

EPITHERMAL GOLD MINERALIZATION IN THE KRASSEN DEPOSIT, PANAGYURISHTE ORE DISTRICT, BULGARIA

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A b s t r a c t: Gold mineralization in the Krassen high-sulphidation epithermal deposit, Panagyurishte ore district, Bulgaria, has been studied in respect to mineral assemblages, gold grain size, fineness and trace elements based on EPMA and LA-ICP-MS studies. Part of the gold in the early massive pyrite dominated ores is submicroscopic in size ($< 0.1 \mu\text{m}$) and could be attached to the so called “invisible” gold. Later fracturing of the early massive pyrite, followed the deposition of Cu-pyrite ore bodies enriched in chalcopyrite, enargite, bornite, galena and sphalerite accompanied by deformation and recrystallization is suggested as a reason for Au and Ag migration to cracks and gold grains coarsening. The electrum fineness in individual grains varies between 882 and 998 ‰. Most commonly observed trace elements in the native gold, electrum grains and gold hosting sulphide minerals are: Cu, Fe, Hg, Sb, Te, Bi, As and Se. The Au content in pyrite varies from 0.35 to 7.83 ppm and in chalcopyrite from 0.97 to 2.78 ppm. Bi-Te-Se and Ga-Ge-In trace elements signature is characteristic feature of the ore minerals and indicator for Au enrichment.

Key words: epithermal gold; Panagyurishte ore district; high-sulphidation gold, LA-ICP-MS

INTRODUCTION

The Panagyurishte ore region (Figure 1) in the Srednogorie zone of Bulgaria that is part the Late Cretaceous Apuseni-Banat-Timok-Srednogorie (ABTS) metallogenic zone, which lies entirely in the western part of the global Tethyan-Eurasian orogenic belt (Janković, 1977; Bogdanov, 1987; Dabovski, 2010; Sillitoe, 2010; Popov et al., 2012). The Srednogorie zone is interpreted as part of an island-arc system, developed during the Late Cretaceous as a result of NE subduction of the Vardar paleocean toward Serbo-Macedonian and Rhodope massifs. (Kamenov et al., 2003; Dabovski, 2010). The Panagyurishte ore region is typified by thick volcanic sequences and relatively shallow

intrusions (Figure 1) that host over 150 Cu-Au epithermal and porphyry-copper deposits and mineral occurrences.

The Krassen epithermal Cu-Au deposit (Figure 2) is located in an 80–100 m thick fault zone, limited by two sub-parallel fault arms with trend of 110–115° and 50–65° dip to the NE. Three gold-bearing stages of mineralization *Early Pyrite stage*; *Main Copper-pyrite stage* and *Late Anhydrite-barite stage* are well developed in Krassen HS epithermal Cu-Au deposit. The gold grade in the pyrite-enargite and Cu-pyrite ores as a rule is between 0.5 and 4 g/t.

GEOLOGICAL SETTING

Elshitsa, Radka and Krassen Cu-Au epithermal deposits, as well as numerous ore occurrences (Chervena Mogila, Izgorelia Vruh, Kaleto, Borova Mogila etc.) are characteristic for the southern part of the Panagyurishte ore region (Figure 1). The

latter consists of: Pre-Upper Cretaceous metamorphic crystalline basement; Triassic sedimentary rocks; Turonian-Lower Senonian volcano-sedimentary rocks, and Upper Senonian, Tertiary and Quaternary sedimentary rock complexes (Figure 1).

The deposits and occurrences are situated in volcano-tectonic depressions that can be interpreted as pull-apart basins (Ivanov, 2001; Drew, 2005). The epithermal deposits are associated with small porphyry copper Cu-Au ore and occurrences, that were formed as a result of the evolution of individual Late-Cretaceous volcano-intrusive centers, such as Elshitsa–Vlaikov Vruh, Radka–Tsar

Assen, Krassen–Petelovo (Bogdanov, 1987; Popov et al., 2012). The intrusion centered complexes have been formed as a result of the development of the following three successive stages of the magmatic activity: volcano-plutonic (with andesite-diorite and dacite-granite phases), subvolcanic (with subvolcanic dacite and granodiorite-porphyry phases) and final volcanic (trachyandesite-basalt).

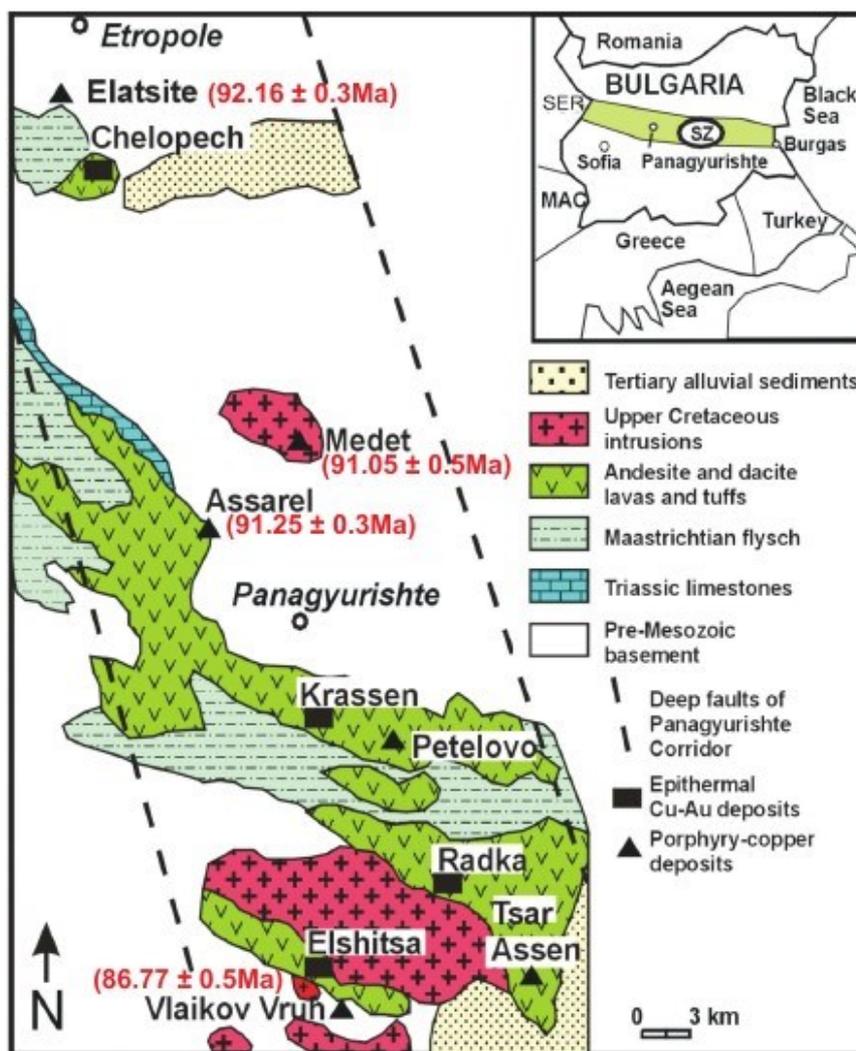


Fig. 1. Sketch map of the Panagyurishte ore region

(Modified from Bogdanov, 1987; Bogdanov, Popov 2003. The Re/Os age data in molybdenite are after Zimmerman et al., 2008.)
SZ – Srednogorie zone

The Krassen epithermal Cu-Au deposit (Figure 2) is hosted in the Krassen–Petelovo volcano-plutonic complex that is located in an 80–100 m thick fault zone, limited by two sub-parallel fault arms with trend of 110–115° and 50–65° dip to the NE (Popov et al., 2012). The pyrite-energite dominated mineralization is tectonically controlled by series of subparallel developed tectonic breccia zones (Figure 3) that host the pyrite-energite ore

bodies (Tsonev et al., 2000; Bogdanov, Popov, 2003). Abundant tectonic clay and sulphide ore brecciated mineralization are evidences for later reactivation of the northern part of the Krassen fault zone (Figures 2 and 3). The N-NE trending diagonal faults that crosscut and displace subequatorial faults have also been developed (Popov et al., 2012).

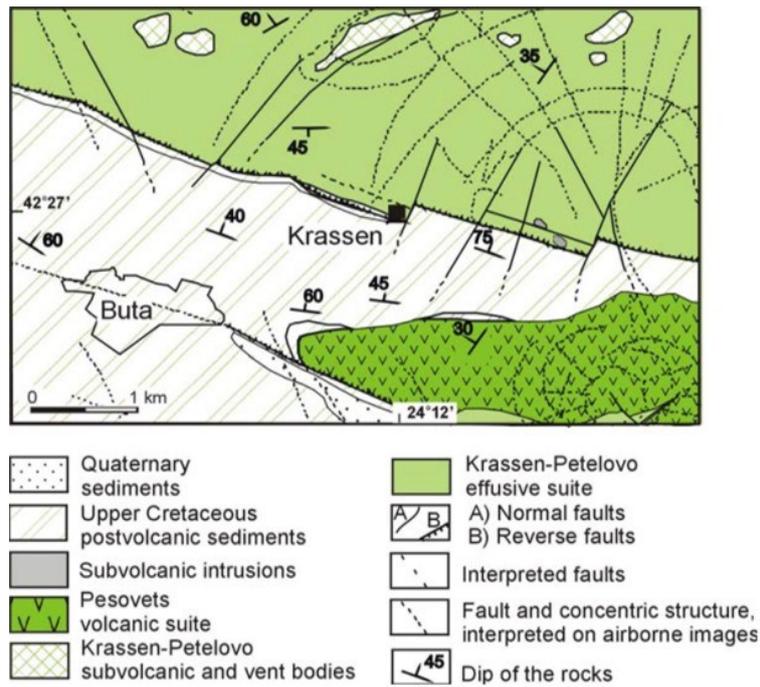


Fig. 2. Geological map of Krsasen epithermal Cu-Au deposit (After Popov and Popov, 2000)

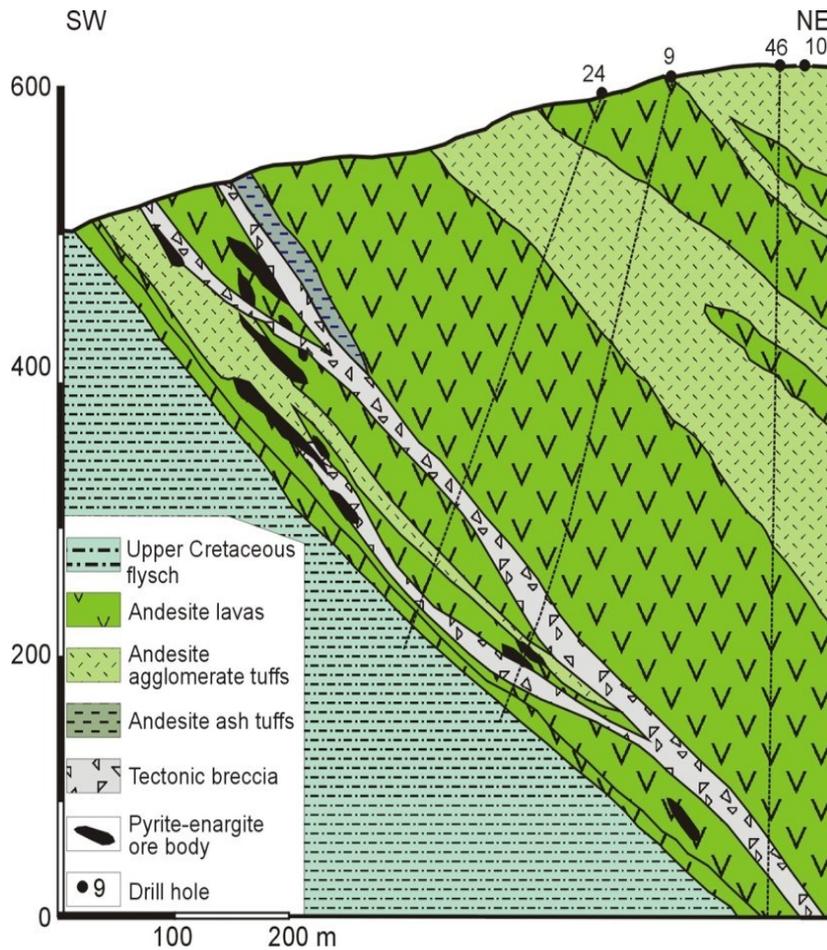


Fig. 3. Cross-section of Krassen epithermal deposit (After Tsonev et al., 2000; Bogdanov, Popov, 2003)

Pipe-like tectonic breccia zone hosts individual vein, lens-like and columnar ore bodies in the Krassen deposit (Figure 3). Quartz-sericite, adularia-sericite, propylitic and advanced argillic quartz-pyrophyllite and kaolinite-alunite host rock alterations are commonly observed around the ore bodies (Radonova, 1967; Popov et al., 2012). The successive high sulphidation hydrothermal phases of the Krassen–Petelovo volcano-plutonic system overprinted the pyrite ore bodies with chalcopyrite, enargite, bornite, tennantite chalcocite and gold mineralization in the Krassen epithermal deposit.

HOST ROCK ALTERATIONS

Both the volcanic and intrusive rocks in the southern part of the Panagyurishte ore region have been affected by abundant pervasive propylitic (albite-chlorite-epidote \pm calcite) alterations. Adularia-sericite, acid-sulphate, propylitic, and advanced argillic styles of alterations are the most common ones in the andesitic and dacitic volcanic rocks in addition to the K-feldspar, sericite and chlorite alterations of the subvolcanic diorite and granite porphyry intrusions (Radonova, 1970; Tsonev et al., 2000). Quartz-sericite, acid-sulphate and advanced argillic alterations of the volcanic and subvolcanic rocks in the Radka, Krassen (Fig-

The ore zone is developed along subvolcanic andesite intrusion and has 300 to 100 m in size ellipse-like section, NE dip of about 50° and is traced down to 700 m in depth. Most of the ore bodies are lenticular and lense-like shaped (Figure 3). The enargite-chalcopyrite-bornite mineralization is dominating in the western part of Krassen deposit. Massive pyrite ore, replaced by chalcopyrite along the contacts, and low-grade vein pyrite mineralization are commonly observed in the eastern part of the deposit (Bogdanov, Popov, 2003; Popov et al., 2012).

ures 1 and 2), Chervena Mogila and around the peaks of Petelovo, Borova Mogila, and Pessovets are represented by epidote-chlorite-albite, quartz-sericite, sericite-diaspore, quartz-pyrophyllite, quartz-kaolinite-dickite, quartz-topaz, quartz-alunite and monoquartzite facies (Kanazirski, 2011). Mushroom shaped and lens-like (700 \times 300 \times 300 m) pervasive acid-sulphate (quartz-alunite and quartz-diaspore) alterations are most commonly observed around the peaks of Petelovo, Borova Mogila, and Pessovets, while the quartz-sericite alterations are characteristic for Elshitsa and Radka (Popov et al., 2012).

ORE MINERALOGY

The ore-forming processes in the Krassen deposits have been developed into three stages of mineralization with seven mineral assemblages: *Early Pyrite stage* with (1) Pyrite-quartz assemblage; *Main Copper-pyrite stage* with (2) Chalcopyrite-pyrite; (3) Enargite-covellite; (4) Bornite-tennantite and (5) Galena-sphalerite-chalcopyrite; *Late Anhydrite-barite stage* with (6) Quartz-pyrite (7) Pyrite-marcasite and (8) Anhydrite-barite assemblage. The early massive pyrite-quartz assemblage (Figure 4a) is overprinted the enargite-covellite (Figure 4b) chalcopyrite-pyrite, bornite-tennantite and galena-sphalerite-chalcopyrite assemblages (Figure 4c). All of them are crosscutted and overprinted by late quartz-pyrite, pyrite-marcasite and anhydrite-barite assemblages (Figure 4d). The pyrite stage is represented mainly by colloform-textured pyrite (Figure 4a), quartz, and submicroscopic gold intergrown with pyrite. The Cu-pyrite stage consists of several mineral assemblages (chalcopyrite-pyrite, enargite-covellite, bornite-tennantite, galena-sphalerite-chalcopyrite). Enargi-

te is the dominant sulphide (Figure 4b), while chalcopyrite, tennantite -tetrahedrite mineral series, bornite, covellite and chalcocite are subordinate. Enargite is abundant in the Krassen deposit and together with covellite is evidence for high sulphidation mineralization style (Bogdanov, Popov, 2003; Sillitoe, Hedenquist, 2003; Popov et al., 2012). Native gold and silver that are of primary economic importance for the studied area, are commonly observed as microscopic blebs and irregular grains associated with enargite, chalcopyrite, bornite, tennantite, covellite, galena and sphalerite, or situated along their grain boundaries. The galena-sphalerite-chalcopyrite mineral assemblage is represented by galena, clausthalite, sphalerite, pyrite, bornite, chalcopyrite and the tennantite tetrahedrite mineral series. Aikinite, clausthalite, roquesite, germanite, gallite and hessite have been rarely observed (Bogdanov, Popov, 2003).

All the sulphide minerals are Bi, Ge, Ga, In, Te and Se bearing. The late quartz-anhydrite stage consists of quartz-pyrite, barite, anhydrite veins

cross-cutting and replacing all the minerals from the earlier mineralization stages. In Krassen, the late quartz-pyrite vein assemblage is not so abundant. The late anhydrite-barite mineral assemblage is represented by anhydrite, gypsum, barite and calcite veins and lenses replacing the sulphide ore (Figure 4c,d). The sulphate minerals are accompa-

nied by minor amounts of pyrite, chalcopyrite, galena, tennantite, sphalerite, gold, silver, calcite, fluorite and quartz. Marcasite found in assemblage with pyrite indicates that the hydrothermal conditions were quite acid in the late mineral assemblages.

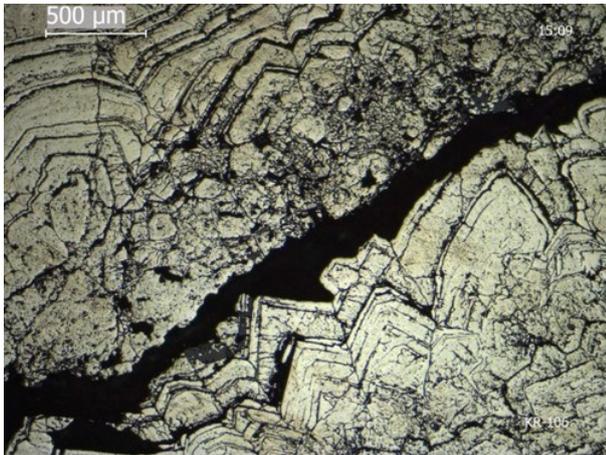


Fig. 4a. Early massive pyrite with zoning pattern

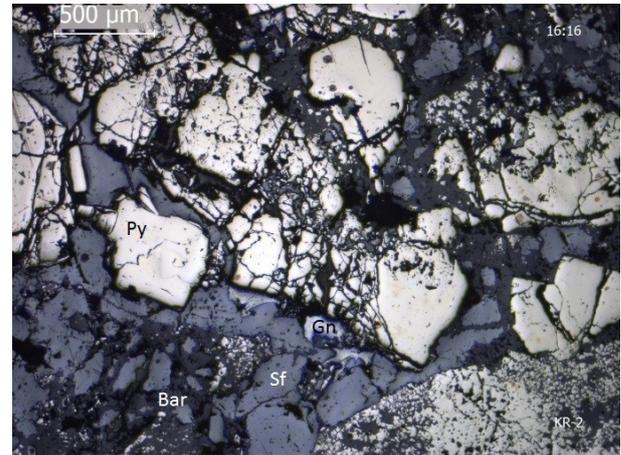


Fig. 4c. Galena (Gn), sphalerite (Sf) and barite (Bar) overprinting brecciated early pyrite (Py)

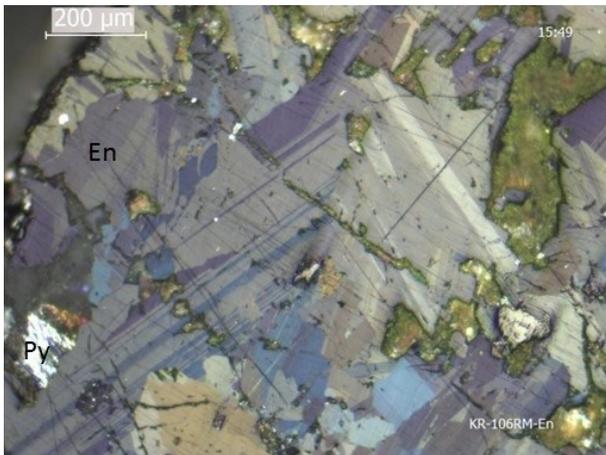


Fig. 4b. Enargite (En) replacing pyrite (Py)

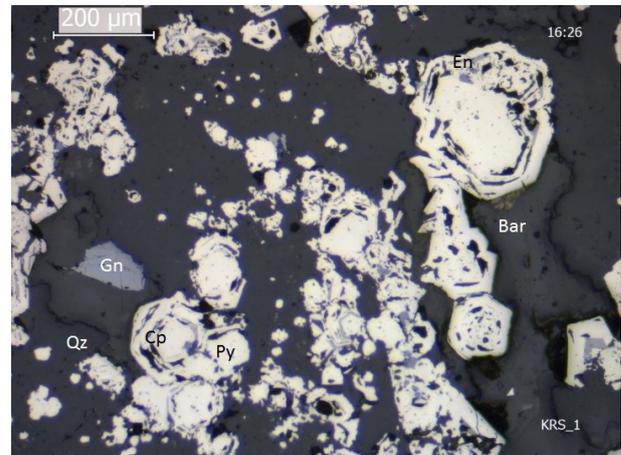


Fig. 4d. Chalcopyrite (Cp), enargite (En) and galena (Gn) replacing pyrite (Py) and all overprinted by barite (Bar) and quartz (Qz)

THE GOLD-SILVER MINERALIZATION

Native gold and electrum are commonly observed in the enargite-pyrite and galena-sphalerite-chalcopyrite assemblages in Krassen HS epithermal deposit. The pyrite veinlet and disseminated ore bodies are poor in gold and other precious metals, as compared to the ore bodies with more complex mineral composition. The gold grade in the Cu-pyrite ores is 0.1–5 g/t, rarely up to 10–12 g/t.

Morphology and grain size

Native gold and electrum often occur in the form of microscopic anhedral grains that do not exceed 100 µm. Individual grains up to 1–2 mm in size are rarely found. Gold is generally found as a microscopic irregular single grains, commonly flattened or elongate. Electrum and gold occur as

blebs or irregular particles in the sulphide minerals. Microscopic anhedral grains, rosary arranged blebs or irregular particles of native gold and electrum are commonly associated with pyrite, enargite, bornite, tennantite, chalcopyrite, sphalerite, galena, clausthalite, covellite and chalcocite. Most of the observed gold grains are microscopic and do not exceed in size 2–20 μm . Gold blebs in size of 40–100 μm have been rarely found.

Trace elements in gold

The most commonly found trace elements in the native gold and electrum grains are: Cu, Fe, Hg, Sb, Te, Bi, As and Se. Their content varies in the range from 0.1 to 2.21 wt%. The Bi content in gold and electrum (Table 1) varies from 0.51 to 0.05 wt%, Te from 0.15 to 5.75 ppm.

Gold fineness

The gold fineness in the studied gold grains from Krassen epithermal deposit as a rule is high and varies between 882 and 998 ‰ (Figure 5, Table 1). The early quartz-pyrite assemblage is characterized by gold fineness within the range of 930–963 ‰. In the chalcopyrite-pyrite, enargite-pyrite, bornite-tennantite and galena-sphalerite-chalcopyrite assemblages of the main copper-pyrite stage the gold fineness is between 880 and 984 ‰. In the late anhydrite-barite assemblage the gold fineness varies between 990 and 995 ‰. Kustelite and native silver have been also found in the late anhydrite-barite assemblage (Table 1).

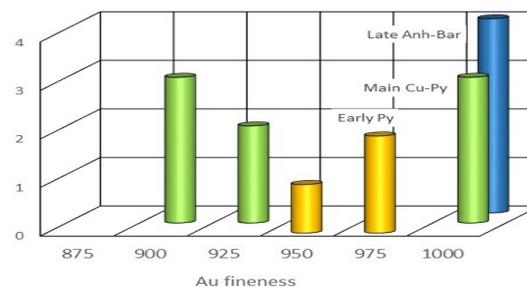


Fig. 5. Gold fineness in the early, main and late stages of mineralization in Krassen epithermal deposit

Trace elements in the gold host minerals

Titanium, Ge, As, Sb, Ag and Tl (Table 2) are the most common trace elements in the early massive pyrite that has been replaced by enargite, chalcopyrite, galena and sphalerite. LA-ICP-MS data (Perkin-Elmer ELAN DRC with New Wave UP193FX LA device, Geological Institute – BAS) indicate the presence of refractory submicroscopic type (<1 μm) gold mineralization in the early massive pyrite ore bodies (Figure 6). The Au content in pyrite (Table 2) varies from 0.35 to 7.83 ppm, Ag from 0.15 to 5.75 ppm. High As (2224–2407 ppm) grades are probably due to mineral inclusions of enargite, while the Ti content could be due to rutile inclusions. Cobalt, Te and Tl rarely more than 50 ppm LA-ICP-MS data (Table 3) indicate Pd, Ni, Sn and In as most common trace elements in the chalcopyrite from the main Cu-pyrite mineralization stage. The Au content in chalcopyrite (Table 3) varies from 0.97 to 2.78 ppm. Cadmium, Mn, Ga, In, Tl, Ag and Hg are common trace elements in sphalerite with grades between 8 and 228 ppm (Table 3).

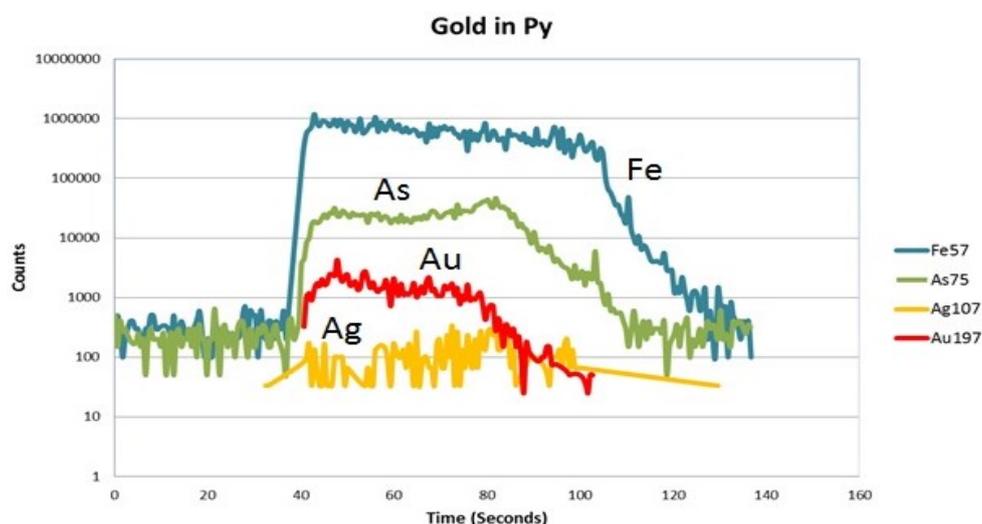


Fig. 6. LA-ICP-MS profile in early pyrite from Krassen epithermal deposit

Table 1

Microprobe data for gold-silver mineralization from Krassen epithermal deposit (in wt%)

Stage of mineralization	Au	Ag	Cu	Te	Bi	Hg	Sb	As	Fineness. ‰
Early pyrite	94.07	3.05	1.36	0.66	0.51		0.4		940
	93.35	3.48	2.21	0.32	0.91		0.1		930
	95.21	3.25	1.14	0.12	0.13		0.2	0.05	951
	94.07	3.05	1.36	0.51		0.66	0.4		940
	96.71	2.48	0.93		0.21		0.12		963
Main Cu-pyrite	98.17	0.66	0.14	0.15	0.36		0.24		984
	97.77	0.96	0.29	0.35	0.29		0.19		979
	97.77	0.96	0.29	0.29		0.35	0.19		979
	91.4	6.45	1.39	0.22	0.14		0.15		916
	88.65	8.95	1.6	0.2		0.31	0.33		886
	88.65	8.95	1.6	0.31	0.2		0.33		886
	88.2	11.75	0.24	0.05					880
	99.02	0.2	0.06	0.08	0.1		0.1	0.05	995
Late anhydrite-barite	98.44	0.46	0.14	0.07	0.05	0.15	0.05	0.1	990
	98.89	0.36	0.06			0.22		0.05	993
	99.02	0.2	0.06		0.08	0.1	0.1	0.05	994
	20.03	79.12	0.14	0.05		0.08	0.23	0.1	201
	6.51	91.46	1.71	0.12		0.15			65
	4.05	94.37	1.02	0.15		0.3			41

Table 2

LA-ICP-MS data for early pyrite from Krassen epithermal deposit (in ppm)

Sample	Krs 1131	Krs 1132	Krs 1133	Krs 1134	Krs 1135	Krs 1136	Krs 491	Krs 492	Krs 493
Ti	41.84	49.07	495.05	54.58	61.66	61.55	57.02	87.42	23.10
V		2.37	1.62	3.68	2.68	138.46	11.62	8.38	1.70
Cr	370.73	27.55	39.49	27.88	40.70	29.96		40.36	35.30
Co	3.71		3.38	3.07	0.71	0.91	1.42		0.70
Ni	3.20					6.74			
Ga		0.521				10.32			
Ge	6.95		7.33	6.42	5.19	58.04			6.42
As	10.18	26.8	982.4	2306.9	2407.9	2224.4	7.46	48.28	5.87
Se	325.6			18.21		40.92		19.73	
Mo				0.63		8.33		1.40	
Ag	0.15	2.77	0.27	0.97	4.55	5.75		0.87	
Cd			0.94	0.84				1.91	
In						0.38			
Sn			0.76	1.04		6.16			
Sb	2.92	0.91	10.24	20.22	66.09	370.30		1.60	
Te	6.42		18.94		9.20	18.82		10.93	
W			3.45		1.70	5.69			
Au		0.35	3.15	0.69	7.83	6.79			
Hg		0.93		0.68	2.60	1.23		2.57	
Tl		12.89	12.64	20.68	55.83	7.52		32.89	
Bi	4.15	1.03	10.52	18.69	7.50	31.29			

Table 3

LA-ICP-MS data for chalcopyrite and sphalerite Krassen epithermal deposit (in ppm)

Mineral Sample	Chalcopyrite				Sphalerite	
	Krs 46\1	Krs 46\2	Krs 46 \3	Krs 46\4	Krs121	Krs122
Ti	36.43	61.24	63.65	82.75	129.64	443.76
V	1.50					29.42
Cr	22.09	25.47		27.19		
Mn	25.67	27.22	26.96	28.60	13.09	13.87
Co			193.86	210.63		7.29
Ni	176.08	223.08				
Ga	3.92	11.65		11.71	170.34	127.58
Ge			4.19			62.84
As			511.98	172.43	35.40	197.40
Se	395.12	330.96		2.11		99.83
Mo			8.18	9.28		15.45
Pd	6.77	7.18	31.98	27.65		
Ag	29.30	37.47			11.12	31.56
Cd	2.11		7.71	8.45	1232.64	1210.02
In	6.50	16.94	218.65	32.33	12.79	8.28
Sn	104.30	514.00	8.38	1.65		
Te				2.56		28.23
Au			2.78	0.97		
Pb	5.62					
Bi	1.79					
Hg					218.45	228.80

DISCUSSION AND CONCLUSION

The andesite dominated island arc magmatic activity in the Panagyrishte structural corridor (Figure 1) has a life span of about 10 Ma, while the formation of the porphyry-epithermal system of Krassen–Petelovo seems to be limited to more narrow time interval of 1–3 Ma (Von Quadt et al., 2005; Zimmerman et al., 2008; Popov et al., 2012). Small shallow basins with carbonate and flysch sedimentation are characteristic for the Maastriichtian when the volcano-plutonic activity has ended. Most of the porphyry-epithermal systems are attached to positive gravitational anomalies located along deep fault submeridional structure framing the Panagyrishte structural corridor (Figure 1). The intersections of the NW trending (310–320°) faults framing the pull-apart basins with the submeridional Panagyrishte deep fault are regarded as an important magmatic and ore-controlling structure (Ivanov, 2001; Drew, 2005; Popov et al., 2012). The formation of discrete coeval volcano-plutonic centers with porphyry-epithermal mineralization is triggered by strike-slip faults in the margin of narrow pull-apart basins. (Ivanov, 2001; Popov et al., 2012).

Gold in the early pyrite dominated ores in Krassen is of submicroscopic type (< 0.1 µm) and can be attached to the so called “invisible” gold (Cook and Chryssoulis, 1990; Bogdanov et al., 1997). The electrum fineness in individual grains is high and varies between 880 and 995 (Figure 5) that is close to the fineness for the epithermal class of mineral deposits (Morrison et al., 1991; Bogdanov et al., 1997). Fracturing of the early massive pyrite prior to recrystallization in response to strain-induced deformation, followed by tectonic brecciation and recrystallization of the Cu-pyrite ore bodies has led to Au and Ag migration to cracks and gold grains coarsening. Cu, Te, Sb and Bi are the most common trace-elements in gold and electrum in Krassen epithermal Cu-Au deposit. The observed trend for increasing of the gold fineness (Figure 5) from the early pyrite to the late anhydrite–barite stage could be due to ore brecciation and recrystallization. Characteristic Bi-Te-Se and Ga-Ge-In trace elements signature in the minerals and ores of Krassen deposit is an important indicator for Au enrichment in high sulphidation environment.

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Резиме

**ЕПИТЕРМАЛНА МИНЕРАЛИЗАЦИЈА НА ЗЛАТО ВО НАОЃАЛИШТЕТО КРАСЕН,
РУДНА ОБЛАСТ ПАНАЃУРИШТЕ, БУГАРИЈА**

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Клучни зборови: епитермално злато; рудна област Панаѓуриште; високо-сулфидно злато, LA-ICP-MS

Минерализацијата на злато во епитермалното наоѓалиште на висока сулфидизација Красен, рудна област Панаѓуриште, Бугарија, е проучувана во поглед на минералниот состав, големината на златните зрна, чистотата и

елементите во траги врз база на анализите на електронска микропроба (EPMA) и индуктивно сврзана плазма-масена спектрометрија со ласерска аблација (LA-ICP-MS). Дел од златото во доминантните примарни масивни пирити е со

субмикроскопска големина ($< 0.1 \mu\text{m}$) и може да му се придружи на таканареченото “невидливо” злато. Подоцнежните напукувања во раните масивни пирити биле подложени на таложење на Си-пиритни рудни тела богати со халкопирит, енаргит, борнит, галенит и сфалерит и проследено со деформации и рекристализација, со што се овозможиле миграцијата на Au и Ag во пукнатините и окрупнувањето на златните зрнца. Чистотата на електрумот во одделните зрна варира помеѓу 882 и 998 ‰. Најчес-

ти елементи во траги одредени во самородното злато, во зрната на електрумот и сулфидните минерали во кои е сместено злато се: Cu, Fe, Hg, Sb, Te, Bi, As и Se. Количеството на Au во пиритот варира од 0.35 до 7.83 ppm, а во халкопиритот од 0.97 до 2.78 ppm. појавата на асоцијациите на елементи во траги Bi-Te-Se и Ga-Ge-In е карактеристична за рудните минерали и е индикатор за присуството на Au.