grains showed good rhombohedral shapes (Fig. 1). Rhombohedral forms were also observed on crystals of Tl_3AsS_3 synthesized from melts by slow cooling through the freezing temperature (325°C) followed by holding at 310°C for three weeks. Small grains enclosed or intergrown with the assemblage of lorandite, getchellite, christite, and realgar are about 0.4 to 1 mm in length. Grains of ellisite in the veinlets and patches, intergrown with other arsenic minerals, range from about 0.2 to 0.5 mm in longest dimension.

Ellisite has hackly fracture and good to excellent rhombohedral cleavage. The color is dark gray, and luster on fresh surfaces is metallic and slightly less bright than that of stibnite. Streak is light brown with a tinge of orange. No natural material was found that was suitable for density measurement. The density of synthetic Tl₃AsS₃, measured with a Berman balance. is 7.10(5) g cm⁻³, which is slightly lower than 7.18 g cm⁻³ calculated from the cell volume. The Vickers hardness of ellisite determined with a Leitz hardness indenter with a 50-g load ranged from 36.4 to 44.0 and averaged 39.3 kg mm⁻² (10 determinations); the hardness of the mineral using a 25-g load ranged from 41.0 to 51.5 and averaged 45.9 kg mm⁻² (8 determinations). Both average values correspond to about 2 on the Mohs scale of hardness.

Ellisite is opaque. In reflected light it is light gray with a distinct purple tint (light purplish-gray), very weakly bireflectant with colors ranging from light purplish-gray to light pinkish-gray, and anisotropic. Polarization colors range from blue-purple to redpurple to brownish-orange. Broken grains display a deep red to deep orange-red internal reflection. Re-

Crystallography

Ellisite has trigonal symmetry, but the lattice is hexagonal. X-ray powder diffraction data for ellisite and synthetic Tl₃AsS₃, using silicon as an internal standard, are given in Table 2. The hexagonal unit cell constants, determined from X-ray powder diffraction data, using the Appleman and Evans (1973) cell parameter least-squares program, are: a = 12.324(2), c = 9.647(2)A, and $V = 1269.0(4)A^3$; $Z = 7[Tl_3AsS_3] = 21[TlAsS_3]$.

The crystal structure of ellisite has not been determined. However, it appears to be related to the crystal structure of carlinite, Tl_2S , (Z = 27) which consists of sheets of Tl atoms developed parallel to (001), in which S atoms occupy octahedral interstices between pairs of planes of Tl atoms (Ketelaar and Gorter, 1939). Removal of one of the planes of Tl atoms from each pair, and the substitution of one small As³⁺ (0.58A) for each three large Tl⁺ (1.47A) removed, as required for charge balance, would cause compaction along c from the original value for carlinite (18.175A) to the c value for ellisite (9.647A, about one half of the original distance) without much disturbance of the a parameter from the value for carlinite (12.12A).

No identifiable forms were observed on ellisite grains, but euhedral, roughly equant rhombohedrons were observed on synthetic Tl_3AsS_3 .

Chemical composition

Five grains of ellisite were analyzed by standard electron microprobe techniques. Analyses of 4 to 5 spots on each grain showed the grains to be uniform in composition. Data from these analyses and the analytical conditions used are given in Table 3. Because synthetic Tl₃AsS₃ was used as a standard, no matrix corrections were necessary. Results of the microprobe analyses show the composition of ellisite to be Tl₃AsS₃. Small hand-picked grains and fragments were combined and analyzed by the standard semiquantitative spectrographic technique of the U.S. Geological Survey (Table 4). The only additional elements present at significant levels but below the

	Ellisite			Synthetic Tl ₃ AsS ₃		
	Calculated*	* Observed** Observe		Observed**	Observed	
hkl	d _{hkl} A	d _{hkl} A	I	d _{hkl} A	I	
100	10.673		10000			
001	9.647					
101	7.157					
200	5,337	5.333	37	5.340	38	
	5 102					
002	2.193					
201	4.670			14.45.00	** ** **	
102	4.396		-			
210	4.034			and the first		
112	3.798	31175		0000		
211	3.722					
202	3.578			2 550		
300	3.558	3.559	20	3.338	20	
301	3.338					
003	3.216	3.214	53	3.218	50	
212	3.095			10000		
220	3.081			the last of the		
310	2.960				212	
221	2.035	(1000)	00 (00 Mail)			
302	2.863			<u>11</u>		
113	2.851	2.854	9	2.853	13	
311	2.830			****		
203	2.754	2.757	10	2.755	14	
400	2.668	2.669	100	2.667	100	
222	2.597					
401	2.572					
312	2.523					
	0.110					
320	2.449		323			
303	2.286	2.387	2	2.385	3	
321	2.373					
104	2.353	7777-				
402	2.335					
410	2.329	2.327	28	2.326	33	
411	2.264					
114	2.246		500			
223	2.225	0.005	-	100 A 100		
204	2.198	577		7 1 6 1		
322	2.183			2.181	- L	
500	2.1/8					
412	2.133	2.098	1		100.000	
501	2.084	2.086	2	2.090	6	
214	2.070		-		10110	
330	2.054	2012				
403	2.053		-	555	2070	
420	2.017					
331	2.009				***	
510	1.917	1.916	3	1.918	3	
115	1.841	1.839	2	1.838	3	
600	1.779	1.780	15	1.//0	17	
520 423	1.709	1.707	4	1.707	5	
600	1 ((0	1 670	4	1 666	6	
002	1.608	1.608	4	1.607	6	
530	1.525	1.524	7	1.526	10	
007 53	3 1.378]	1.377	-	1 376	c	
406	1.377	1.3/1	4	1.3/0	8	
410 801	1.323	1.323	2	1.323	3	

*All calculated hkl's listed for d_{jkl} >2.000 A. All observed hkl's for d_{jkl} >1.300 A 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 $\alpha = 12.324$ and $\alpha = 9.647$ A. **X-ray diffractometer conditions are: Ni-filtered Cu radiation; Cu Kar = 1.540598 A; silicon internal standard; scanned at $1/2^{\circ}$ 20 per minute.

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Table 2. X-ray powder diffraction data for ellisite and synthetic Tl_3AsS_3

Table 3.	Microprobe	chemical	analy	ses of ellis	site
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		Weight Percent		
Grain	T1	As	S	Total
1	78.4	9.8	12.3	100.5
2	77.9	9.5	12.2	99.6
3	78.5	9.6	12.1	100.2
4	78.2	9.6	12.3	100.1
5	77.8	9.7	12.4	99.9
Ave.	78.2	9.6	12.3	100.1
Tl ₃ AsS ₃	78.18	9.55	12.27	100.00

ADP crystal, 15 KV; (2) Arsenic, Lα characteristic line, ADP crystal, 15 KV; Sulfur, Kα characteristic line, ADP crystal, 15 KV. Synthetic Tl₃AsS₃ used as standard. Analyst: R. Wittkopp

sensitivity level of microprobe analysis were Sb (500 ppm) and Fe (200 ppm).

Synthesis

Crystalline Tl_3AsS_3 can be made by reacting mixes of Tl_2S and $TlAsS_2$ in stoichiometric proportions in evacuated sealed glass tubes at 300°C for one day. The product is a fine-grained holocrystalline mass. Coarser material is obtained by using longer reaction times; as mentioned earlier, 3 weeks produced visible rhombohedral crystals on the outer surface of the charge and inside cavities.

Phase relations

Phase relations in the system $Tl_2S-As_2S_3$ have been studied by Canneri and Fernandes (1925) by differential thermal analysis, and by Peterson (1976) by DTA and sealed glass-tube methods (Fig. 2). Ellisite melts congruently at 325°C. It exhibits solid solution toward Tl_2S (carlinite) of about 3 mole percent, but not toward $TlAsS_2$ (lorandite). The limited solid-solution behavior between ellisite and carlinite is consistent with the proposed difference in crystal structure wherein smaller As^{3+} replaces larger Tl^+ . Eutectic relations are displayed toward carlinite at 81 mole percent Tl_2S and 305.5°C and lorandite at 63 mole percent Tl_2S and 235°C. All crystalline phases found in Peterson's experiments occur at Carlin.

Assemblages of sulfosalt and sulfide minerals

Ellisite is the fifth new mineral discovered at the Carlin gold deposit by Radtke and his co-workers in samples taken from 1965 to 1975 for routine mineralogical and chemical study. The presence of the unusual minerals was not suspected until christite was first noted in 1974 (Radtke *et al.*, 1974a; 1974b), by which time many localities already had been destroyed. One locality, in the central area in the East Pit, remains, but it is in a location inaccessible at the time of this writing (fall, 1978). Consequently, detailed and systematic observations and sampling have not been possible. However, a summary statement, integrating results of mapping and collecting by Radtke with subsequent geochemical and mineralogical studies, is presented in the following paragraphs.

The mineral localities are shown on the simplified geological map (Fig. 3). Minerals unique to the Carlin deposit are: ellisite (E), Tl₃AsS₃; carlinite (Ca), Tl₂S (Radtke and Dickson, 1975); christite (Ch), TlHgAsS₃ (Radtke et al., 1977); weissbergite (W), TISbS₂ (Dickson and Radtke, 1978); and frankdicksonite (F), BaF₂ (Radtke and Brown, 1974). Rare minerals include: native arsenic (A), As (Hausen and Kerr, 1968); lorandite (Lo), TlAsS₂ (Radtke et al., 1974b); getchellite (Ge), SbAsS₃; galkhaite (Ga), approximately HgAsS but with important amounts of Tl and Sb; and avicennite (Av), Tl₂O₃ (Radtke et al., 1978). More common minerals are: orpiment (O), As₂S₃; realgar (R), AsS; stibnite (St), Sb₂S₃; and cinnabar (Ci), HgS. Most of the minerals have compositions close to their stoichiometric formulae, except for some specimens of stibnite, orpiment, and realgar (Radtke et al., 1973; Dickson et al., 1975). A preliminary statement on the sulfide and sulfosalt minerals was presented by Dickson and Radtke (1977). Basemetal sulfides of probable different timing also occur erratically in veins and in the disseminated zones.

Spatially associated sulfide and sulfosalt minerals occur disseminated in carbonate rocks and in veins. The disseminated minerals occur in large volumes of

Element	Weight percent	Element	Weight percent	Element	Weight percent
Ag	0.0001	Fe	0.020	Ti	0.0005
As	Major	Mn	0.005	Т1	Major
Co	0.0003	Ni	0.0007	v *	0.0003
Cr	0.0002	Pb	0.001	Zn	0.001
Cu	0.0015	Sb	0.05		

[Sought but not found: Al, Au, B, Ba, Be, Bi, Ca, Cd, Ce, Eu, Ga, Ge, Hf, In, K, La, Li, Mg, Mo, Na, Nb, P, Pd, Pt, Re, Sc, Si, Sn, Sr, Ta, Te, Th, U, V, W, Y, Yb, Zr. Analyst: A. S. Radtke]



Fig. 2. Reconnaissance phase diagram of the system $Tl_2S-As_2S_3$ as a function of bulk composition and temperature, at the vapor pressures of the assemblages, after Peterson (1976). Symbols: Ca = carlinite, Tl_2S ; E = ellisite, Tl_3AsS_3 ; Lo = lorandite, $TlAsS_2$; O = orpiment, As_2S_3 ; L = liquid, $Tl_2S-As_2S_3$; SS = solid solutions.

rock as dispersed grains, seams, patches, and irregular masses. The disseminated zones trend more or less parallel to the stratiform ore bodies (Fig. 3 and 4). Four disseminated mineral assemblages occur: the gold assemblage, gold + pyrite (Py) + quartz (Q) of the ore bodies; the thallium-arsenic assemblage, E + Ch + Lo + Ge + R + A + Q; the mercury assemblage, Ga + Ci + Q; and the carlinite assemblage, Ca + Q + Py + hydrocarbons (HC), scattered through brecciated carbonate rock. Two types of veins crosscut the country rocks, ore and the disseminated zones: these are near-vertical, barite (B) veins with minor quartz, and quartz veins. Vein mineral associations include: Ch + Lo + O + R + B + Q; W + St + Q; and F + Q. The barite veins occur dominantly in fractures in upper oxidized and leached rocks, but they narrow downward and terminate in the top of the underlying unoxidized rocks. The quartz veins tend to occur in the hanging wall at or immediately above the disseminated zones. The veins are spatially separate from each other. Figure 5 shows the compositional relationships of the minerals on a Tl-Hg-As-Sb diagram.

With regard to the timing, the disseminated zones clearly formed earlier than the sulfosalt- and sulfidebearing veins. However, timing of the spatially associated minerals within each disseminated zone cannot be deduced directly because the minerals rarely occur in contact. The persistent association of sets of minerals within common volumes of rock is consistent with but does not prove simultaneity. Indeed, phase-rule considerations suggest that it is improbable that all the minerals of the disseminated thallium-arsenic assemblage formed simultaneously. In



Fig. 3. Simplified geologic map of East ore zone of the Carlin gold deposit at the 6,400 foot level showing spatial relations between sulfide and sulfosalt assemblages and their relationships to prominent geologic features. Narrow zones with short bar pattern are pre-ore igneous dikes; heavy lines are faults; gray areas are gold ore bodies. Stippled areas contain dispersed thallium-arsenic minerals; lined pattern denotes areas containing galkhaite and cinnabar; cross-hatch pattern denotes areas containing weissbergite and stibnite, carlinite, or frankdicksonite. The occurrences of W + St, Ca and F in the hanging wall above ore have been projected down onto the plane of the map.



Fig. 4. Cross section along line A-A' (Fig. 3) showing vertical positions of sulfide-sulfosalt assemblages and geologic features. Symbols correspond to those of Fig. 3. Zones containing carlinite (C) and weissbergite (W) + stibnite (St) above zone of disseminated thallium-arsenic minerals are projected into plane of section. Geologic formations are thin-bedded carbonate beds of the Roberts Mountains Formation (DSrm) overlain by thick-bedded carbonate beds of the Popovich Formation (Dp).

contrast, the vein minerals are intergrown in textures typical of simultaneity, and no unlikely constraints are encountered by application of the phase rule.

The presence of distinctly different assemblages between separate unconnected veins strongly suggests that either the veins were formed at different times, or, if they were synchronous, that efficient flow of fluid was impaired in the fracture system. The retention of minerals such as carlinite, which oxidizes easily, and frankdicksonite, which is relatively soluble and reactive to components available from wall rocks (calcium, carbonate, sulfate), testifies to the efficiency of sealing of the veins from ground-water attack.

The ore-forming fluids at Carlin were derived from meteoric waters, according to D/H ratios of vein minerals and altered rocks, and they contained sulfur of marine affinity (Dickson *et al.*, 1978). The ore fluids also transported hydrocarbon compounds, now found in intimate association with ore but also dispersed in hydrothermally altered unmineralized rocks. The ore and gangue components could have been leached from deeper sedimentary rock terranes by fluids set into circulation by heat released from Tertiary volcanic rocks (Dickson *et al.*, 1978).

The temperature and pressures attending ore deposition are difficult to establish because most of the hydrothermal ore minerals are exceedingly finegrained and not suitable for fluid inclusion studies, or are intergrown with other minerals, making separations for isotopic analysis almost impossible. However, fluid inclusions in quartz in some small seams containing pyrite and gold, collected deep in primary ore, had filling temperatures ranging from 160°-180°C (A. S. Radtke, unpublished data). In contrast, fluid inclusions of later vein minerals show temperatures above 200°C [realgar, 210°C to >230°; sphalerite, 266°-299°; and quartz, 235°-285° (A. S. Radtke, unpublished data)]. The partitioning of ³⁴S between barite and sulfide vein minerals indicates depositional temperatures in the range 250°-290°. possibly as high as 305°C. We visualize that at the beginning of the episode the hydrothermal solutions were cool, and initially their passage through the host rocks did little except dissolve some calcite and deposit small amounts of quartz. The ore assemblage formed later as the temperatures increased and the hotter fluids permeated the prepared horizons. The deposition of the disseminated sulfide and sulfosalt minerals probably was during the late stages of this moderate-temperature event, judging by the tendency of the zones containing the disseminated minerals to parallel or envelop the ore bodies. The lower-temperature stage gave way abruptly to a higher-temperature stage of vein formation. The flow of the fluid switched from slow lateral permeation through large volumes of rock to rapid movement



Fig. 5. Representation of compositions of minerals in the system Tl-Hg-Sb-As-S at Carlin. Heavy lines join or encompass associated minerals. Solid heavy lines join or enclose disseminated assemblages, ellisite (E) + christite (Ch) + lorandite (Lo) + getchellite (Ge) + realgar (R) + native arsenic (A), and galkhaite (Ga) + cinnabar (Ci). Carlinite (Ca) not associated with other sulfides is shown as a single dot. Broken heavy lines join synchronous minerals of late veins, christite (Ch) + lorandite (Lo) + orpiment (O) + realgar (R), and stibnite (St) + weissbergite (W).

along faults and near-vertical fractures. The event could have been triggered by sudden breaking of the fluids through a near-surface impermeable barrier, allowing the fluids to vent at the water table or at the earth's surface. The hypothesized sudden release of pressure would promote rapid flow, fed from regions of higher temperatures at greater depths. The temperatures of the vein-forming fluids at the level of the deposit rose to a maximum before the rapid flow damped away and the deposition of vein minerals ceased.

The deposition in the veins probably was largely in response to supersaturation induced by decreased temperatures and pressures of the fluids during their vertical ascent. Some deposition was caused by boiling in the upper portions of the vein system. An interesting possibility is that emplacement of high-temperature fluids in fractures cutting mineralized rocks soaked with lower-temperature fluids set up increasing temperature and decreasing pressure gradients in the rocks; these gradients were directed toward the fractures. Some components originally in the ore body and mineralized zones may have been leached by interstitial fluids. Delivery to the vein system could have happened in stagnant fluids by diffusion up the temperature gradient. More likely, fluids migrated to the vein system by flow down the pressure gradient. The geometries of the vein systems and crushed zones are complicated. Fractures and openings were poorly interconnected in places. Specialized processes took place locally in response to boiling or to the delivery to the fractures of fluids of unusual composition. Therefore, ellisite and the ore and exotic minerals at Carlin represent the integrated effects of a hydrothermal cycle triggered by the intrusion of hot igneous material into water-containing rocks of the upper crust.

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