Arsenian nagyagite from Sacarimb, Romania: a possible new mineral species

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Abstract

Arsenic-rich nagyagite (up to 5.5 wt % As) has been discovered in samples from the Au–Ag–Te deposit of Sacarimb, western Romania. Optical properties of the mineral show small but distinct differences to nagyagite (ss). Chemical analyses indicate a substitution of As for Sb, and chemical zonation suggests rapid changes in the chemistry of the mineralising fluids.

KEYWORDS: nagyagite, arsenic, Sacarimb, Romania.

Introduction

NAGYAGITE is a complex sulpho-telluride of lead, gold and antimony which was named after the type locality of Nagyag (now Sacarimb) Romania (Udubasa *et al.*, 1992). Although the mineral was first described in 1772, and has been the subject of several studies, there is still a large amount of uncertainty about its structure and exact chemical composition.

Several chemical analyses of nagyagite have previously been published, but those presented by Giusca (1937) and Stumpfl (1970) seem to be the most reasonable. Giusca (1937) proposed a chemical formula of $Pb_7Au(Te_3Sb_2)_{\Sigma=5}S_9$, in which the dominant substitution is between Te and Sb, while Stumpfl (1910) suggested $Pb_5Au_{0.66}Sb_{1.06}(Te_{2.33}S_{5.40})_{\Sigma=7.63}$, with the main substitution between Te and S. Until now no significant contents of arsenic have been recorded in nagyagite. Here we present data for a number of nagyagites from Sacarimb which contain significant amounts of As.

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Occurrence

The Sacarimb gold-telluride deposit is located in the southeastern part of the Metaliferi Mountains, western Romania. Although now close to exhaustion, it must rank as one of the most famous Neogene epithermal deposits in the world, not only because of its size (around 85t of Au and Ag so far extracted; Udubasa *et al.*, 1992) but also because it is the type locality for several telluride minerals (petzite, stutzite, krennerite, nagyagite and muthmannite).

The telluride-bearing veins are located in a volcanic body consisting of hornblende- and pyroxene-bearing quartz-andesites of Neogene age. More than 230 mineralised veins have been found, in a relatively small area of around 1 km^2 and to a depth of about 600 m. The number of

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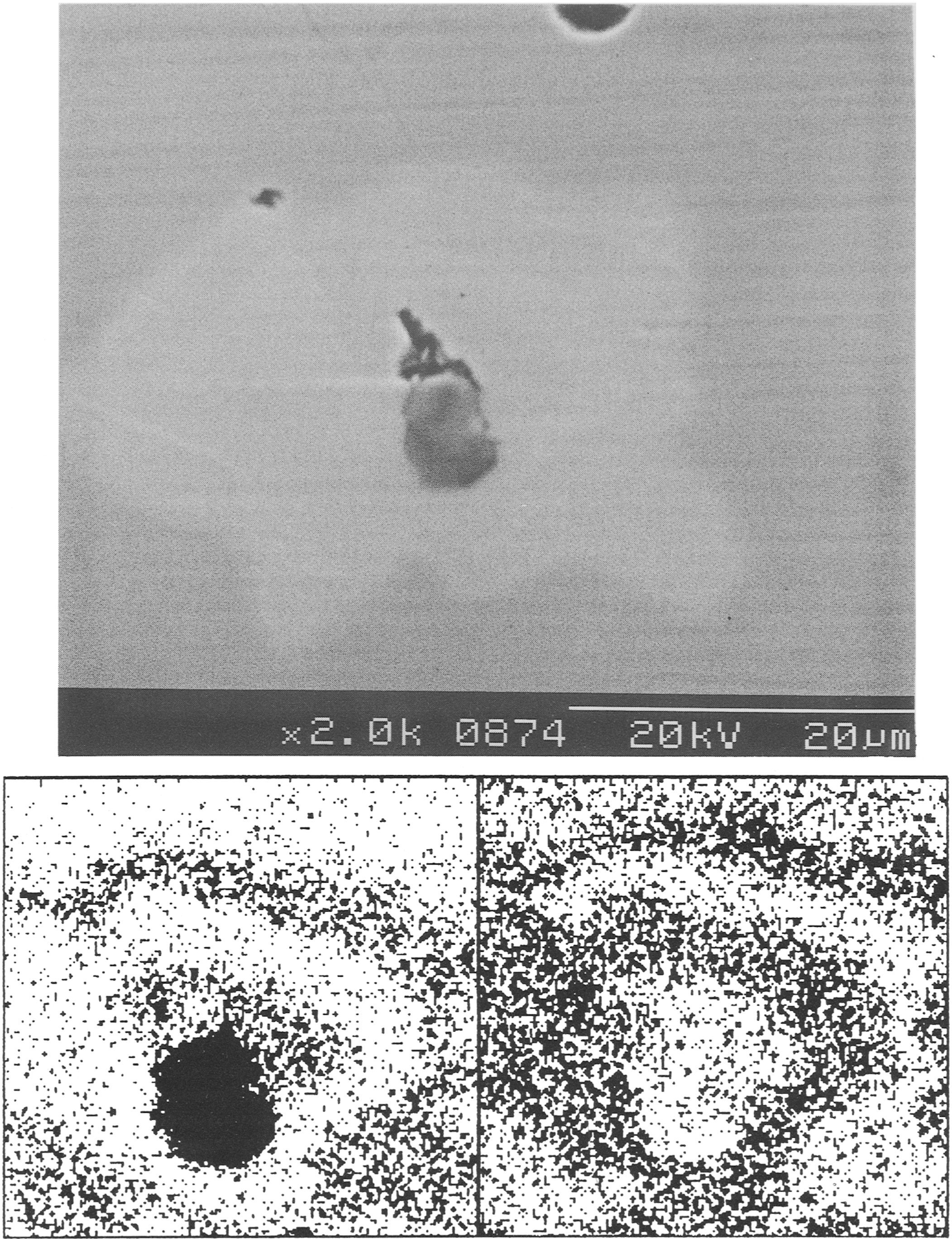

FIG. 1. (*a, top*) Secondary electron image of grain of arsenian magyagite (white) from Sacarimb, showing slight zonation and twinning. The mineral contains an inclusion of arsenopyrite, and is surrounded by petzite (dark grey). (*b, bottom*) X-ray intensity maps for As (left) and Sb (right) for the grain shown in (*a*).

veins decreases markedly with depth. The length of the veins is typically about 500–600 m, with a thickness varying from a few mm to 2 m (average around 0.3 m). The host volcanic rocks exhibit a pervasive propylitic alteration, whilst argillic alteration is dominant adjacent to the veins.

Ghitulescu and Socolescu (1941) summarised the dominant mineral assemblages in the main vein groups of Sacarimb:

- quartz, rhodochrosite, nagyagite and abundant base-metal sulphides, in the Magdalena vein group (SE part of the mine).
- quartz, baryte, rhodochrosite, sylvanite, krennerite, gold and subordinate base metal sulphides, in the Longin vein group (NE part of the mine).
- calcite, petzite and alabandite, in the Nepomuc vein group (SW part of the mine).

The samples containing the arsenian nagyagite were not found *in situ* but came from the collection in the University of Bucharest, and were labelled simply 'Tellurides, Sacaramb'. However, the mineral assemblages suggest that these samples might have come from the Nepomuc vein group. The arsenian nagyagite is associated with altaite, petzite, stutzite, sylvanite I (primary), hessite, sylvanite II (as a decomposition product of the γ and/or χ phase), tellurantimony, nagyagite (ss), coloradoite, and bournonite. Minor amounts of krennerite, arsenopyrite, and boulangerite have also been noticed. The gangue minerals consist of calcite and quartz.

Ore microscopy

Textural features. The arsenian nagyagite usually occurs as rims, 20–50 µm in width, coating the early-formed minerals altaite, petzite, stutzite, sylvanite I, hessite + sylvanite II, tellurantimony and coloradoite, or as apparent inclusions in stutzite. Sometimes it is rimmed by a thin layer of boulangerite and/or bournonite.

Often the mineral shows a clear growth zonation with the normal nagyagite, thus resem-

TABLE 1. Reflectance and colour values for nagyagite and arsenian nagyagite from Sacarimb

Wavelength (nm)	1	2	3	4
400	44.1	45.0	45.5	49.4
420	43.7	44.6	44.9	48.9
440	43.2	44.2	44.4	48.2
460	42.7	43.7	43.8	47.3
470	42.5	43.4	43.6	46.8
480	42.2	43.1	43.3	46.4
500	41.7	42.5	42.8	45.5
520	41.1	41.9	42.3	44.7
540	40.6	41.3	41.8	43.9
546	40.4	41.1	41.7	43.7
560	40.1	40.7	41.4	43.3
580	39.5	40.2	40.9	42.8
589	39.3	39.9	40.7	42.6
600	39.0	39.6	40.4	42.3
620	38.4	39.1	39.9	41.9
640	37.8	38.6	39.5	41.4
660	37.2	38.1	39.0	40.7
680	36.6	37.6	38.5	39.9
700	35.9	37.1	38.0	38.7
Quant. Colour				
x	0.301	0.300	0.302	0.300
y	0.309	0.308	0.309	0.306

1. Arsenian nagyagite (R min)
2. Arsenian nagyagite (R max)
3. Nagyagite (R min on basal section)
4. Nagyagite (R max on basal section).

bling the common zonation seen in tetrahedrite and tennantite (and seen in the Sacarimb veins). Several alternating layers with totally different As and Sb contents may clearly be seen (Fig. 1). Whilst the arsenian nagyagite which occurs as rims around the other minerals is always anhedral, in the zoned crystals the morphology varies from anhedral to euhedral, with a hexagonal form.

Optical properties. In plane polarized light, the arsenian nagyagite is moderately reflective, with very low bireflectance and pleochroism. Colours of R_1 and R_2 are nearly the same: R_1 is a slightly darker greenish grey, whilst R_2 is a slightly lighter greenish grey. When observed near altaite it is grey, near nagyagite it is darker and shows a grey coloration with a slightly greenish tint, and near stutzite it appears slightly lighter and greenish.

Under crossed polars, the mineral shows distinct anisotropism, similar to nagyagite, but slightly stronger, and clearly lower than stutzite. With rotation the tints range from a yellowish brown to purplish grey, resembling those of nagyagite, but lighter. Uncrossing the nicols emphasises the differences between these anisotropy colours: arsenian nagyagite shows brighter colours than those of nagyagite, ranging from bright brownish yellow to bright blue, while the colours for nagyagite range from brown to blue-grey.

Reflectance and quantitative colour. Reflectance measurements were made using a Reichert Zetopan Reflectivity Microscope, and a Zeiss calibrated WTiC standard (ref.no.400). The data (Table 1 and Fig. 2) are consistent with the visual impresson of very low bireflection and pleochroism, and with the observation that its reflectivity appears lower than that of nagyagite. (The nagyagite used for comparison has a similar Au content to that of the arsenian nagyagite (10 wt. %) as we have found that the content of Au has a marked effect on the reflectivity of this mineral.) The colour values for illuminant C are listed in Table 1 for both arsenian nagyagite and nagyagite.

Chemistry

Chemical analyses were obtained by wavelength-dispersive methods, using a Cameca SX50 electron microprobe coupled to a Link AN 10000 energy-dispersive system, installed at the Dept. of Earth Sciences, University of Cambridge. An accelerating voltage of 20kV, a beam current of 60nA, and a proprietary (LINK) implementation of the ZAF matrix correction method were used. The standards used in this study were pure elements for Au, Ag, As, Sb and Te, and PbS for Pb and S. Preliminary SE and BSE images showed that the arsenian nagyagite is quite homogeneous in composition. More than fifty analyses have been obtained on separate grains of the mineral, and on zoned crystals, and these are summarised in Table 2.

These analyses show that the zoned crystals consist of two discrete phases which are different

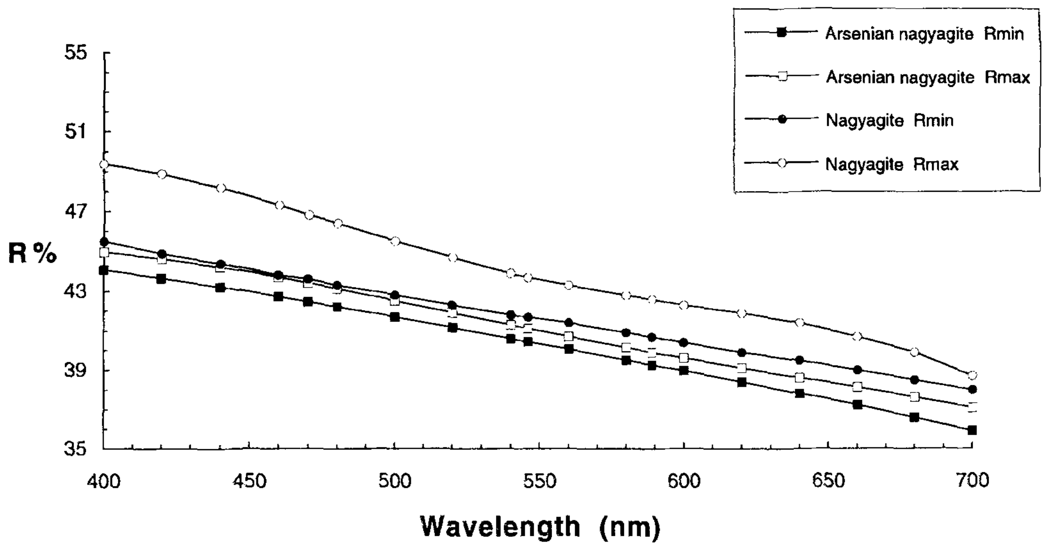

FIG. 2. Reflectivity curves for arsenian nagyagite and nagyagite. Data taken from Table 1.

TABLE 2. Chemical analyses of arsenian nagyagite from Sacarimb

wt.%	1	2	3	4
Pb	54.2	55.0	53.1	55.8
Au	11.5	10.3	10.1	10.5
Ag	0.1	0.1	0.1	<0.1
As	5.5	2.2	4.0	0.1
Sb	0.4	5.8	3.2	7.4
Te	17.5	16.1	18.1	15.0
S	11.5	11.0	11.2	10.9
Total	100.7	100.5	99.7	99.8
No. of atoms				
Pb	5.00	5.00	5.00	5.00
Au	1.12	0.98	1.00	0.99
As	1.40	0.55	1.04	0.04
Sb	0.06	0.90	0.51	1.14
Te	2.62	2.38	2.76	2.18
S	6.85	6.45	6.81	6.28

1. 'High As' arsenian nagyagite
2. 'Low As' arsenian nagyagite
3. Mean of 53 analyses of arsenian nagyagite
4. Mean of 47 analyses of 'normal' nagyagite (with 10% Au)
(Number of atoms in the formula is based on 5 Pb)

not only in optical properties but also in chemical composition (Fig. 1). It is interesting to note that the zoned crystals consist of an alternation of a phase containing Sb, with one containing only As. Nagyagite with intermediate Sb and As contents has mostly been found in the rims. Normal nagyagite from Sacarimb typically contains 0.2% As or less (Table 2).

The empirical formula calculated from the average of 53 analyses of As-bearing nagyagite (no. 3, Table 2), and based on 5 atoms of Pb, is $Pb_{5.0}Au_{1.0}(Sb_{0.5},As_{1.0})_{\Sigma 1.5}Te_{2.8}S_{6.8}$

Discussion

The optical properties, reflectivity, quantitative colour, and chemical analyses suggest that the arsenian nagyagite represents a new mineral species. Unfortunately, the small size of the

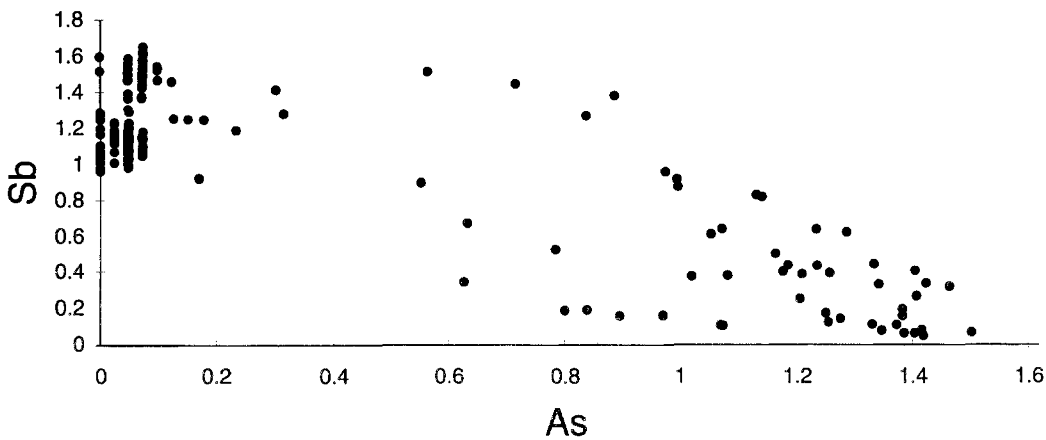

FIG. 3. Plot of As against Sb (number of atoms in formula unit, based on 5 Pb).

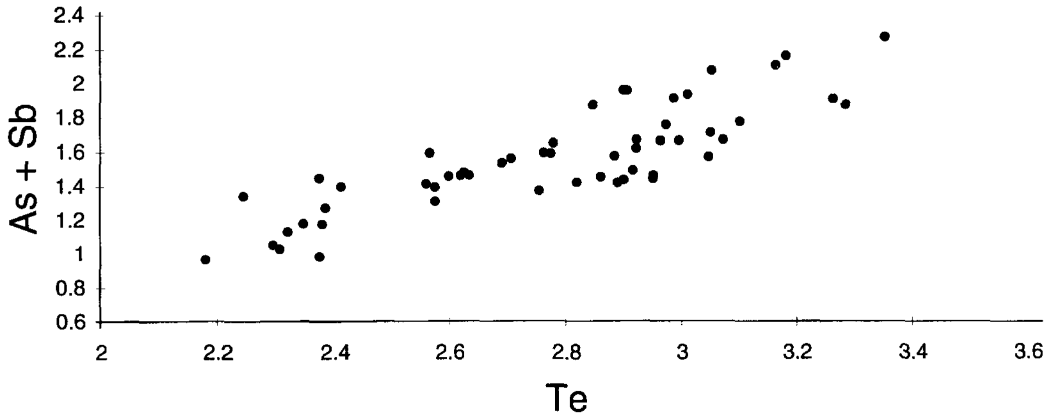

FIG. 4. Plot of (As + Sb) against Te (number of atoms in formula unit, based on 5 Pb).

mineral and its zonation with normal nagyagite make it impossible to carry out any X-ray studies.

The chemical analyses (Table 2) and the plot of As against Sb (Fig. 3) suggest the presence of a solid solution series between nagyagite and arsenian nagyagite, with approximately 1.5 atoms of (As + Sb) for every 5 atoms of Pb. This solid solution is presumably similar to that observed in the tetrahedrite–tennantite series. In addition, Fig. 4 indicates that there is a positive correlation between (Sb + As) and Te, and thus we do not favour the proposal of Giusca (1937) that Sb and Te substitute for each other.

The alternation of the nagyagite and arsenian nagyagite in zoned crystals suggests a rhythmic change in the chemistry of the mineralising fluids, especially in their Sb and As contents. On the other hand, nagyagite and arsenian nagyagite clearly formed after the deposition of the myrmekytic intergrowth of hessite and sylvanite II assemblage, which represents the decomposition product of the γ and/or χ phase (Cabri, 1965; Kelly and Goddard, 1969). If the mineralisation at Sacarimb was deposited under a regime of steadily decreasing temperature, the nagyagite and arsenian nagyagite must have been deposited below 120°C (the decomposition temperatures of the γ and/or χ phase).

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