

Kostovite and its argentian varieties: Deposits and mineral associations

In memory of Ivan Kostov (1913-2004)
and Georgy Terziev (1935-1972)

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Abstract. Whereas the main Au- and Ag-tellurides were known as minerals long ago, many of them as early as in the 19th Century, kostovite, AuCuTe₄, was found much later, in 1966, by G. Terziev. The mineral, named after the eminent scientist Bulgarian Ivan Kostov, was firstly described from the Chelopech Au-Cu deposit, Bulgaria. More recently, it was reported by other authors from several, predominantly Au, Au-Cu, Au-telluride and complex gold-polymetallic epithermal ore deposits of different geographic and geologic positions, including Kochbulak (Eastern Uzbekistan), Commoner mine (Zimbabwe), Kamchatka (Russian Far East), Ashanti (Ghana), Buckeye Gulch (Leadville, Colorado, USA), Bisbee (Arizona, USA), Kutemajärvi (Finland), Coranda-Hondol (Romania), Glava (Sweden), Bereznjakovskoje (Southern Urals, Russia), Moctezuma (Sonora, Mexico), Panormos Bay (Tinos Island, Greece), etc. The ages of deposits vary from Archean-Proterozoic and Late Hercynian (Permian) up to Cretaceous (Senonian), Tertiary and Neogene. The mineral is commonly found in small amounts. Due to the microscopic and X-ray similarity with sylvanite, it is possibly often overlooked.

The individual deposits display paragenetic similarities, yet some notable differences. They usually include early quartz and pyrite together with main Cu-sulphides (chalcopyrite, tetrahedrite/tennantite, bornite, etc.), native gold of high fineness and often also galena and sphalerite. Kostovite, closely associates with native Te, Au- and Ag-tellurides (calaverite, sylvanite, krennerite, hessite, petzite, montbrayite), and with other tellurides and selenides (altaite, goldfieldite, clausenthalite, kawazulite). The telluride assemblages are commonly deposited in a later mineralization stage, at specific physicochemical conditions: relatively high oxidation potential (e.g., association with barite, sulphides of highly oxidized semimetals: Te⁴⁺, As⁵⁺, Sb⁵⁺, hypogene hematite/goethite); high $f\text{Te}_2/f\text{S}_2$ ratio and low $f\text{S}_2$; rather low T of formation (250-200-170°C).

In the first original analysis, kostovite is nearly Ag-free, but in many later determinations, Ag occurred in variable concentrations, outlining an apparently wide area of intermediate compositions along the kostovite-sylvanite join. For now, it seems that a compositional gap exists between these two end members, with the continuous series: Au(Cu_{1.0-0.3}Ag_{0-0.7})Te₄ (crossing the 50% point) for kostovite and argentian kostovite, and Au(Ag_{1.0-0.85}Cu_{0-0.15})Te₄ for sylvanite and cuprian sylvanite; Sb admixtures are common. Some deviations in stoichiometry from the Au/(Ag+Cu) = 1 ratio also exist in nature. It seems, however, that miscibility in this series, like in other similar lower- T hydrothermal tellurides of Au and Ag, is not realised by simple Ag-Cu substitution, but by forming of incommensurate or commensurate modulated structures with different degrees of ordering. The problem can be resolved by further investigations on natural specimens by microprobe and HRTEM analyses.

Key words: kostovite, sylvanite, telluride minerals, goldfieldite, ore genesis

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Абстракт. Докато главните Au- и Ag-телуриди са известни като минерали още от 19-ти век, костовитът, AuCuTe_4 , е установен много по-късно, през 1966 г. от Г. Терзиев. Минералът, наречен на името на известния учен българинът Иван Костов, е открит в Au-Cu находище Челопеч, България. По-късно е доказан и в редица други, предимно Au, Au-Cu, Au-телуридни и комплексни златно-полиметални епитермални рудни находища с различно географско разположение и геоложка позиция. Това са Кочбулак (Източен Узбекистан), Комънър Майн (Зимбабве), Камчатка (Руския Далечен изток), Ашанти (Гана), Бъкей Гълч (Ледвил, Колорадо, САЩ), Бисби (Аризона, САЩ), Кутемайарви (Финландия), Коранда-Хондол (Румъния), Глава (Швеция), Бережняковское (Южен Урал, Русия), Моктецума (Мексико), Панормос Бей (остров Тинос, Гърция) и пр. Възрастта на находищата варира от архайско-протерозойска и къснохерцинска до кредна (сенон), терциерна и неогенска. Минералът е намиран в малки количества. Поради микроскопското и рентгенографското му сходство със силванита вероятно често е пропускан.

Отделните находища на минерала показват парагенетични сходства, но и някои забележими различия. Обикновено в тях се съдържат ранен кварц и пирит заедно с важни Cu-сулфиди (халкопирит, тетраедрит/тенантит, борнит и др.), самородно злато с висока пробност, често галенит и сфалерит. Костовитът тясно асоциира със самороден телур, със Au- и Ag-телуриди (калаверит, силванит, кренерит, хесит, петцит, монтбраит) и с други телуриди и селениди (алтаит, голдфилдит, клаусталит, кавазулит). Телуридните парагенези обикновено са образуват в късен минерализационен стадий при специфични физикохимични условия: относително висок окислителен потенциал (напр. в асоциация с барит, сулфиди на високоокислени полуметали: Te^{4+} , As^{5+} , Sb^{5+} , хипогенен хематит/гьотит); високо отношение $f\text{Te}_2/f\text{S}_2$ и ниска $f\text{S}_2$; умерено ниска T на отлагане (250-200-170°C).

В първия оригинален анализ костовитът е почти стехиометричен, без Ag. В много от по-късните определения обаче се установява Ag в различни концентрации, очертаващи широка област от междинни състави по протежение на костовит-силванитовата линия. Изглежда, че съществува прекъсване в смесимостта между тези два крайни члена, с непрекъсната серия $\text{Au}(\text{Cu}_{1.0-0.3}\text{Ag}_{0-0.7})\text{Te}_4$, пресичаща 50%-вата граница за костовит и сребърен костовит, и $\text{Au}(\text{Ag}_{1.0-0.85}\text{Cu}_{0-0.15})\text{Te}_4$ за силванит и меден силванит. Sb е често срещан примес. Наблюдават се и отклонения от стехиометричното отношение $\text{Au}/(\text{Ag}+\text{Cu}) = 1$. Изглежда обаче, че подобно на другите нискотемпературни хидротермални телуриди на Au and Ag, смесимостта не се проявява като просто заместване Ag-Cu, а чрез формиране на несъразмерни и съразмерни модулирани структури с различна степен на подреждане. Този проблем може да се реши само чрез детайлно изследване на природни образци чрез микросондови и електронномикроскопски HRTEM анализ.

Introduction

Telluride minerals represent a substantial form of concentration of the noble metals in ore deposits. The tellurides of gold and silver are especially interesting among them, both in respect to the minerogenetic and economic aspects. Most ore telluride minerals were first discovered a long time ago, many of them already in the Nineteenth Century; in some cases even earlier. The type deposits of several of these minerals are in Transylvania, in the "Golden Quadrilateral" of Apuseni Mts., Romania (e.g., Cook et al., 2004; Ciobanu et al., 2004b).

The gold-copper telluride kostovite, AuCuTe_4 , was found much later, in the Chelopech deposit of the Bulgarian Srednogorie by Terziev*¹ (1966). The mineral was named after the eminent scientist Ivan Kostov, former President of the International Mineralogical Association - IMA. The main reason for the relatively late discovery of kostovite is the commonly small size of its grains. Recognition and characterization of the mineral only became possible with the application of the electron microprobe in mineralogical studies. Indeed, kostovite was

¹*Dr. Georgy Terziev, who gave substantial contributions to the mineralogy and geology of the Chelopech deposit, untimely died in a road accident in India.

one of the first minerals defined and characterized by using of microprobe. Additional data for kostovite from the Chelopech type locality were obtained by Kovalenker et al. (1986) and Petrunov (1994).

Today, the mineral is known from dozen ore deposits all over the world and from various ages and geological settings. Detailed studies identified the mineral in some well known abandoned mines and its real occurrence is probably wider still. Despite this, published data for this mineral are scarce and incomplete. Since it is nearly always found in small amounts, and due to its microscopic and X-ray similarity to sylvanite, microprobe analysis is required to confirm identification. It is possibly commonly overlooked.

In this contribution, we will try to bring together available published data for kostovite, together with our own observations on the distribution, composition, mineral relationships and conditions of formation of the mineral in various ore deposits.

Deposits of kostovite

In addition to the type deposit (Chelopech, Bulgaria), kostovite is reported in Kochbulak, Eastern Uzbekistan (Kovalenker et al., 1979), Commoner mine, Zimbabwe (Twemlow, 1984), Kamchatka Peninsula, Russian Far East (Sakharova et al., 1987), Ashanti, Ghana (Bowell et al., 1990), Campbell mine, Bisbee, Arizona, USA (Anthony et al., 1990; Graeme, 1993), Buckeye Gulch, Leadville, Colorado, USA (Anthony et al., 1990), Kutemajärvi, Orivesi, Finland (Anthony et al., 1990), Coranda-Hondol, Romania (Udubaşa et al., 1992), Bereznjakovskoje, Southern Urals, Russia (Lehmann et al., 1999), Moctezuma, Sonora, Mexico (Braith et al., 2001), Glava, Sweden (Cook, Ciobanu, 2000, unpublished), Panormos Bay, Tinos Island, Greece (Tombros et al., 2004), etc.

The ages of deposits vary from Archean, up to Neogene. The various deposits display

paragenetic similarities, yet also some notable differences.

Chelopech

The major Chelopech gold-copper deposit of Upper Cretaceous (Senonian) age is located in the northern part of the Panagyurishte region of the Srednogie metallogenic zone, in the central sector of the extensive Tethyan-Eurasian copper belt. The basement in the area consists of gneisses and amphibolites (Precambrian?), and Palaeozoic phyllites and diabases overlain by Turonian conglomerates, sandstones and shales. The ore deposit is located in the central part of the Chelopech volcanic structure of early subvolcanic dacitic-andesitic rocks, later andesitic lavas and tuffs, and latest dykes and subvolcanic bodies of andesites, dacites and porphyrites. The orebodies include massive steeply-dipping lenses and pipes, stockwork zones of sulphide veinlets and disseminated ore in altered wall rocks. Characteristics of the deposit and ore mineralization are given by many authors (Terziev, 1964, 1968; Petrunov, 1994, 1995; Andrew, 1997; Popov et al., 2000; Moritz et al., 2001; Bonev et al., 2002; and others).

Together with the main elements, Cu, Au, Fe, S, As and Ba, the geochemical and mineralogical specificity and complexity of the deposit is defined by a number of other elements - Sb, Bi, Te, Se, Ag, Pb, Zn, Sn, In, Ga, Ge, Tl. Pyrite is the dominant mineral. Also very important are chalcopyrite, tennantite, bornite, enargite, luzonite, sphalerite and galena, with a large number of subordinate and minor minerals: tetrahedrite, goldfieldite, famatinite, digenite, covellite, etc. The ore-forming process is complicated due to overlapping and interpenetration of the mineral associations of three successive main ore stages (Petrunov, 1994, 1995): iron-sulphide, copper-arsenic-sulphide, and polymetallic-sulphide. During the first stage massive pyrite lenses, veinlets, nests and impregnations are formed accompanied by chalcedonic silica and, in depth, by anhydrite.

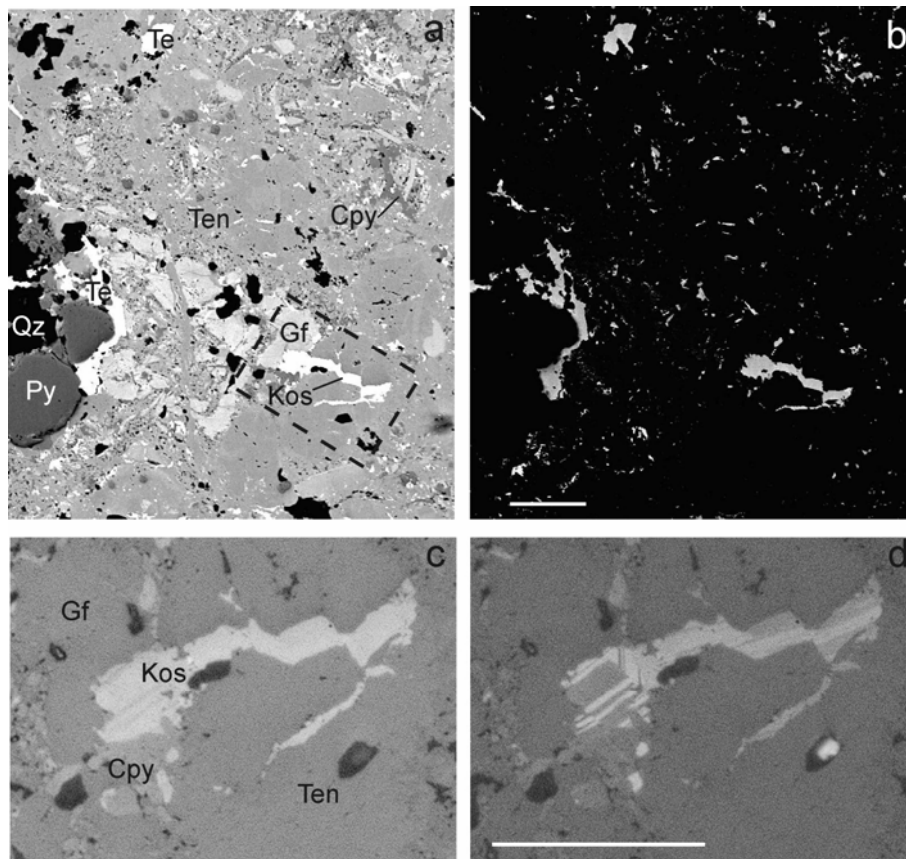


Fig. 1. Relationships of kostovite (Kos) with goldfieldite (Gf), tennantite (Tn), native tellurium (Te), chalcopyrite (Cpy), pyrite (Py) and quartz (Qz): a) and b) in back-scattered electron image (BSEI) at low (a) and high contrast (b), in the last case only the heaviest Te minerals (kostovite and native Te) being exposed; c) and d) enlarged optical views of twinned kostovite grains in reflected microscopy (inset from a), revealing the pleochroism ($// N$, in c), and the characteristic anisotropy ($\times N$, in d). Scale bar in all pictures is 30 μm , the Chelopech deposit

Фиг. 1. Отношения на костовита (Kos) с голдфилдит (Gf), тенантит (Tn), самороден телур (Te), халкопирит (Cpy), пирит (Py) и кварц (Qz): a) и b) изображения в обратно-отразени електрони (BSEI) при нисък (a) и висок контраст (b), при което в последния случай видими са само тежките Те-минерали - костовит и самороден Те; c) и d) увеличени микроскопски изображения в отразена светлина на вдвойникувани зърна от костовит (детайл от a), показващи рефлексивния плеохроизъм ($// N$, в c) и характерната анизотропия ($\times N$, в d). Скала - 30 μm , находище Чelopeч

The most important and complex main copper-arsenic-sulphide stage reflects a high-sulphidation (acid-sulphate) epithermal system developed in subaerial conditions and controlled by intensive fracturing and brecciation. It forms advanced argillic and silicic type of

alteration with alunite and dickite, diaspore, andalusite, quartz, anhydrite and barite, overprinting earlier propylitization and argillization. The chalcopyrite-tennantite, enargite-luzonite, quartz-pyrite, bornite-digenite parageneses form the massive orebodies,

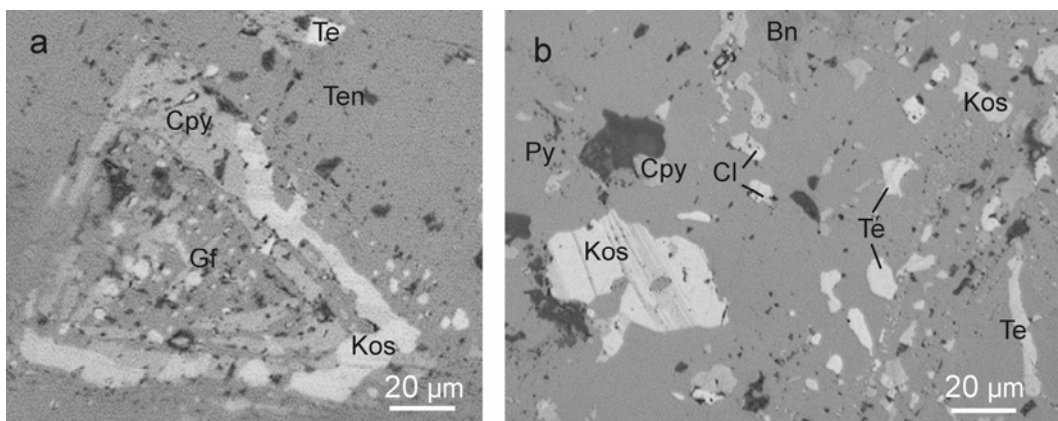


Fig. 2. a) Kostovite (Kos) and chalcopyrite (Cpy) selectively replacing the peripheral part of a tetrahedral crystal of goldfieldite (Gf) in tennantite mass (Ten); b) fine grains of pleochroic kostovite (Kos), disseminated together with native tellurium (Te), clausthalite (Cl), bornite (Bn) and pyrite (Py) in tennantite mass. Reflected light, // N, Chelopech

Фиг. 2. а) Костовит (Kos) и халкопирит (Cpy), селективно заместващи периферни части на тетраедричен голдфилдитов кристал (Gf) в масата на тенантит (Ten); б) финозърнест плеохроитен костовит (Kos), разсеян заедно със самороден телур (Te), клаусталит (Cl), борнит (Bn) и пирит (Py) сред тентитовата маса. Отражена светлина, // N, Чelopeч

stockworks and veinlets with different spatial distribution.

Highly oxidised sulphides and tellurides occur in the upper levels of deposit, including goldfieldite (Kovalenker et al., 1986), and different tellurides, followed at depth by enargite-luzonite, tennantite with chalcopyrite, and bornite-digenite-annilite. Late-stage sphalerite-galena ores associated with barite are prevailing especially in the higher and outer zones. Various rare copper sulphides of Sn, Ge, V, Mo, were also found in the deposit, including hemusite ($\text{Cu}_6\text{SnMoS}_8$), germanocolusite and stibicolusite (type deposit), stannoidite, colusite, mawsonite, arsensulvanite, nekrasovite and renierite. A number of Bi minerals also occur, including native Bi, bismuthinite, aikinite and wittichenite.

Goldfieldite is the earliest and main tellurium-bearing mineral. Its tetrahedral {111} crystals included in pyrite-sulphide mass have patchy and zonal structure with more arsenian cores (5-12 wt.%) and highly tellurian rims with only 2-4 wt.% As. Crystals are cracked and intensively replaced by tennantite and

minor chalcopyrite. *Kostovite*, often in association with native tellurium, is found in the neighbourhood of goldfieldite crystals, usually replacing their peripheral zones. Typical complex fine-grained tennantite-goldfieldite-chalcopyrite-pyrite ore from Chelopech is shown in Fig. 1, with two back-scattered electron micrographs displayed at different contrasts (a and b), and with two enlarged micrographs in reflected light (c and d). Mineral relationships are presented in Fig. 2 a and b. Apart from native Te, kostovite is associated with other telluride minerals such as sylvanite, petzite, montbrayite, altaite, tellurobismuthite, and also with clausthalite, kawazulite, součekite, and mawsonite.

Microprobe analyses of kostovite have established that in Chelopech, together with the nearly stoichiometric AuCuTe_4 (as found in the first analysis of the type material by Terziev, 1966), argentine varieties of the mineral also occur, with up to ~3 wt.% Ag (Table 1). In most cases the mineral also contains Sb, up to 1.58 wt.% (0.10 to 0.20 *apfu*). Although As is the prevailing metalloid component of the

Table 1. Representative microprobe analyses of kostovite, wt.%
 Таблица 1. Представительни микросондови анализи на костовит, тегл. %

Deposit	Theoretical		Chelopech														Bisbee	Tinos	Ashanti	
	AuCuTe ₄	Au(Cu _{0.5} Ag _{0.5})Te ₄	Terziev 1966	Kovalenker, 1986	New data															Criddle, Stanley, 1993
			Type	Ch1	Ch2	PK96	PK2	PK4	PK10	PK3	K94	PK6	PK3a	P4	K99	296	Mean	Mean		
Au	25.55	24.84	25.2	25.93	25.54	24.75	23.15	22.64	24.76	22.97	24.29	24.05	23.83	26.31	23.15	27.2	27.20	24.40		
Ag	-	6.80	0.4	1.04	1.35	1.02	1.03	1.09	1.17	1.26	1.54	1.60	1.62	1.87	2.97	0.5	0.50	0.74		
Cu	8.24	4.00	7.7	7.07	7.00	9.09	8.84	8.50	7.60	8.56	8.13	8.77	9.07	7.80	7.74	7.3	7.85	9.20		
Te	66.21	64.36	67.6	64.82	65.70	66.68	66.11	66.03	65.70	65.60	67.05	65.43	64.90	63.66	66.72	64.4	64.24	66.05		
Sb	-	-	-	-	-	0.49	1.16	1.58	1.15	0.91	0.54	0.65	0.70	-	0.43	-	-	-		
Σ	100.00	100.00	100.9	99.18	99.68	102.03	100.29	99.84	100.38	99.30	101.55	100.50	100.11	99.64	101.01	99.6	99.99	100.39		
<i>apfu</i>																				
Au	1	1	0.98	1.03	1.01	0.94	0.78	0.88	0.97	0.89	0.93	0.93	0.92	1.04	0.89	1.08	1.07	0.94		
Ag	-	0.5	0.03	0.08	0.10	0.07	0.09	0.08	0.08	0.09	0.11	0.11	0.11	0.13	0.21	0.04	0.04	0.05		
Cu	1	0.5	0.93	0.87	0.86	1.06	1.12	1.02	0.92	1.03	0.97	1.04	1.08	0.95	0.92	0.90	0.96	1.09		
Te	4	4	4.06	3.99	4.02	3.90	3.94	3.92	3.96	3.93	3.96	3.88	3.85	3.88	3.95	3.95	3.89	3.92		
Sb	-	-	-	0.03	0.01	0.03	0.07	0.10	0.07	0.06	0.03	0.04	0.04	-	0.03	-	-	-		
Sum	6	6	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00		

Additionally reported (in wt.% / at.%, respectively): Ch1: Se 0.32 / 0.03 ; Ch2: Se 0.09 / 0.01, S 0.1 / 0.02; Bisbee: Fe 0.10 / 0.01, S 0.1 / 0.02; Tinos: S 0.20 / 0.04

orebodies (compared to Sb in, for example, tennantite-goldfieldite, enargite, etc.), As is never included in kostovite. In some analyses Se is also found (Table 1). Individual kostovite grains do not show any chemical heterogeneity. Formation of kostovite and associating telluride minerals is related to overprinting of later Au-bearing fluids of lower fS_2 .

Kochbulak, Uzbekistan

The major Kochbulak epithermal gold-telluride deposit is located on the north slope of the Kuramin Ridge, Middle Tien Shan Mountains, Eastern Uzbekistan (Kovalenker et al., 1997). The deposit is hosted in a local caldera of andesite-dacitic volcanic rocks (C_{2-3} age), cross-cut by subvolcanic dacite-, diorite- and granosyenite porphyry stocks and dykes of granodiorite-porphyry and diabase (C_3-P_1). The hydrothermal system is related to processes of uplifting with intensive volcanic activity and introducing of porphyric granitoid bodies. Three main morphogenetic types orebodies are developed: steep veins, low-angle sloping NE-striking quartz ore veins, 1-2 m thick (up to 5-20% sulphides), and explosive-hydrothermal breccia pipes with high-grade quartz-gold-sulphide-telluride bonanza ores, with lens-like or irregular horizontal sections and from several up to 100-120 m long axis (up to 10-30% sulphides). The ore mineralization extends from near surface down to the roof of a hypoabyssal porphyry stock. The K-Ar and Rb-Sr dating of ore mineralization determine about 280 ± 8 Ma for the breccia pipe ore, and 270 ± 8 Ma for the vein ores. Many authors have studied the very complex geology, ore mineralogy and conditions of formation of the deposit. Important contributions and summaries are given by Kovalenker et al. (1997). More than 110 ore minerals and unnamed phases have been identified and for several of these, this is the type deposit. The intensive wall-rock alterations mostly belong to the quartz-sericite \pm adularia type along the steep and sloping veins, and to the quartz-diaspore \pm alunite type along with the steep pipe ore bodies.

During the complex and prolonged mineralization process, three main productive ore stages are distinguished, with several parageneses in them. Fine-grained and recrystallized quartz, together with pyrite, is prevailing mineral, accompanied by chalcopyrite, sphalerite, galena, and variable members of the tetrahedrite-tennantite series. Interesting and important is goldfieldite, the tellurian member of the series, commonly associated with famatinite-luzonite and telluride minerals. Gold is disseminated in nearly all parageneses, as in quartz, as well as within pyrite and other minerals. It is especially enriched in bonanza ores within quartz-pyrite aggregates, together with variety of Au-Cu-Ag-Bi tellurides, and Cu-Pb-Bi-Ag sulphosalt minerals. In such ore nests, which can reach several cm in size, coarse-grained native tellurium, calaverite, sylvanite, *kostovite*, petzite, hessite, altaite and tellurantimony have been found. These finds enabled detailed investigation of the physical, optical, crystallographic and crystallochemical characteristics of kostovite (Kovalenker et al., 1979). Kostovite from Kochbulak is argentiferous (Table 2, Fig. 4).

Systematic fluid inclusion studies (Kovalenker et al., 1997) established that the main ore-forming stages in Kochbulak started at high temperatures ($>400^\circ\text{C}$) and continued at decreasing T by low-concentrated (0.5 M Cl), periodically boiling Cl-Na-K solutions of moderate acidity (pH 3-4). The productive Au and Ag telluride and sulphosalt mineral associations are attributed to the 250-200°C temperature range, after repeated involvement of concentrated (up to 5M Cl) F-Cl-Te-bearing solutions at decreasing fS_2 .

Kamchatka, Russian Far East

Sakharova et al. (1987) described gold-tellurian vein mineralization in the volcanic belt of Central Kamchatka. Orebodies are included within a large volcanic structure of Neogene age. The hydrothermal veins and mineralized zones are of quartz and quartz-adularia-carbonate compositions. They contain scarce sulphides, mainly pyrite, chalcopyrite, galena

Table 2. Representative microprobe analyses of argentinean kostovites, wt. %
Таблица 2. Представителни микросондови анализи на сребро-съдържащи костовити, тегл. %

Sample	Kochbulak										Kamchatka						Glava	
	KB5/3	KB5/1	K21	K22	K23	K24	K17	P198	S1	S2	S3	S4	S5	S6	A19.3	A19.1	A19.4	
Au	26.68	27.37	27.82	27.93	26.40	25.71	25.04	26.11	24.9	25.8	26.2	27.2	27.0	25.9	24.82	22.69	21.15	
Ag	3.60	4.03	3.64	3.19	4.36	3.25	9.18	3.19	4.5	3.7	4.9	4.7	5.0	4.7	5.85	6.00	7.55	
Cu	4.79	4.85	4.34	4.56	4.94	5.57	2.30	6.62	4.3	5.1	4.3	4.2	3.7	4.1	4.85	7.50	4.30	
Te	64.84	63.97	64.41	64.31	64.14	64.27	63.88	64.17	64.2	64.1	63.9	63.0	64.1	63.8	61.87	64.16	61.05	
Sb	0.34		0.28	0.32	0.33	0.31	0.54	0.18	0.4	0.3					0.53	0.47	0.47	
Σ	100.25	100.22	100.49	100.31	100.17	99.11	100.94	100.55	98.8	100.1	99.3	99.1	99.8	98.5	98.20	100.35	95.15	
<i>apfu</i>																		
Au	1.08	1.10	1.13	1.13	1.06	1.04	1.01	1.03	1.02	1.04	1.07	1.12	1.11	1.07	1.02	0.87	0.88	
Ag	0.27	0.30	0.27	0.24	0.32	0.24	0.68	0.23	0.34	0.27	0.37	0.35	0.37	0.35	0.43	0.42	0.58	
Cu	0.60	0.61	0.55	0.58	0.62	0.70	0.29	0.81	0.55	0.63	0.54	0.53	0.47	0.52	0.58	0.90	0.56	
Te	4.03	3.99	4.03	4.03	3.98	4.00	3.99	3.90	4.06	4.02	4.02	4.00	4.05	4.06	3.92	3.81	3.93	
Sb	0.02		0.02	0.02	0.02	0.02	0.03	0.01	0.03	0.02					0.03			
Sum	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	6.00	

Additionally reported (in wt. % / at. %, respectively): P198: Se 0.21/0.02; S1: Bi 0.3/0.01, Fe 0.20/0.02; S2: Bi 0.9/0.03; Fe 0.20/0.02; A19.3: Hg 0.51/0.02, Se 0.04/0; A19.4: Hg 0.5/0.02, Se 0.09/0.01, S 0.04/0.01

and sphalerite, together with gold of rather high fineness (850-960). The tellurian mineralization consists of goldfieldite, calaverite, petzite, hessite, altaite, *kostovite* and rucklidgeite. Kostovite occurs as rounded or skeletal grains, 5-20 μm in size, together with Cu-bearing sylvanite, and is closely associated with the goldfieldite-chalcocopyrite aggregates in ore nests of the veins. Kostovite systematically shows increased Ag content, in the range 3.7-5.0 wt.% (Table 1, Fig. 4). Sylvanite has up to 1.5 wt.% Cu (Table 3). The volcanogenic gold-telluride deposits of Kamchatka were formed at shallow depth.

Ashanti, Ghana

The Ashanti mine in Ghana is one of the major gold deposits in Africa, and has been intensively worked during the past century. The gold mineralization in quartz veins (Bowell et al., 1990) is related to large shear zones in the late Archean and early Proterozoic rocks of the West African craton of the Birimian series (>2 Ga). The Lower Birimian tightly-folded metasediments comprise phyllites, greywackes and tuffs, which are followed by the more competent metamorphosed and metasomatically altered Upper Birimian volcanics, and then by the Tarkwanian metasediments. The main ore-controlling NNE-trending shear zone follows the ill-defined contact between the two Birimian series. The three significant known mineralized steeply dipping shears, the Cote d'Or, Obuasi, and Ashanti, converge at depth to form the 'Main Reef Fissure'.

Bowell et al. (1990) describe the auriferous ores occurring both as massive and ribboned quartz veins, 1-3 meters and thicker, occupying dilatational zones in the major ductile-brittle shear zones, and as disseminated sulphides and sulpharsenides. The productive ore zones are followed at a length of 8 km, and at depth of 1650 m; gold is confined to well-defined ore-shoots.

Four mineralization stages have been recognized: I) massive smoky blue-grey quartz, the dominant component in the veins, and in

small vein-offshoots, which host most of the gold, together with arsenopyrite, sphalerite, galena, chalcocopyrite, pyrite, tetrahedrite, bornite and pyrrhotite; II) ribboned white quartz, which has partly replaced early quartz and in places contains high concentrations of gold, and includes telluride minerals; the later III) barren quartz-calcite, and IV) barren quartz stages in veins and veinlets transecting the earlier ore mineralizations. The wallrock alterations include sericitization, sulphidation, carbonatization and silicification.

Telluride mineralization occurs rarely as local podiform aggregates up to 15-20 cm in size, within white quartz of II stage in complex assemblages with hypogene hematite, goethite, chalcocite and calcite. 11 telluride minerals have been identified by Bowell et al. (1990), and in which altaite, coloradoite, petzite and hessite occur as macroscopic (i.e., mm-sized) grains. The pods with gold-telluride mineralization have a distinct zoning:

- Core mineralization (stage IIa) consisting mainly of coloradoite + chalcocite + goethite. Fine symplectitic intergrowths of calaverite, sylvanite, *kostovite* (with 0.71-0.79 wt.% Ag - Table 1), and petzite intergrown with goethite and hematite occur in this zone. Some symplectites partially replaced gold. Chalcocite and coloradoite grains comprise complex intergrowths of rickardite, weissite, henryite, stützite, hessite and petzite.

- Peripheral mineralization composed of dominating altaite, with minor petzite, hessite and gold.

- Some later cross-cutting quartz-calcite veinlets (stage IIb) include altaite, petzite and gold.

Fluid inclusion geothermometry gave the following temperature T_h ranges for the successive stages: I) 340-267°C for early gold-bearing quartz from two phase L-G inclusions, IIa) 220-165°C for quartz accompanying the telluride mineralization, from three-phase H₂O-CO₂ inclusions with low salinity of ~5 wt.% NaCl equiv., and IIb) 175-145°C for cutting veinlets from similar inclusions, III) and IV) 220-120°C for the late barren quartz.

When the main quartz stage in Ashanti mine includes sulphides, sulpharsenides, sulphosalts and early gold (low-sulphidation style of mineralization), the later overprinted in the reactivated shear zone lower- T quartz-telluride mineralization is characterized by considerable change of environment – increased fO_2 (goethite and hematite), marked increase of fTe_2/fS_2 ratio, and very low fS_2 , with instability and extensive replacement of the early Cu and Fe sulphides. Bowell et al. (1990) supposed that, in such conditions, the hydrothermal transport of gold may be realized by hydrotelluride, rather than as hydrosulphide complexes at low fluid pressure. It is supposed that Te-rich fluids are connected with the emplacement of a synorogenic granitoid intrusion and so the deposit is of plutonic-hydrothermal origin.

Commoner mine, Zimbabwe

The similar but much smaller Commoner mine is situated in the Midlands greenstone belt of Zimbabwe (Twemlow, 1984). The intensively folded host volcano-sedimentary rocks include tuffs and carbonaceous siltstones, interbedded with minor andesitic lavas, carbonaceous phyllites and cherts, belonging to the Archean Bulawayan Group (2.7-2.9 Ga). Several hydrothermal quartz-carbonate veins are known in shared zones. The location of the main ore shoots (Bulawayo, Salisbury, Gokwe, and C18) is strongly controlled by the fold structures. Two distinct stages of gangue mineralization have been recognized.

The early mineralization predominantly of massive quartz occurs in lenses and encloses fragments of wall rocks. It contains the main economic gold concentration, together with an early-disseminated sulphide assemblage of pyrite, chalcopyrite, sphalerite, galena, and tetrahedrite, together with scheelite and with early hessite. The second mineralization occurs as laminated quartz-carbonate zones along partings within the main veins or in the hanging wall contacts. Gold is also related to a later fracture-controlled telluride mineralization, which partially replaced the early gold-

bearing sulphides, and on places deposited with hematite and chalcocite in open cavities and zones of later brecciation. The main tellurides are altaite, coloradoite, petzite and hessite, with minor sylvanite, rickardite, weissite, stützite, *kostovite*, and native tellurium. Symplectitic intergrowths of gold tellurides and hematite envelop and replace the early gold grains. The association of hessite + sylvanite is considered as indicative for maximum T of deposition of the telluride mineralization of 170°C.

Campbell mine, Bisbee, Arizona

Campbell is one of the several limestone-hosted hydrothermal breccia and replacement orebodies related to the Sacramento stock of the porphyry copper deposits at Bisbee, SE Arizona, USA (Criddle et al., 1989; Graeme, 1993). It was mined for a long period producing firstly base metals, Cu, Pb and Zn, and later Au and Ag. The cigar-shaped orebody is bedded within a sequence of Paleozoic limestones (of Cambrian, Devonian, Mississippian and Pennsylvanian-Permian age), controlled by the large Campbell fault zone of a series of closely spaced fractures and partly occupied by a porphyry dyke. The deposit consists of a core of brecciated pyrite, silica and silicified limestones clasts, surrounded by a zone of base metal sulphides, formed by multiple influxes of complex mineralizing fluids.

In the lower levels brecciated pyrite and silica-carbonate matrix contains early W-Sn-Fe-V oxides, and sulphides of the same metals (stannite, stannoidite, k esterite, mawsonite, colusite, kiddcreekite, etc.) in association with Cu-Fe sulphides (Criddle et al., 1989). An extensive suit of telluride minerals is characteristic. Dominating are the tellurides of Au and Ag, often associated with native gold and tellurium, but also with tellurides of Pb, Cu, Bi and Ni. These minerals form very complex intergrowths between and with base metal sulphides, and occur as filling fractures and inclusions in pyrite. The tellurides represent a later and lower temperature stage of mineralization where Te dominated S. In the upper levels of the orebody hematite-silica

breccias and replacements are developed together with another extensive assemblage of Bi-Cu-Ag sulphides and sulphosalts, and with native elements, Au, Ag, Cu and Bi. In this late low-temperature stage Te is absent, its place taken by Bi. Near to the surfaces low-temperature Cu and Fe sulphides and oxides, sulphates and other oxidation products are superimposed.

Kostovite occurs as small greyish grains associated with pyrite, chalcopyrite, goldfieldite, altaite and melonite (Criddle et al., 1989; Criddle, Stanley, 1993; Graeme, 1993). The mineral composition is nearly stoichiometric (Table 1, Fig. 4). Other tellurides in the deposit are sylvanite, calaverite, volynskite, krennerite, rickardite, rucklidgeite and stützite; detailed information is, however, absent.

Bereznjakovskoje, Southern Urals, Russia

The Bereznjakovskoje gold trend is one of the numerous ore deposits in the southern Urals. It belongs to the East Uralian zone south of Cheljabinsk, which consists of a Silurian to Early Carboniferous volcano-sedimentary sequence on Proterozoic basement, intruded by post-orogenic Permian granites. According to Lehmann et al. (1999), the deposit is hosted by dacitic to andesitic volcanic-subvolcanic units of Late Devonian age and consists of a system of E-W-striking irregular lens-like bodies several m in width, and up to 200 m long with a dip of 40-70°. Epithermal gold mineralization is of the sulphide-rich low-sulphidation (adularia-sericite) type with metal spectrum of Au-Ag-As-Sb-Cu-Zn-Pb, and a characteristic Te component. In general, it consists of an early barren pyrite stage of impregnations and brecciated vein filling, as a part of intensive propylitic alteration, followed by the main base and precious metal-rich sulphide-telluride stage. The dominated sulphide is high Cu-tennantite with minor tetrahedrite and low-iron sphalerite. They are related to quartz-sericite alteration with some adularia and carbonates. The tellurides are closely associated with fahlore, overprinting and replacing it or

occurring as later phases in cracks and in vugs. The most abundant telluride is altaite, followed by petzite. They are finely intergrown with rare clausthalite, galena, tellurobismuthite, and locally with calaverite, hessite, sylvanite, weissite, coloradoite, and *kostovite*, also with gold. The telluride-gold mineralization is considered formed from a short impulse of high-Te and low-S fluids of magmatic origin developed mainly in the central parts of the hydrothermal system. Enrichment in native gold is related to secondary low-temperature, redistribution process.

Moctezuma, Mexico

The Moctezuma district in Sonora, Mexico (Deen, Atkinson, 1988; Braith et al., 2001) is constituted by vitreous rhyolite ignimbrites and dacite lavas interlayered with thin limestone and siliceous beds of Upper Cretaceous-Tertiary age. The ore mineralizations are controlled by a group of several NNW faults and include a skarn Pb-Zn deposit in limestones (Oposura), and various silver- and gold-bearing quartz veins. Of particular interest are three gold-tellurian epithermal vein deposits, formed at shallowest depths: Bambolla mine (or Moctezuma mine), Bambollita mine and San Miguel Prospect. These famous ore veins, located in a hardly accessible wild region of Northern Mexico, have been explored firstly for gold, later for tellurium, but were abandoned several decades ago. Their unique mineralogy has been studied by many authors, and includes more than 150 primary and secondary minerals, 23 of which are new mineral species (!), first described from the district. Among these are the tellurium-bearing minerals bambollaite $\text{Cu}(\text{Se},\text{Te})$, burkhardtite $\text{Pb}_2(\text{Fe}^{3+},\text{Mn}^{3+})\text{Te}^{4+}[\text{O}_2(\text{OH})_2\text{AlSi}_3\text{O}_{10}]\cdot\text{H}_2\text{O}$, benleonardite $\text{Ag}_8(\text{Sb},\text{As})\text{Te}_2\text{S}_3$, and cervelleite Ag_4TeS .

Native tellurium is the main ore mineral in the quartz veins of Bambolla mine, and is often found as massive granular aggregates and large crystals, accompanied by fine-grained gold-bearing pyrite, tetrahedrite-goldfieldite, and a great variety of tellurides. Barite, dickite and kaolinite are, together with quartz, the

main gangue minerals. The telluride association includes tellurides of gold (krennerite, *kostovite* and montbrayite), of Cu and Ag (hessite, benleonardite, bambollaite, rickardite, henryite and cervelleite), Hg (coloradoite), Pb (altaite), Bi (tetradyomite). Some selenides also occur (klockmannite, bohdanowiczite, tiemannite). Tellurides formed fine-grained inclusions within tellurium, in some places also finely intergrown masses. Bambollita and San Miguel are Ag-rich deposits, with galena, bornite, hessite and tetrahedrite-goldfieldite, also with Bi minerals. The oxidation zone of the deposits is known with remarkable mineral diversity.

Glava, Sweden

The abandoned Cu-Ag deposit Glava in Värmland, South-central Sweden, is small in scale, but metal-rich and mineralogically rather complex (Scherbina, 1941; Oen, Kieft, 1984). Together with others in the Vänern-Mjøsa belt (Alm, Sundblad, 1994), Glava belongs to the Southwest Scandinavian domain. The deposit is hosted within a suite of 1.6-1.1 Ga granitoids, gneisses and metasediments. The ores are generally inferred to be Sveconorwegian (~0.9 Ga). Glava is one of several bornite-chalcopyrite-dominated vein systems, characterized by conspicuously low Zn and Pb, and high Cu/S ratios (Alm, Sundblad, 1994). The new investigations on samples from surface outcrops reveal a diverse abundance of tellurides and selenides as >10-100 µm clustered inclusions within bornite, and as lenticular telluride 'swarms' in gangue. Telluride inclusions in bornite are remobilized within zones of sealing and adjustment synchronous to pulses of micro-shear in the host.

The mineral assemblage contains chalcocite, digenite, wittichenite, tellurobismuthite, cobaltite, hessite, volynskite, altaite, native tellurium, petzite, sylvanite, gold, electrum, stützite, greenockite, cobaltite, clausthalite and

cobaltian melonite. The presence of native silver, calaverite, empressite, naumannite and weissite, was also confirmed by the new microanalyses. Several other species not reported previously from Glava were also identified: rucklidgeite, tetradyomite-kawazulite solid solution, mckinstryite, and also *kostovite*. The dominant telluride in the assemblages is tellurobismuthite, present as early segregation product of high-temperature bornite solid solution (Fig. 3).

Selenides (naumannite, clausthalite) are relatively rare throughout the ore, and Te-Se-bearing phases (e.g., kawazulite), are very much subordinate. Sulphides accompanying the above assemblages are few: with the exception of bornite and chalcocite, only a single grain of mckinstryite has been identified. Galena, argentite and bismuthinite are conspicuous by their absence.

At Glava, *kostovite* is closely associated with native gold and calaverite and occurs as minor (<10 µm) grains. Microprobe analyses of *kostovite* found that varying, but significant contents of Ag are incorporated in the mineral (Table 2, Fig. 4), suggesting extensive solid solution between *kostovite* and sylvanite (or krennerite). The following assemblages of tellurian minerals are important:

- Tellurobismuthite, the dominating telluride, presented as early segregation product of bornite solid solution (initial Te and Bi);
- Later segregations: hessite, volynskite, tsumoite, native tellurium (input of Ag + Pb to the initial Te + Bi);
- Consequent mineral phases: gold, petzite, kawazulite, sylvanite and *kostovite* as reaction products, chalcocite replaced bornite, hematite also occurs (input of Au + Cu to earlier Te + Bi + Ag);
- The latest assemblage consists of Co-melonite (+ Ni, Co). Hematite indicates a high fO_2 .

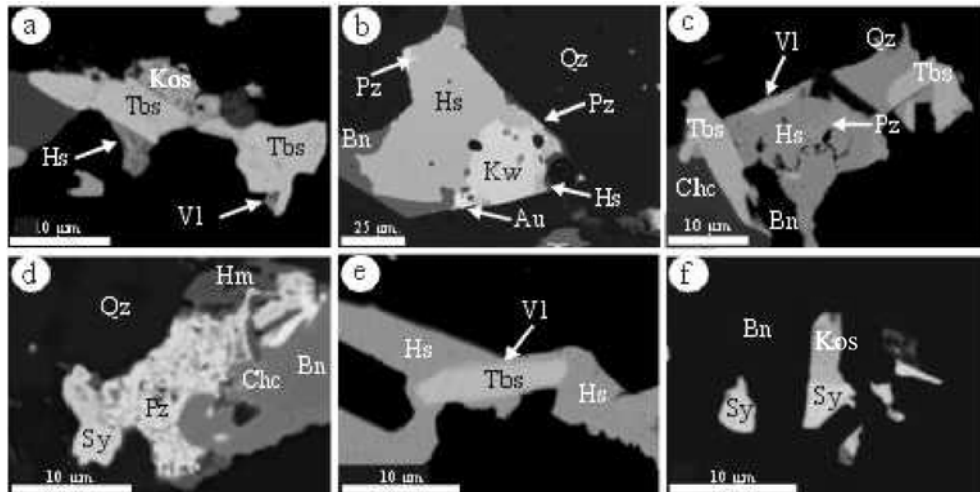


Fig. 3. BSE images of complex exsolutions of gold- and silver- bismuth-tellurian minerals within Glava bornite (Bn) (a-e). Different intergrowths of tellurobismuthite (Tbs), hessite (Hs), kostovite (Kos) and sylvanite (Sy), together with minor gold (Au), petzite (Pz), kawazulite (Kw) and volynskite (Vl). Qz - quartz, Chc - chalcocite, Hm - hematite

Фиг. 3. BSE изображения на сложни отсмесвания от златни и сребърни бисмутово-телуридни минерали сред борнита (Bn) от Глава: (a-e) различни прораствания на телуробисмутит (Tbs), хесит (Hs), костовит (Kos) и силванит (Sy), заедно с малко злато (Au), петцит (Pz), кавазулит (Kw) и волинскит (Vl). Qz - кварц, Chc - халкоцит, Hm - хематит

Panormos Bay, Tinos Island, Greece

The size and economic importance of the polymetallic Au-Ag-Te occurrence on the Greek Tinos Island is still not well known but its mineralogy was studied in detail by Tombros et al. (2004). In the area, thick-bedded dolomitic Triassic marbles are exposed. These are overlain by schists and marbles of the Blueschist Formation of the Cyclades Islands, which was successively metamorphosed to the blueschist facies in the Lower Eocene, and then to the greenschist facies in the Oligocene-Miocene. The polymetallic mineralization is related to quartz vein-stockwork systems of nearly 30 steep veins, controlled by two conjugated strike-slip faults. The older veins, up to 2 m wide, trend NE to NNE and consist of milky quartz with sulphides (pyrite, tetrahedrite, chalcopyrite, and bornite). The younger veins, of width >0.5 m, trend NW to WNW, formed by open space

filling and consist of clear quartz with some galena. The Tinos granodiorite-leucogranite pluton of Miocene age is located some 16 km to the east.

As shown by Tombros et al. (2004), the polymetallic mineralization has a complex mineralogy and multistage formation in 8 hypogene paragenetic stages. The first five stages are associated with milky quartz, calcite, talc, brucite, and muscovite, and are presented by common sulphides and sulphosalts, including: I - pyrite, minor arsenopyrite and pyrrothite; II - tetrahedrite, tennantite, goldfieldite; III - chalcopyrite and bornite; IV - Cu, Sn and Pb sulphosalts and Ni and Co arsenides (stannite, mawsonite, boulangerite, bournonite, luzonite, famatinite, niccolite, etc.).

Stage V includes a specific telluride mineral association, without sulphides, subdivided to three successive sub-stages, presented by Ag-, Cu-, and Au- tellurides, respectively. The early sub-stage V includes:

hessite and sylvanite, often together with altaite, stützite, native Te, and an unnamed Ag-Cu sulphotelluride, $(\text{Ag,Cu})_{12}\text{Te}_3\text{S}_2$. The middle stage V includes melonite, rickardite, together with vulcanite, weissite, and native Te. The late stage V includes μm -sized *kostovite* associated with rickardite, krennerite, petzite, calaverite, and an unnamed Ag-Au-Cu sulphotelluride, probably $(\text{Ag,Au,Cu})_9\text{Te}_2\text{S}_3$. *Kostovite* has a low silver content (Table 1).

The next mineralization stages, related to clear quartz in the second group of veins, includes: in stage VI - galena, betekhtinite and argentite; in stage VII - sphalerite, together with greenockite and magnetite; and in stage VII - native Au, Ag, Cu, As, with stromeyerite and pyrrargyrite. Supergene oxides, carbonates, sulphides, and sulphates characterized the oxidation zone.

Microscopic observations established the presence of primary two-phase liquid- vapour and minor three-phase CO_2 bearing fluid inclusions. For the early milky quartz the measured T_h range is between 245 and 290°C, with a mean $275 \pm 5^\circ\text{C}$, whereas for the clear quartz the respective values are 219 and 250°C, with a mean of 235°C. The mean fluid salinities are 4.21 and 3.77 wt.% NaCl equiv.

Tombros et al. (2004), considering the paragenetic and temperature data, the evident high $f\text{Te}_2$ and low $f\text{S}_2$, and the modelling of Cooke and McPhail (2001), supposed a preferential transport of the tellurium species in vapour phase and telluride deposition, after condensation of the magmatically derived vapours into deep-level chloride waters in the surrounding area of the Tinos or similar leucogranite.

Other deposits

Occurrences of *kostovite* are also reported or mentioned from a number of other deposits:

- Buckeye Gulch, Leadville, Colorado, USA: Pb-Zn-Cu-Ag-Au ore deposit (Anthony et al., 1990).
- Kutemajärvi (also referred to as Kutema), Finland: a metamorphosed epithermal Au-Te ore deposit (e.g., Kojonen et al., 1999) in the

Orivesi District, near the city of Tampere is located within the Svecofennian Domain. *Kostovite* is one of a number of abundant tellurides in the gold ore, which occurs within five vertical ore 'pipes'.

- Coranda-Hondol, Metaliferi Mountain, Romania: Cu-Au-Te ore deposit (Udubaşa et al., 1992).

- Guilaizhuang gold deposit in Pingyi, Shandong, China (Liu Guangzhe, 1994).

- Our database of reported telluride occurrences in the Fennoscandian Shield (Ciobanu et al., 2004a) includes mentions (without microprobe data) from Enåsen (Sweden), Järvenpää and Jokisivu (Finland).

Although these deposits have been studied in detail by various authors, we have found little more information about the occurrence of *kostovite* within them.

Chemical composition and crystal chemistry

The compositional relationships between *kostovite* and sylvanite are especially interesting. In the first original microprobe analysis of Terziev (1966), *kostovite* is nearly Ag-free, containing only 0.5 wt.% (0.03 *apfu*) Ag. Subsequent determinations found that Ag is a common component of the mineral, and occurs in variable concentrations (Tables 1 and 2). Plotted on a $\text{CuTe}_2\text{-AgTe}_2\text{-AuTe}_2$ diagram, as a characteristic section of the general Au-Ag-Cu-Te diagram, all available published (~20) and our own unpublished (~40) analyses outline a wide area of intermediate compositions along the *kostovite*-sylvanite join. In fact, the points cross the "50% boundary" between the two compounds and cover, close to uninterrupted, the range between the pure *kostovite*, AuCuTe_4 , and $\text{Au}(\text{Cu}_{0.3}\text{Ag}_{0.7})\text{Te}_4$ (analysis K17 in Table 3). As is seen on Fig. 4, each deposit displays a distinct range of *kostovite* composition; this is especially wide in the case of Glava.

Kostovite often contains some Sb (Tables 1 and 2). In Chelopech it is in the range 0.43-1.58 wt.%. Although As strongly prevails in the environment of the deposit, it is absent in

Table 3. Representative microprobe analyses of cuprian sylvanites, wt.%
Таблица 3. Представителни микросондови анализи на мед-съдържащи силванити, тегл. %

Deposit	Kochbulak	Ashanti		Kamchatka	Zhana Tyube		
Authors	Kovalenker et al., 1979	Bowell et al., 1990		Sakharova et al., 1987	Spiridonov et al., 1974		
Sample	K18	E1277	RB59	S7	ZT-1	ZT-2	ZT-3
Au	23.98	24.60	24.69	28.0	24.99	25.31	25.17
Ag	11.99	11.12	11.06	8.9	12.04	10.56	10.50
Cu	0.68	1.17	1.07	1.5	1.03	1.14	1.16
Te	62.28	62.55	62.40	61.7	63.59	61.87	63.95
Sb	0.34			0.50			
Σ	99.27	99.44	99.22	100.6	101.65	98.88	100.78
<i>apfu</i>							
Au	0.99	1.02	1.03	1.16	1.01	1.06	1.03
Ag	0.91	0.84	0.84	0.67	0.89	0.81	0.78
Cu	0.09	0.15	0.14	0.19	0.13	0.15	0.15
Te	3.99	3.99	3.99	3.94	3.97	3.98	4.04
Sb	0.02			0.04			
Sum	6.00	6.00	6.00	6.00	6.00	6.00	6.00

kostovite. Up to 0.34 wt.% Sb was found and is also reported in kostovite from Kochbulak (Kovalenker, 1979). Minor Se (up to 0.32 wt.%) also occurs in some analyses from Chelopech. Bi substitution (up to 0.9 wt.%) is known from the Kamchatka occurrence.

Sylvanite, on the other side, occasionally contains some content of Cu, as has been mentioned by Spiridonov et al. (1974), Sakharova et al. (1987) and Howell et al. (1990) (Table 3). Such compositions thus cover the compositional range between AuAgTe₄ and Au(Ag_{0.85}Cu_{0.15})Te₄. As can be seen in Fig. 4, one analysis even falls into the "gap" between these two areas. At present, however, there is clearly insufficient evidence for the existence of a continuous solid solution series between the Cu- and Ag- golden ditellurides.

Another peculiarity is the spread (Fig. 4) of chemical compositions in a rather wide zone along the straight line kostovite-sylvanite, which traces out the stoichiometric ratio Au/(Cu+Ag) = 1. The variations outline an upper zone in which this ratio exceeds 1, reaching even up to 1.2-1.3 and expressing some excess of Au, and a lower zone with ratio <1, even below 0.8, expressing Au deficiency.

Such a distribution (if not related to analytical imprecision in the different laboratories) indicates a far more complex character to the isomorphic relationships.

Systematic compositional deviations in sylvanite, expressed as a dominance of Au over Ag, were noticed by Cabri (1965), for natural, as well as for synthetic sylvanite phases obtained in his experiments. Recent studies on the classic Au-Te deposit of Sacarimb, Romania (Ciobanu et al., 2004b), established that such distinct compositional non-stoichiometry with Ag/Au < 1, is typical for sylvanite. Three categories of sylvanite were found in this locality as lamellar intergrowths: 1 - close to stoichiometric (0.7 to 1.0 *apfu* Ag), 2 - low-Ag (0.7 to 0.4 *apfu* Ag), and 3 - very low-Ag (0.4 to 0.2 *apfu* Ag), the latter compositionally overlapping with krennerite. The structural nature of these categories and their intergrowths are not clear enough and demand further work.

The crystal structures of the gold-silver ditellurides, sylvanite, calaverite and krennerite, as shown by Tunnel and Pauling (1952) and Pertlik (1984a, b), are basically similar, although they have three different

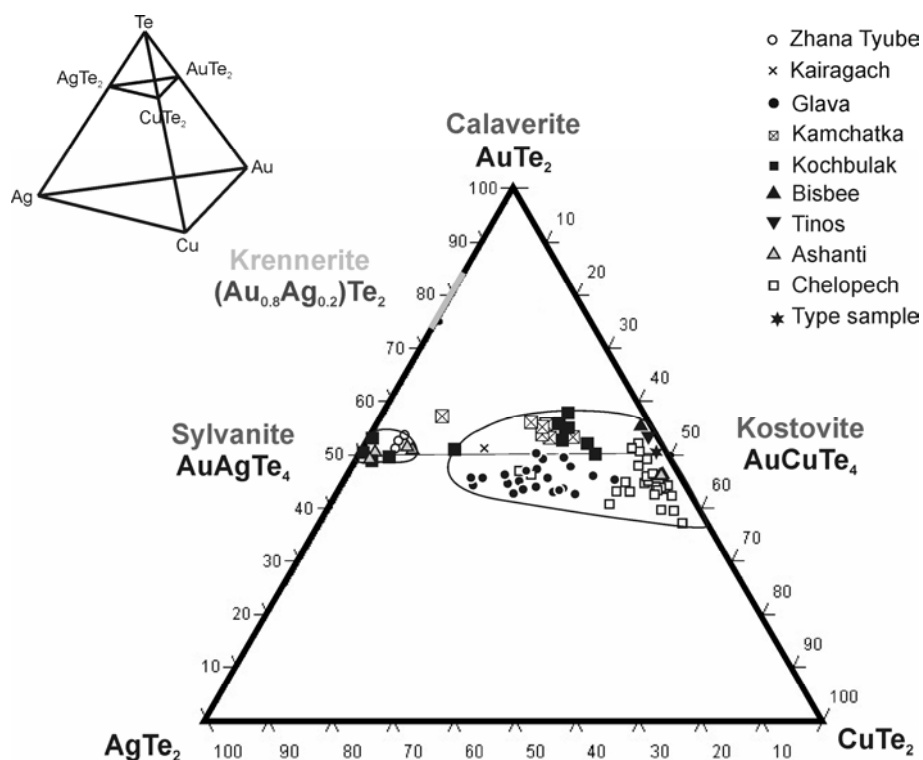


Fig. 4. Composition of kostovite-sylvanite minerals plotted on the AgTe_2 - CuTe_2 - AuTe_2 ternary diagram. Data of: Terziev (type sample, 1966), Kovalenker et al. (1981, 1986), Sakharova et al. (1987), Howell et al. (1990), Chvileva et al. (1990), Criddle & Stanley (1993), Petrunov (1994), Tombros et al. (2004), Spiridonov et al. (1974), and our new data

Фиг. 4. Състав на костовит-силванитови минерали, представени на триъгълна диаграма AgTe_2 - CuTe_2 - AuTe_2

space-group symmetries. Terziev (1966), on the base of composition and the similarity of the powder diffraction patterns, assumed that kostovite is isostructural with the monoclinic sylvanite. Later, Kovalenker et al. (1979) reported orthorhombic symmetry for kostovite material from Kochbulak. The determined unit cell parameters a 16.50(5), b 8.84(2) and c 4.42(2) Å, are directly comparable with those of krennerite. However, on the kostovite Weissenberg $h0l$ X-ray pattern, as mentioned by these authors, some additional reflexes do not correspond to the space group $Pma2$ of krennerite. Despite, these data and this space group are widely accepted as representative for

kostovite (PDF # 080020; Anthony et al., 1990; Gaines et al., 1997).

VanTendeloo and Amelinckx (1986) provided detailed high-resolution transmission electron-microscopic (HRTEM) investigations on synthetic kostovite, sylvanite, calaverite and krennerite. For kostovite, they proposed the structural model of orthorhombic sylvanite, as determined by Tunnel and Pauling (1952), in which Cu substitutes the Ag atoms. They mention however, that founding no evidence for a Cu homologue of krennerite, cannot discuss the structure of such a phase. The electron diffraction and HRTEM studies found commensurate and incommensurate modulated

structures of kostovite and sylvanite depending on composition and non-stoichiometry, and on heat treatment. Although, these phases have been produced from high-temperature (550°C) melts at non-geological conditions, it is highly probably that similar structures can be formed in lower-temperature hydrothermal telluride minerals.

For now, however, we can conclude that the details of crystal structure of the mineral kostovite are insufficiently understood. The crystallography of the characteristic intensive twinning of kostovite is a further aspect demanding explanation. Analytical difficulties are exacerbated by the typically small size of the natural crystal grains available for study up until now.

Based on the information currently available, it seems more realistic that the miscibility in the discussed natural solid solution series is between kostovite and sylvanite (and not krennerite). This probably does not reflect a simple Ag-Cu substitution alone, but the presence of far more complex modulated structures with non-stoichiometric domains and different sequences of ordering/disordering. If a gap in this series really exists, we can accept that compositions $\text{Au}(\text{Cu}_{1.0-0.3}\text{Ag}_{0.7})\text{Te}_4$, belong to kostovite (and argentian kostovite), while those of composition $\text{Au}(\text{Ag}_{1.0-0.85}\text{Cu}_{0.15})\text{Te}_4$ belong to sylvanite (and cuprian sylvanite). Some intermediate compositions determined as cuprian sylvanite, like $\text{Au}(\text{Cu}_{0.4}\text{Ag}_{0.6})\text{Te}_4$, described by Kovalenker et al. (2003) in the Kairagach deposit from the region of Kochbulak, should thus be considered as argentian kostovites (Fig. 4).

The variations in composition, of course, influence and the physical properties of kostovite but systematic observations are absent. However, by comparing the published reflectivity data for a nearly Ag-free kostovite from Bisbee (with 0.5 wt.%, and 0.04 *apfu* Ag: Criddle, Stanley, 1993) and for the Ag-rich kostovite from Kochbulak (3.9 wt.% and 0.23 *apfu* Ag: Chvileva et al., 1988) is seen that the argentian kostovite shows some lower values, deviating up to -4.7% in the central area of the

optical spectrum. Additionally, and the Au content can be important.

Discussion: Telluride parageneses

Up until now, kostovite was found in various predominantly gold, gold-copper, gold-telluride and complex gold-polymetallic epithermal ore deposits from different geographic and geologic positions. The ages of these deposits vary from Archean-Proterozoic (Ashanti, Glava, Commoner mine) and Palaeozoic (Kochbulak, Bisbee, Bereznyakovskoje), up to Cretaceous (Senonian for Chelopech), Tertiary (Moctezuma, Panormos Bay) and Neogene (Kamchatka).

As noticed by *Bowell et al.* (1990), these deposits can be assigned to two major genetic types: volcanic-hydrothermal and plutonic-hydrothermal. The volcanic deposits are clearly related to volcanic terrains and volcanic structures, which is the case with Chelopech, Kamchatka and Bereznyakovskoje. The plutonic-hydrothermal deposits with quartz veins and replacement bodies are often spatially associated with granodiorite or similar intrusions and have a greater vertical continuity than the volcanic type, in some cases extending over intervals of 2000 m or more. Ashanti and Commoner mine are typical representatives of this type (*Bowell et al.*, 1990). Panormos Bay and Glava would appear to be similar. The Campbell orebody at Bisbee can be considered intermediate between the plutonic and volcanic types of mineralization (*Criddle et al.*, 1989); Kochbulak (*Kovalenker et al.*, 1997) also fits such a category.

The various deposits display paragenetic similarities yet some notable differences. The main ore mineralization in these deposits is presented by early quartz with native gold of high fineness, commonly with pyrite, abundant copper-iron sulphides (chalcopyrite, bornite, chalcocite, tetrahedrite-tennantite, enargite-luzonite-famatinite, etc.), also with galena and sphalerite (Tabl. 4).

Kostovite, always associated with Au-Ag-Cu tellurides is a later, lower-temperature reaction product formed by overprinting of the

Table 4. Mineral associations in the deposits of kostovite: main sulphide minerals and overprinted tellurides
 Таблица 4. Минерални асоциации в находища на костовит - главни сулфидни минерали и наложени телуриди

Minerals / deposits		Chelo- pech	Koch- bulak	Kam- chatka	Ashanti	Glava	Bisbee	Mocte- zuma	Tinos
Pyrite, quartz	FeS ₂	+	+	+	+	+	+	+	+
Chalcopyrite	CuFeS ₂	+	+	+	+	+	+	+	+
Enargite	Cu ₃ AsS ₄	+	+			+	+	+	
Luzonite	Cu ₃ AsS ₄	+	+			+	+		+
Famatinite	Cu ₃ SbS ₄	+	+			+	+		+
Tennantite-tetr.	Cu ₁₂ (As,Sb) ₄ S ₁₃	+	+		+	+	+	+	+
Bornite	Cu ₅ FeS ₄	+	+		+	+	+	+	+
Chalcocite	Cu ₂ S	+	+		+	+	+	+	+
Sphalerite	ZnS	+	+	+	+	+	+	+	+
Galena	PbS	+	+	+	+	+	+	+	+
Clausthalite	PbSe	+				+		+	
Pyrrhotite	Fe _{1-x} S	+	+		+		+		+
Arsenopyrite	FeAsS				+	+			+
Bismuthite	Bi ₂ S ₃	+	+			+	+	+	
Aikinite	CuPbBiS ₃	+	+			+	+		
Wittichenite	Cu ₃ BiS ₃	+	+			+	+	+	+
Hematite/goethite	Fe ₂ O ₃ /FeOOH				+	+	+	+	
Native gold	Au	+	+	+	+	+	+	+	+
Native tellurium	Te	+	+		+	+	+	+	+
Goldfieldite	Cu ₁₀ Te ₄ S ₁₃	+	+	+		+	+	+	+
Altaite	PbTe	+	+	+	+	+	+	+	+
Volynskite	AgBiTe ₂		+			+	+		
Calaverite	AuTe ₂	+	+	+	+	+	+	+	+
Sylvanite	AuAgTe ₄	+	+	+	+	+	+	+	+
Kostovite	AuCuTe ₄	+	+	+	+	+	+	+	+
Krennerite	(Au _{0.8} Ag _{0.2})Te ₂	+	+			+	+		+
Petzite	Ag ₃ AuTe ₂	+	+	+	+	+	+	+	+
Montbrayite	(Au,Cu) ₂ Te ₃	+	+				+	+	
Nagyagite	Pb ₅ Au(Te,Sb) ₄ S ₈	?	+		+				
Henryite	Cu ₄ Ag ₃ Te ₄				+		+	+	
Rickardite	Cu _{4-x} Te ₂		+		+	+	+	+	+
Weissite	Cu ₂ Te		+		+	+			+
Vulcanite	CuTe								+
Bambollaite	Cu(Se,Te) ₂							+	
Tellurobismuthite	Bi ₂ Te ₃	+	+			+	+		
Tetradymite	Bi ₂ Te ₂ S	+	+			+	+	+	
Kawazulite	Bi ₂ Te ₂ Se	+				+			
Rucklidgeite	(Bi,Pb) ₃ Te ₄			+		+	+		
Hessite	Ag ₂ Te		+	+	+	+	+	+	+
Stützite	Ag ₂ Te _{1,2}		+		+	+	+	+	+
Empressite	Ag ₅ Te ₃		+			+			
Cervelleite	Ag ₄ TeS							+	
Tellurantimony	Sb ₂ Te ₃		+						
Coloradoite	HgTe	+	+		+		+	+	
Melonite	NiTe ₂		+		+	+	+		+
Geochemistry:		Cu, Au, Te, Ag, Bi, Se, As, Pb, Sn	Au, Ag, Te Cu, Bi, Sb, Pb, Se	Au, Ag, Te, Cu	Au, Te, Ag, Cu	Cu, Ag, Bi, Te, Se	Cu, Pb, Zn, Ag, Au, Te, Bi	Cu, Pb, Zn Au, Ag, Te Bi, Sn	Cu, Au, Ag, Te, Pb
T°C: Main ores		300-200	400-150		340-267	335-145			320-280
Late tellurides			250-200		220-165				260-230

main ores by later stage fluids with changed composition and character. These telluride assemblages always have complex composition, representing mutual intergrowths of several mineral phases, favoured by the high mobility of Au and Cu.

In most cases, some earlier specific Te-bearing minerals precede the deposition of the more complex late telluride association with kostovite, thus indicating an evolution of fluid composition.

Cu-Te. In the major Cu-Au deposits, Chelopech and Kochbulak, the association of goldfieldite-tetrahedrite-tennantite with Au and Ag tellurides is decisive. *Goldfieldite*, as the earliest and very important Te mineral from the main ore stage (arsenian goldfieldite in Chelopech, and antimonian goldfieldite in Kochbulak), is replaced by kostovite by means of later introduced Au-bearing fluids. Various other tellurides also formed, including calaverite, sylvanite, krennerite, petzite, montbrayite, altaite, coloradoite, as well as native Te, some Bi-sulphotellurides (telluro-bismuthite, tetradyomite), Ag-Pb-Bi sulphosalts, and some selenides (clausthalite, kawazulite). Because the goldfieldite grains are dispersed into the early ores, dispersed are also the new-formed tellurides. Similar, but more uniform is the mineralization in Kamchatka (goldfieldite, with sylvanite, kostovite, petzite, hessite, altaite, rucklidgeite).

Bi-Te. In Glava the earliest and dominating telluride, *tellurobismuthite*, formed segregations in the main high-*T* bornite ore. By successive reactions with introduced **Ag + Pb** it was replaced by hessite, volynskite, tsumoite and native tellurium, after later **Au + Cu** input was followed by gold, petzite, kostovite, sylvanite, kawazulite, and at last by melonite. Hematite also occurs.

Te. Peculiar is the case of Moctezuma where the coarse-crystal *native tellurium* is the prevailing early ore tellurian mineral, which proposes the suitable environment for kostovite, and a great variety of other tellurian minerals precipitated also and directly from the fluids.

In some other deposits the early low-sulphidation mainly pyrite sulphide bodies are overprinted by later Te-bearing associations of complex mineral composition.

Ag-Pb-Te. In Ashanti, the early quartz-arsenopyrite-pyrite-gold mineralization does not include Te minerals. The time-restricted input of Te-rich fluids replaced the early gold, thus formed localized small pods of Au-Cu-Te mineralization. This later *copper-telluride association* includes chalcocite, bornite, altaite, coloradoite, hessite, stützite, as also sylvanite, kostovite, and calaverite in fine symplectitic intergrowths with goethite and hematite. Analogous is the case with Commoner mine. Similarly, in Panormos Bay the late telluride association overprinted on the early, copper-polymetallic mineralization, includes successively formation of **Ag-Pb** tellurides, followed by **Cu** tellurides, and then by **Au-Cu** tellurides, in which kostovite associated with calaverite, krennerite, petzite, rickardite and an unnamed Ag-Au-Cu sulphotelluride.

In Bereznyakovskoe early pyrite and following it Cu-tennantite are overprinted by an association of Pb-, Bi- and Au-Ag-Cu tellurides replacing the early sulphides and forming fine intergrowths also with native gold.

In accordance with the thermodynamic calculations, diagrams and main conclusions of Afifi et al. (1988), the telluride associations containing kostovite in all these deposits are characterized by similar and specific physico-chemical environment:

- relatively highly oxidation conditions, as indicated by the abundance of hypogene hematite or goethite (Ashanti, Commoner mine), barite (Chelopech, Kochbulak, Moctezuma), highest oxidation state of semimetals (Te^{4+} , As^{5+} , Sb^{5+}) in goldfieldite, enargite, famatinite, etc.;
- high $f\text{Te}_2/f\text{S}_2$ ratio and low $f\text{S}_2$, indicated by the usual absence of sulphides in the telluride parageneses, with only rare sulphotellurides in some cases (as in Panormos Bay);
- rather low *T* of formation (Table 4), mostly in the 250-200-170°C range, as estimated by the different authors, based on studies of fluid inclusions and paragenetic relationships.

Interesting and important are problems of the origin, transport and deposition of Te, Au, and other components of the hydrothermal fluids and mineral parageneses. Afifi et al. (1988) argued that geological and physicochemical data are suggestive of magmatic origin of tellurium, the Te-enriched fluids being limited in volume and time, which corresponds to the observations reported here. Cooke and McPhail (2001) presented a numerical model of telluride deposition in the epithermal Acupan Au-Ag telluride deposit, Philippines. They proposed that Te is transported in the magmatically-derived vapour phase rather than the aqueous phase and that its condensation into precious metal brines is the main mechanism for deposition of telluride ores. This mechanism is accepted by Tombros et al. (2004) for the Te mineralization in Tinos Island.

Shackleton et al. (2003) insist that multiple mechanisms of gold-telluride deposition can operate (phase separation, wallrock interaction, cooling, boiling and throttling, condensation and fluid mixing), accounting for the heterogeneous distribution of tellurides in many deposits. Evidently, as they point out, a complex interaction of factors can be evaluated in each case, including host-rock composition, structural conditions, fluid characteristics and composition, detailed mineral relationships, alteration assemblages, regime of T and P , etc.

Conclusions

Kostovite is a specific minor component of the rich telluride parageneses in a number of Au, Au-Cu, and complex Au-polymetallic epithermal deposits, formed in Cu-rich environment at conditions of increased fO_2 , high fTe_2/fS_2 and low fS_2 . The mineral is probably more widespread than currently perceived.

Kostovite is formed in the late, low-temperature mineralization stages, mostly as a reaction product after earlier high-Te minerals, mostly of:

- Cu: *goldfieldite* in Chelopech, Kamchatka, Kochbulak,

- Bi-Te: *tellurobismuthite* in Glava,
- *native tellurium* in Moctezuma.

In other deposits, the early pyrite and copper sulphide assemblages are overprinted by a later complex Au-telluride mineralization, commonly in fine mineral intergrowths, as is the case with Ashanti, Commoner Mine and Panormos Bay.

Extensive incorporation of Ag in natural kostovite is established in the kostovite-sylvanite series, ranging between 0 and 70 at.%. In sylvanite, exchange of Ag by Cu is much more limited, not exceeding 15 at.%. Some deviations in stoichiometry from the $Au/(Ag+Cu) = 1$ ratio also exist. Often, some Sb is also included.

The composition of kostovite from the different deposits has own specificity: in Chelopech, Ashanti and Bisbee, formed in high-Cu (and Au) environment, it has low Ag-content, in Kochbulak and Kamchatka the Ag content is higher, and in Glava (in Cu-Ag-Bi-rich environment) the variations are largest. As concerning Au, some deficit is found in Glava and Chelopech, and some excess, in Kochbulak.

Although requiring confirmation, isomorphism in the telluride minerals has, most probably, a complex character with the existence of incommensurate modulated structures of different degree of ordering. Extensive disordering has been revealed by the detailed HRTEM investigations carried out by Van Tendeloo et al. (1986) on high-temperature synthetic kostovite and other Au-Ag tellurides.

Some problems for future studies

- Systematic investigation of gold-telluride mineralogy, paragenetic relationships and genesis of Au-Ag-Te deposits. Identification and characterization of macroscopic-scale kostovite, necessary for additional crystallographic, physical and other studies.
- Experimental investigation of the systems Au-Cu-Te and Au-Ag-Cu-Te and determination of stability fields for the different mineral phases.

- Refinement of the crystal structure of natural and synthetic kostovite and explanation of the widespread crystal twinning.
- High-resolution electron microscopic (HRTEM) studies on natural kostovite and sylvanite specimens of different composition and conditions of formation for explaining their structural modulations and character of the miscibility and non-stoichiometry.

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