# NEOGENE MINERALIZATIONS OF BUCIUM RODU-FRASIN Au-Ag DEPOSITS, SOUTH APUSENI MOUNTAINS, ROMANIA

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**Abstract:** The Miocene Bucium Rodu-Frasin deposit is a maar-diatreme complex structure hosting gold-silver epithermal LS mineralizations, in association with quartz-andesite intrusions. There is an intimate spatial and temporal association between the mineralization stages and the Miocene intrusions of the Frasin quartz-andesite. In Rodu area, the mineralizations follow the western diatreme contact with the Cretaceous flysch. In contrast, the Frasin structure mineralizations are focused in or near the NW and NE dome contacts with polymictic breccias. The ore deposition from hydrothermal fluids had a pulsating character, developed in three stages, gold being placed at shallow levels and during the last two epithermal LS stages, from alkaline liquid rich fluids.

The main mineral assemblages are: 1) magnetite (hematite) - pyrite (marcasite) -quartz and pyrite - quartz ± base metal sulfides in the first stage; 2) carbonates (calcite, aragonite, dolomite, ankerite, ± rhodochrosite ± kutnohorite) - quartz - adularia, in the epithermal low sulfidation second stage; 3) quartz - pyrite - marcasite - carbonates (dominant rhodochrosite) - Au and alabandite - rhodochrosite - quartz in the third, epithermal low sulfidation stage.

The mineralizing hydrothermal fluids had near neutral pH and gold was probably transported as a bisulfide complex; first boiling seems to be the main trigger of gold precipitation. Alterations as adularia, phyllic and carbonatizations show close relationships with the gold mineralization.

**Keywords:** Neogene Au-Ag mineralization, veins, stockworks, breccias, South Apuseni Mountains.

## Introduction

The Bucium Rodu-Frasin Neogene volcanic structure and related Au-Ag deposits (Fig. 1) are located in the north-eastern part of South Apuseni Mountains, within the so called "Golden Quadrilateral". They are situated about 10 km east of the Abrud city and 5 km south of the Rosia Montana deposit, being part of the Rosia-Bucium-Baia de Aries metallogenetic district.

The Au-Ag ore deposits have been exploited from old times, traces of those works being provided by large surface excavations. The total resources for the Rodu and Frasin deposits currently stand 43.3M t with 1.3 g/t Au and 3 g/t Ag, equals 1.8 M oz of gold, and 4.7M oz of silver (Verbeek, 2005, in Hewson et al., 2005).

In this brief review, we highlight the basic features of the mineralizations, and the relationships between volcanic structures, hydrothermal alterations and mineralization styles from Bucium Rodu-Frasin area.

## Geological setting

The Apuseni Mountains have been considered as an isolated massif situated inside of the Carpathian arc, in the western part of Romania (Sandulescu, 1984; Balintoni, 1994), or as an interior segment (Seghedi et al., 2004) in the Alpine-Carpathian-Balkan orogenic system. They are composed of two major units: North Apuseni Mountains and South Apuseni Mountains. The South Apuseni Mountains consist of Palaeozoic and older metamorphic rocks, Mesozoic and Neogene igneous and sedimentary rocks.

The Neogene magmatic products (15-7 Ma, Rosu et. al., 1997) have a calc-alkaline character (Ianovici et al., 1976; Cioflica et al., 2002), with some adakite-like features (Seghedi and Downes, 2011), being prevalently andesitic in composition.

In Mesozoic-Tertiary times, the Bucium Rodu-Frasin area underwent a multi-stage evolution characterized by a strong mobility regime explained, at least partially, by the presence of the north-western segment of an important fault, designated here as Vintu-Aries dextral strike-slip fault.

Structural interpretation of detailed mappings, mining and exploration drilling works, magnetotelluric sounding data and laboratory studies (Ianovici et al., 1976; Borcoş and Vlad, 1997; Leary et al., 2004; Nadasan and Hewson, 2005) suggest that Bucium Rodu-Frasin area represents a small Upper Cretaceous-Lower Miocene collapse basin and a complex maar-diatreme structure, resulted from normal reactivation of older, steeply dipping fault structures related to the Vintu-Aries Fault (Berbeleac et al., 2016).

The geology of Bucium Rodu-Frasin area consists mainly of the following lithological units: 1) Lower Cretaceous marine sediments; 2) Badenian Frasin quartz-andesite, volcano - sedimentary deposits, epiclastic deposits and volcaniclastic products (mono- and polymictic breccias); 3) Badenian Frasin hornblende-biotite-quartz andesite intrusions; 4) Barza type Sarmatian(?) andesite lavas, 5) Detunata basalt - andesite; 6) intra - maar - diatreme and subsequent sedimentary deposits and 7) Quaternary alluvial deposits.

Bucium Rodu-Frasin maar-diatreme complex volcanic structure consists of two separate eruptive craters, each excavated 400-500 m into Cretaceous sediments, filled with pyroclastic and epiclastic sequences. One is the Rodu maar-diatreme, crosscut by the Abruzel River, and the other one, the Frasin maar - diatreme structure, crosscut by Seasa Valley (Fig. 1). Both structures cover about 5 $km^2$ and are situated in a high mobility zone with intense deformation related to the Vintu-Aries Fault (Hewson et al., 2005). The Rodu maar-diatreme has an elliptical shape ~ 1500/1000 m with its long axis striking N-S and the southern and eastern sides exhibiting an irregular contour. In contrast, the Frasin maar-diatreme is nearly circular in shape with the diameter of about 1100 m.

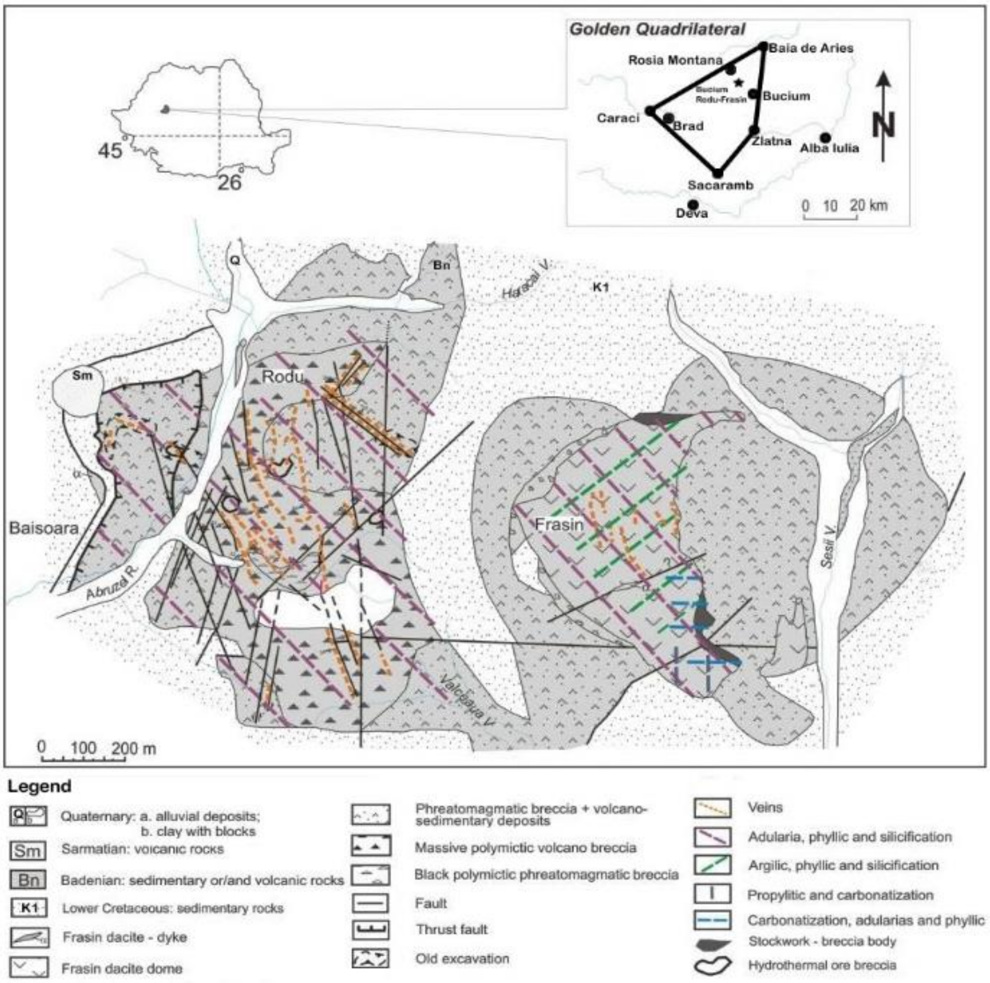


**Fig. 1.** Geological map of Bucium Rodu-Frasin area (after R.M.G.C. data with additions, modified after Berbeleac et al., 2016).

## Hydrothermal alteration

Hydrothermal alteration in Bucium Rodu-Frasin area is pervasive and widespread throughout the volcanic structure and surrounding Cretaceous formations. Altered rocks contain multiple overprinted hydrothermal mineral assemblages, as well as minerals formed by weathering. Five main types of hydrothermal alteration were distinguished (Fig. 2, 3): propylitic, potassic, phyllic, carbonatization and silicic. Argillic alteration is present just locally.

_Propylitic alteration:_ At the surface, this alteration type was recognized in the andesite dike on the right side of Abruzel River and in the south-eastern half of the Frasin quartz-andesite flow dome. Its color is greenish and carbonate, chlorite and minor amounts of albite, sericite, epidote, pyrite and rutile are the most common new mineral assemblages. The Ca-plagioclase is replaced by albite ± chlorite, calcite, and epidote. Pyrite, chlorite, calcite, epidote and rutile replaced hornblende and biotite.

_Potassic (Adularia) alteration:_ This alteration type is quite widespread. Its main characteristic is a total or partial replacement of plagioclase and mafic minerals by adularia, sericite and carbonates ± quartz and pyrite - rutile; the initial rock textures are preserved. The adularia processes seem to have evolved in stages II and III of ore deposition, characterized by following major mineral assemblages (Fig. 2, 3): arsenopyrite (Au?)-base metal sulfides-quartz-sericite-chlorite-adularia and pyrite (Au)-quartz-adularia in stage II and Au-base metal sulfides-carbonates-quartz-adularia with alabandite-rhodochrosite-quartz-pyrite characteristic for stage III. In stage II the feldspar and mafic phenocrystals were substituted by adularia (ad I ± sericite), calcite and pyrite (Fig. 2A-B, F, K), while the III$^{rd}$ stage gave birth to quartz-adularia (- ad II), sericite, carbonates, pyrite ± base metal sulfides, gold and rutile mineral assemblage (Fig. 2D- E, K, F, I).

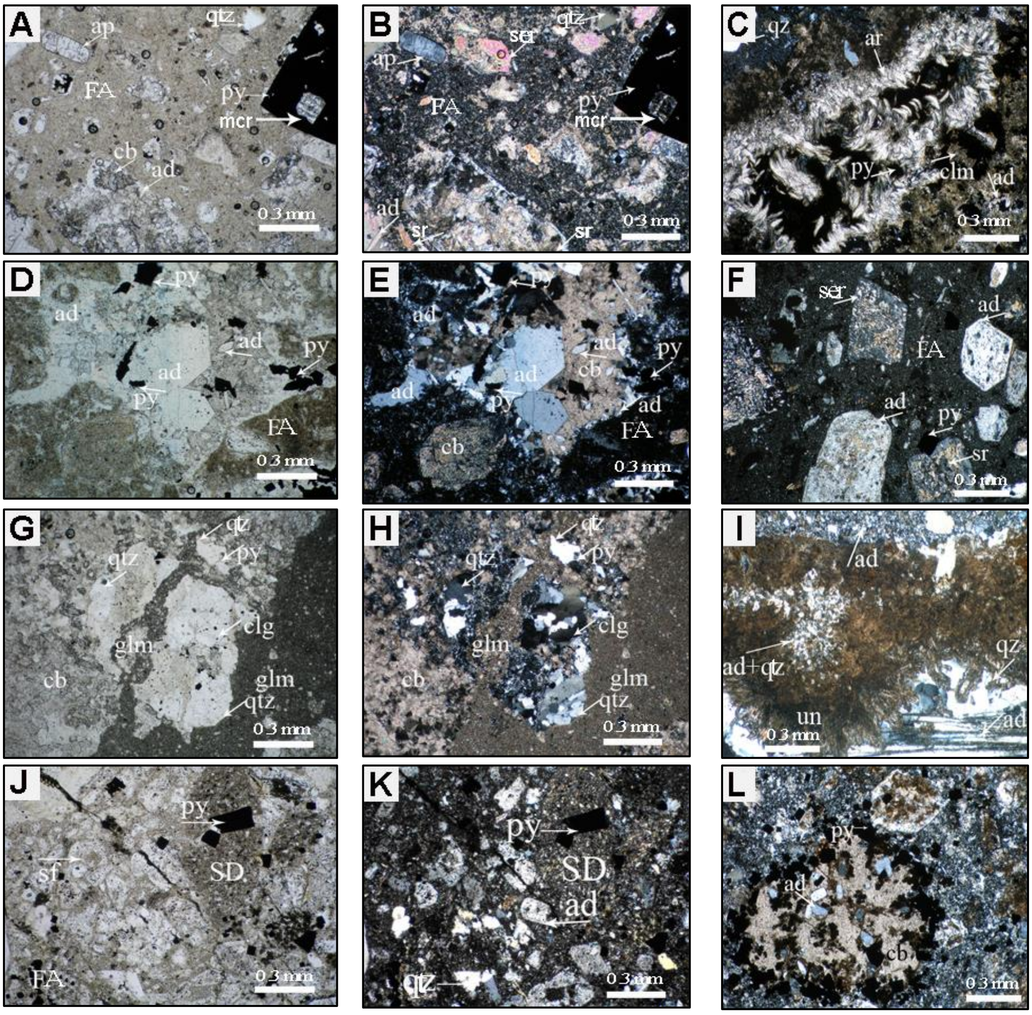


**Fig. 2.** The ore textures and mineralogy of some samples from Bucium Rodu and Frasin gold deposits. A. (N II) and B. (N +). Frasin quartz andesite (FA) with porphyritic and cryptocrystalline groundmass textures. Phenocrysts replaced by adularia (ad), sericite (ser), clay minerals (clm), carbonates (cb) and pyrite (py) (Stage II); C. (N +). Frasin andesite with glassy groundmass and fissure partially filled with aragonite (ar), clay minerals (clm) and pyrite (py) (Stage III); D. (N II) and E. (N +) Vug of jigsaw-fit andesite filled up by quartz (qtz), carbonate (cb), pyrite (py) and adularia II (Stage II); F. (N +). Glassy Frasin andesite (FA) with phenocrysts replaced by adularia (ad), sericite (ser) and pyrite (py) (Stage II); G. (N II) and H. (N+): Polymictic lithoclast breccias with ash clasts. Conglomeratic clast (cgl) consisting of quartzite (qtz), glassy ash clasts and elongate cylindrical glassy vesicles (gvs) filled with quartz, cross-cut by a “glam” (local mining term) fissure (glm). Carbonates (cb), quartz (qtz) and pyrite (py) replace clasts and breccia matrix. I. (N +) Polymictic reworked breccia (Stage II) with a fissure filled with kutnahorite, quartz (qtz) and adularia II (Stage II); J. (NII) Polymictic breccia with interclast relict classical vesicles–sferulites (sf). K. (N+) adularized (ad) vesicles-spherulites and phenocrystals of glassy ash, marginal volcanic sandstone clast (SD) (Stage II); L. (N +). Lithophysae filled with carbonates (cb), adularia (ad) and pyrite (py) in porphyritic quartz andesite clast matrix (Stage II). N II = plane-polarized light; N + = cross-polarized light; all scale bars are 0.3 mm.

<u>*Phyllic alteration*</u> seems to be younger than adularia alteration. Adularia is overprinted, in various amounts, by sericite. The maximum intensity of this alteration type occurs within mineralized zones situated at the eastern contact of Frasin andesite dome with polymictic breccias, as well as in Rodu area. In these areas, there are fine to medium foliated sericite (illite) aggregates, in various percentages, plagioclase and/or adularia remnants (Fig. 2A, B, F).

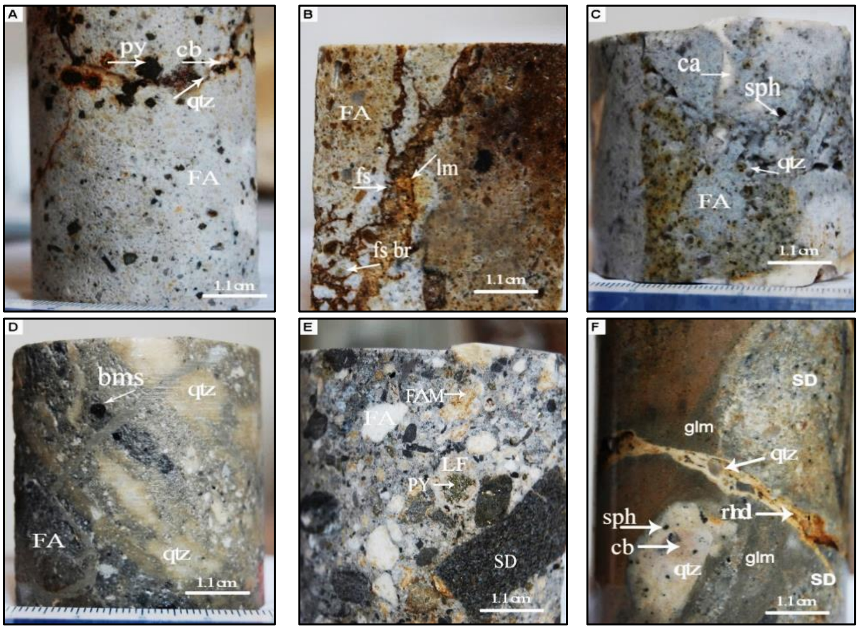


**Fig. 3.** Photographs of some representative core slabs showing the alteration - mineralization stages: A. F1, D 019, m 453.72-473.80. Fissure filled with oxidized pyrite (py), quartz (qtz) and carbonates (cb) in porphyritic Frasin quartz andesite (FA) (stage II); B. D 030, m 94.40-94.45. Fissure (fs) with breccia (br) texture cemented by limonite (lm) in FA (Stage I); C. D 039, m. 107.32-107.37. Stockwork mineralization: sphalerite (sph), quartz (qtz) and carbonates (cb) in vugs of jigsaw-fit texture in fine, moderately porphyritic FA (Stage II); D. D 023, m. 105.85-105.90. A fracture with reworking breccia and bedded texture mineralized with (bms) and quartz (qtz), cut by a (glm) fissure (Stage II + I). E. D 053 A. m. 226-226.05. Massive reworking hydrothermal polymictic breccia with subangular and rounded (FA) clasts altered and mineralized (FAM). Pyrite (py) in lithophysae and angular sandstone (SD) in lithic supported matrix (Stage I?). F. D 005, m. 186.90-186.95. An older" glam" vein and younger vein filled with (qtz), (cb) and (sph) (Stage II), both cut by rhodochrosite (rhd) and (qtz) fissure (Stage III).

*Carbonatization* is subsequent to the adularia and phyllic alterations and in the mineralized zones it is the most widespread. Carbonatization displays a two stages evolution. Partial sometimes up to 60-70% replacement took place in the stage II, feldspar and femic minerals of the quartz-andesite, breccia clasts and Frasin quartz andesite body (Fig. 2A-B) being replaced with fine- to medium-grained carbonates as calcite, ankerite and pyrite. The stage III of alteration is recognized by the presence of litophysae, veins and veinlets with fine- to large-grained carbonates (calcite, dolomite, aragonite, rhodochrosite) and ± quartz, precious metals, base metals sulfides, alabandite, rutile, iron and manganese hydroxides (Fig. 2C-E, G, H; 3C, F).

*Silicic alteration* displays close relationships to the direct pathways of the ore fluids. Its products consist of amorphous silica as dark-grey small fissures and "Chinga" veins and white-brown fine grained quartz crystals ± adularia, sericite, pyrite and argillic minerals (Fig. 3D, F).

*Argillic alteration:* The origin of the argillic alteration products is epigenetic, diagenetic and supergene, the clay minerals outlying, in the mineralized zones, the adularia-sericite and phyllic alteration zones.

White, (2003, in Leary et al., 2004), relying on mineralized zones petrology data, identified two types/phases of alteration: 1. adularia-clay illite-smectite - carbonate Mg-calcite ± kutnahorite ± rhodochrosite ± silica alteration; and 2. later carbonate dolomite ± siderite - kaolinite alteration, due to a low salinity bicarbonate fluid.

## Mineralization

In Bucium Rodu-Frasin area the Au-Ag-base metals mineralization occurs in close genetic relationships with the hydrothermal breccias and phreatomagmatic fracturing. The high-grade mineralization was present mainly in gently dipping veins. Major veins, as in the case of Rodu area, are partly paralleled by preexisting faults. The veins include carbonates, quartz, along with minor pyrite, sphalerite, galena, chalcopyrite, tetrahedrite and gold. Magnetite with minor hematite and sulfides mineralization, probably formed in the mesothermal conditions (?) has been recognized only as metasomatic substitutions of a probable Cretaceous limestone clasts fragmented from the depth.

According to the mineral parageneses, homogenization temperatures and free gold abundance, the mineralization has been considered by White (2003, in Leary et al., 2004) and Szentesy et al., (2004) as LS epithermal style; we think that it probably passes, at larger depths, into the mesothermal domain, or even into skarn in Cretaceous limestone. The ore has been apparently emplaced in subsurface and subaerial conditions. It is situated in the more permeable polymictic breccias at the shallow levels of the Rodu-Frasin maar-diatreme complex structure. In addition, it is present within and immediately adjoining the Frasin quartz-andesite dome, as well as in Cretaceous sediments. The mineralization can be classified, according to the deposition forms, in the following styles: 1) vein which cut the Frasin quartz-andesite dome and volcaniclastic breccias in the Rodu maar-diatreme and Cretaceous sediments; 2) disseminations and hydrothermal breccia in maar-diatreme breccias, quartz-andesite bodies and Cretaceous sediments and 3) stockworks in the Frasin quartz-andesite dome and the contact breccias.

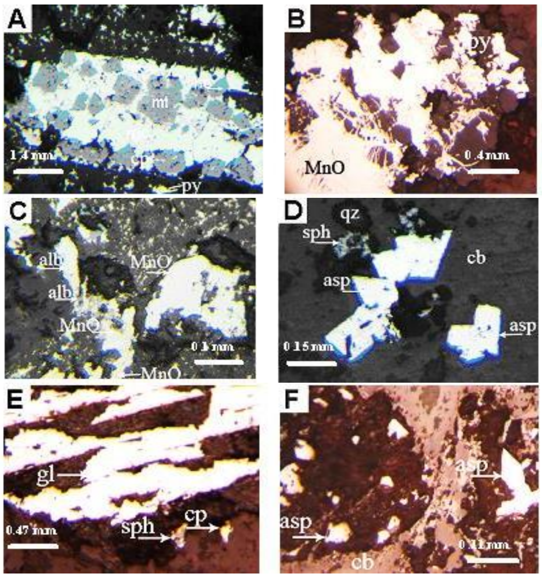


**Fig. 4.** Reflected light photomicrographs of some ore minerals from Rodu and Frasin deposits. A. (N+) Frasin quartz andesite (FA) replaced by euhedral and subhedral coarse-grained magnetite (mt), less substituted by hematite and cemented by marcasite aggregates (mc) and py (stage I); B. (N+) A vug in FA filled with manganese and iron oxides, pyrite and hydrothermal quartz (qtz) (Stage III); C. (N+). Alabandite (alb) near total replaced by manganese oxides (MnO) (Stage III); D. (N +) Fissure filled with arsenopyrite (asp), carbonates (cb), quartz (qtz) and sphalerite (Stage II); E. (N+) Galena (gl), chalcopyrite (cp) and sphalerite (sph) grains, disseminated in polymictic breccia (Stage II). F. (N+) Arsenopyrite (asp) euhedral crystals within lithophysae clast (Stage II).

| Mineral | Zn (Wt%) | As (Wt%) | Fe (Wt%) | S (Wt%) | Au (Wt%) | Ag (Wt%) | Sum (Wt%) |
|---|---|---|---|---|---|---|---|
| **Sphalerite** | 34.20 | 0.00 | 2.30 | 62.12 | 0.80 | 2.30 | 99.42 |
| **Pyrite** | 2.97 | 0.23 | 31.06 | 62.55 | 2.85 | 0.00 | 99.66 |
| **Pyrite** | 2.74 | 0.27 | 29.45 | 63.42 | 3.94 | 0.00 | 99.82 |
| **Arsenopyrite** | 3.55 | 15.84 | 36.19 | 42.59 | 1.10 | 0.00 | 99.26 |
| **Arsenopyrite** | 2.05 | 16.20 | 36.91 | 42.86 | 1.65 | 0.00 | 99.67 |
| **Electrum** | 0.11 | 0.00 | 0.22 | 0.00 | 72.62 | 26.62 | 99.57 |

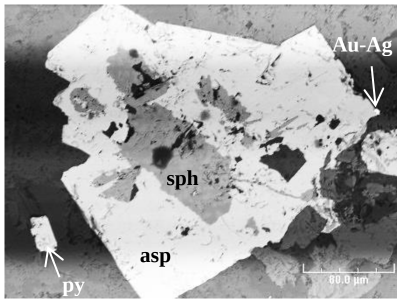


**Fig. 5.** SEM image (right) and SEM-EDS analysis (table, left) of “electrum” (Au-Ag)-arsenopyrite (asp)-sphalerite (sph)-pyrite (py) assemblage.

The ore deposition had a pulsating character (Fig., 3A-D, F) with the evolution occurring, probably, in three stages to which the following mineral assemblages were associated (Fig. 4): a) magnetite (hematite) - pyrite (marcasite) -quartz and pyrite - quartz ± base metal sulfides in the first stage (mesothermal?) (Fig. 10A); b) arsenopyrite - Au -base metal sulfides - quartz - adularia (Fig. 4 D), “Chinga” pyrite - Au - quartz - adularia and base metal sulfides - carbonates (calcite, aragonite, dolomite, ankerite, ± rhodochrosite ± kutnohorite) - quartz - adularia, in the epithermal low sulfidation - second stage (Fig. 4 E) and c) quartz - pyrite - marcasite - carbonates (dominant rhodochrosite) - Au and alabandite - rhodochrosite - quartz in the third, epithermal low sulfidation stage.

_Veins:_ The pyrite-base metal sulfides-gold-carbonates (calcite, dolomite, rhodochrosite, aragonite)-quartz- adularia vein style is especially present in the Rodu maar-diatreme, where it occurs as single veins or/and vein sets, sometimes accompanied by Au - Ag disseminations and hydrothermal breccias. The veins usually have NW-SE strikes and N-S trends, show an “en échelon” tension veins distribution, being related to late normal movement on steeply east-dipping faults (Hewson et al., 2005). The most important veins are: Sperlea, Scaunul Camarii and Scaunele Bradului, Crucile Bradului and Buhaiului” (Ghitulescu and Socolescu, 1941).

_Disseminations and breccias:_ These mineralization styles occur especially on restricted areas of the Rodu shallow-dipping maar-diatreme polymictic breccias and surrounding Cretaceous sediments, and also in shallow-dipping polymictic quartz-andesite breccias situated along the eastern contact of the Frasin andesite dome (Fig. 1). Some parts of the breccias show evidence of multiple events of brecciation, cementation and reworking (Fig. 3E).

*Au-Ag stockwork mineralizations:* The gold is present in various proportions, either as small grains or as sub-microscopic occurrences, within all Rodu-Frasin Au-Ag -base metal sulfide mineralization styles. The individual gold grains in native state have been observed as thin sheets on pyrite, sphalerite and quartz grains or as short wires, and sheets in geodes. It is accompanied by pyrite, sphalerite, calcite, rhodochrosite and quartz. Local gold concentrations are also common at the intersection zones of the so called "chairs" ("scaune") veins with "crosses" ("cruci") veins. According to White (2003, in Larry et al., 2004) and confirmed by our study (Fig. 5) the gold has been geochemically identified as "electrum".

**Conclusions**

The Neogene geologic evolution of the Bucium Rodu-Frasin magmatic-hydrothermal system took place in close relationships with the tectonic, magmatic and metallogenetic activity in the Bucium-Rosia Montana-Baia de Aries district. The ore occurs in a structurally complex environment, typically with some generations of faults or fractures oriented in two or more directions. Ore minerals consist roughly of sulfides, gold, carbonates, adularia and quartz. They have been prevalently emplaced as veins, breccia bodies and disseminations in open fractures and breccias in the Rodu diatreme, and as stockworks, veins and disseminations in relationship to the Frasin dome structure.

**Acknowledgments:** The authors show their gratitude to S.C. Roşia Montană Gold Corporation S.A. especially to Mr. Adrian Minuţ, for the logistic support in the field, and for the access to the primary data. Also, we thank Prof. Dr. Essaid Bilal and Ecole Nationale Supérieure des Mines of Saint-Etienne (France) for the help with SEM-EDS analysis. This study was partially supported by the 29 PCCDI/2018 "GEORES" research project financed by UEFISCDI, Romania.

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