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Spatial and Metallogenic Relationships between Different Hydrothermal Vein Systems in the Southern Arburèse District (SW Sardinia) †

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Abstract: The SW Sardinian basement hosts various ore deposits linked to geological processes active from Cambrian to post-Variscan times. In particular, the Southern Arburèse district hosts several granite-related W-Sn-Mo deposits and a 10 km-long system of Ni-Co-As-Bi-Ag ± Au bearing five-element veins. New investigations into the eastern and central parts of the district (Pira Inferida mine sector) were performed to understand the poorly documented spatial and metallogenic relationships between these systems. The granite-related deposits consist of massive wolframite-quartz (W-Bi-Te-Au) and molybdenite-quartz veins, linked to the early Permian (289 ± 1 Ma) Mt. Linas granite, that are cross-cut by the five-element veins. The wolframite-quartz veins, observed by optical and electron (SEM-EDS) microscopy, show abundant native Bi, Bi-Te phases and native Au suggesting a W-Bi-Te-Au hydrothermal system. The five-element veins exhibit breccia and cockade textures, enveloping clasts of the Ordovician host-rocks and locally small fragments of the earlier W-Mo-quartz veins. The five-element vein paragenesis includes three main stages, from older to younger: (1) native elements (Bi ± Au); (2) Ni-Co arsenides-sulfarsenides in quartz gangue; and (3) Pb-Zn-Cu ± Ag sulfides in siderite gangue. The mineralogical, geochemical and isotopic features of the five-element vein swarm are closely comparable to five-element deposits elsewhere in Europe (Germany, Switzerland, Italian Alps). While the source of Ni and Co is still unknown, the high Bi contents, as well as Au enrichment in the five-element veins, suggest selective remobilization of these elements, and perhaps others, from the granite-related W-Bi-Te-Au veins. The five-element vein system was likely formed during a post-289 ± 1 Ma and post-Variscan metallogenic event.

Keywords: five-element veins; granite-related deposits; Ni-Co arsenides; native Bi; late Variscan metallogenesis

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1. Introduction

In recent years the five element (Ni-Co-As-Bi-Ag) class of hydrothermal vein deposits [1] has been the object of renewed interest, primarily due to its relevant contents in critical elements such as Co and Bi [2]. Past and current mining surveys and exploitation in the major districts worldwide (e.g., Cobalt, Ontario; Great Bear Lake, WT; Bou Azzer, Morocco; Kongsberg, Norway; Erzgebirge, Germany; Jachimov, Czech Republic, Batopilas, Mexico, etc.) revealed the metallogenic intricacies of these systems, commonly characterized by a complex geochemical association (also including Sb, U, Hg and base metals), by exceptional enrichments of Ni-Co arsenides-sulfarsenides and native elements (Ag, Bi) and by a typical carbonate gangue [3]. These peculiar characters raised several questions about the sources of these metals, the chemo-physical conditions regulating their regional-scale transport by low-temperature hydrothermal fluid systems and their deposition in vein-type deposits at shallow crustal levels. In recent studies focused on the genetic aspects of this class of deposits, abrupt redox variations and intake of hydrocarbons in fluids in tectonically active environments have been identified as the main controlling factors [4,5]. This general frame, albeit with differences related to regional and/or local geological conditions, has been confirmed by further studies on European deposits in Germany, Switzerland, Italian Alps and Sardinia [6–8]. The SW Sardinia (Italy) is characterized by multiple metallogenic events that originated different types of ore deposits ranging in age from Cambrian to post-Variscan times [9]. The northern side of this region, the Arburèse district (Figure 1a), hosts several kinds of deposits including: a) skarns, greisens and veins related to the emplacement of the late Variscan (289 ± 1 Ma) Monte Linas granite [10]; b) the large Zn-Pb Montevecchio vein swarm, and a 10-km long five-element vein system formerly considered to be related to the emplacement of the older Arbus pluton at 304 ± 1 Ma [9]. Our study is set in the old Pira Inferida mine, in the central section of the Southern Arburèse district, a key area due to the simultaneous occurrence of large Ni-Co-As-Bi-Ag ± Au five-element veins and a wide granite-related quartz-wolframite vein swarm ("Togoro" veins in Figure 1b,c), both hosted in Ordovician very low-grade metasandstones–metasiltstones. The mine operated during the 1930's and only the Ni-Co arsenide and Pb-Zn sulfide ores have been systematically exploited; the five-element veins of Pira Inferida were included and briefly described in a recent study of the Southern Arburèse district by [8]. Conversely, very little data are available from the poorly studied wolframite-bearing veins. New surveys and mineralogical studies were performed in the area to acquire new data on both types of ores and to investigate their mutual and hitherto poorly constrained spatial and metallogenic relationships.

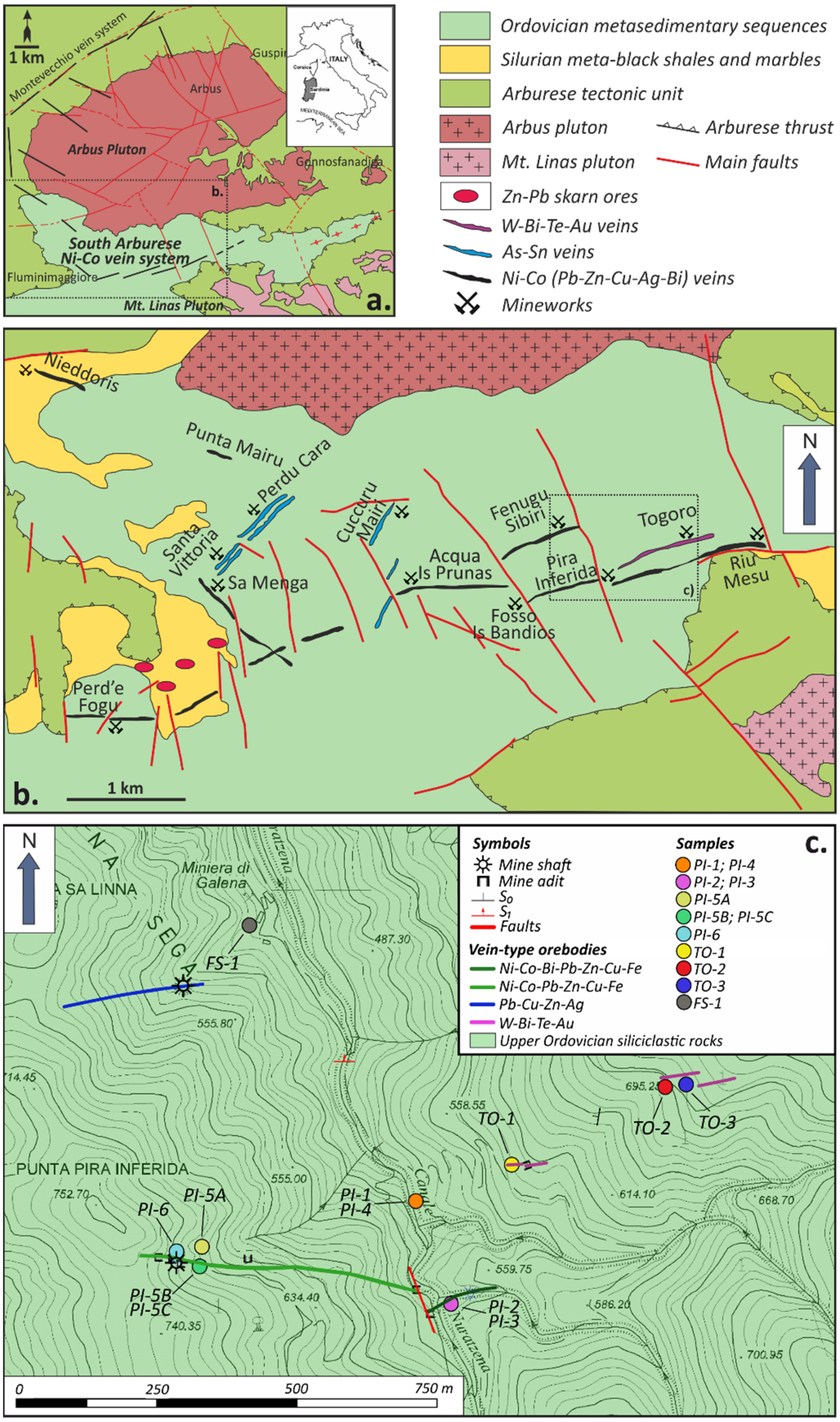

Figure 1. Geological sketch of the South Arburèse area: (**a**) large-scale geological sketch showing the spatial relationships between the Arbus (304 Ma) and Linas plutons (289 Ma) and the orebodies (mod. after Moroni et al., 2019); (**b**) detail of the different vein-type orebodies of the area, including Ni-Co five-element type, As-Sn and W veins (mod. after Moroni et al., 2019); (**c**) small-scale geological sketch of the study area showing the location of the Pira Inferida five-elements type and Togoro W-Bi-Te-Au veins and the sampling sites.

2. Materials and Methods

Field surveys and samplings were performed in the old mine area of Pira Inferida (Figure 1c). Samples were collected at different mine levels, from the outcrops and from the dumps close to the main (mostly collapsed) adits. Several sub-sets of samples (PI-1, PI-2, PI-3, PI-4, PI-5, PI-6) were selected based on their location and different ore and gangue mineral associations. Samples of the quartz-wolframite veins were collected from small mineworks and trenches set at three different levels along the vertical development of the vein swarm (TO-1; TO-2; TO-3). Thin and polished sections from hand-selected samples were studied under transmitted and reflected light (RL) Optical Microscopy (OM). Further investigations were performed by SEM-EDS spot analyses and elemental mapping using a FEI Quanta 200 equipped with a ThermoFisher Ultradry EDS detector at the CeSAR laboratory at Università di Cagliari under high vacuum conditions, acceleration voltage 25–30 Kv, spot size 5 μm.

3. Results

3.1. Field Relationships

The investigated five-element veins comprise a series of 1–3 m thick, E-W striking and S-dipping veins outcropping discontinuously for over 500 m along their strike (Figure 1c). The veins crosscut a thick sequence of very low-grade metasandstones and metasiltites of the late Ordovician age (Rio San Marco Fm. [11]); evidence of contact-metamorphism (i.e., andalusite-bearing spotted schists) are widespread, suggesting a relative proximality of underlying intrusions. A series of mineworks intercepted the orebodies at different levels: in the lower parts, the veins are well exposed with evident banded, brecciated and cockade textures (Figure 2); a thick, well-developed oxidation zone with vuggy quartz can be observed in the uppermost levels (Figure 2a–c). The Togoro wolframite ores occur as a NNE-SSW and N-dipping swarm of sub-parallel quartz veins whose thickness varies from 15 cm to 1 m; the mineralized zone is roughly 50 m wide and can be followed for over 1 km. The veins display a massive texture of white quartz enveloping idiomorphic, millimetric to pluricentimetric wolframite bladed crystals (Figure 2d). Overall, the orientation of the veins suggests crosscutting relationships between the two systems, although the bad outcrop preservation does not allow the direct recognition of their interaction. Such a situation is similar to what is reported in other mineralized sites along the Southern Arburèse district (e.g., Sa Menga and Acqua is Prunas mines [8]), where greisen-type orebodies and five-element vein swarms are contiguous but not well exposed, thereby questioning the succession of mineralizing events.

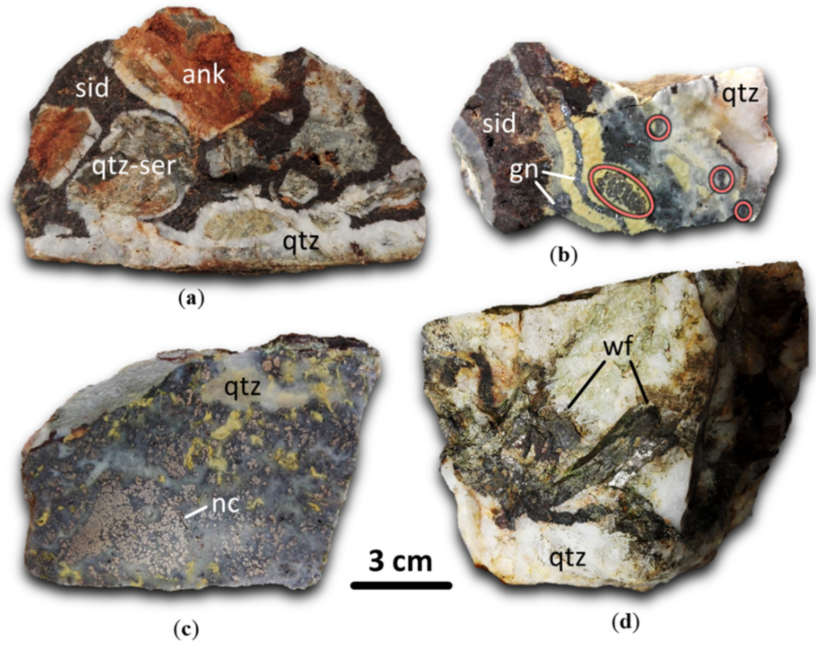

Figure 2. Hand specimens from the five-element veins at Pira Inferida and from the Togoro wolframite-bearing veins: (**a**) typical brecciated textures of the quartz and carbonate (siderite-ankerite) gangue (Pira Inferida, PI-1); (**b**) Bi and Ni-Co arsenide-sulfarsenide nests (in red) and thin galena veins in the siderite-quartz matrix (Pira Inferida, PI-5); (**c**) niccolite aggregates in the quartz matrix (Pira Inferida, PI-2); (**d**) idiomorphic wolframite crystals up to 6 cm in length in quartz gangue (Togoro, TO-2). nc = niccolite; qtz = quartz; sid = siderite; ank = ankerite; gn = galena; wf = wolframite.

3.2. The Five-Element Vein-Type Ore

The mineralogy of the five-element veins of Pira Inferida includes native Bi, Ni-Co arsenides and sulfarsenides, Zn-Cu-Fe-Pb sulfides and quartz-sericite-siderite gangue. Vein textures indicate multiple brecciation events between and during the mineralizing stages. At the selvages, host rocks are diffusely fractured, silicified and sericitized, with fractures filled by thin quartz veinlets. Incipient fracture opening is testified by the abundant host rock fragments into the vein mass. Remarkably, small fragments of older quartz veins were detected into the breccia in many samples; in these fragments, RL OM studies recognized the presence of rutile crystals with high W contents revealed by SEM-EDS. On the basis of the preliminary observations by OM and SEM-EDS, the five-element ore mineralizing stages may be schematized as follows: (1) native element stage (native Bi ± native Au); (2) arsenide-sulfarsenide stage (niccolite ± breithauptite ± löllingite → gersdorffite ± cobaltite ± ullmannite ± bismuthinite ± arsenopyrite) with quartz; (3) sulfide stage (sphalerite ± pyrite → chalcopyrite + tetrahedrite + galena) with siderite-ankerite. Focusing on the first two stages of mineralization, the paragenetic succession of native elements and Ni-Co arsenides-sulfarsenides can be clearly inferred from the zoned textures of the ore nodules, with native Bi cores extensively overgrown by niccolite aggregates with a mosaic or radial texture, in turn rimmed and substituted by gersdorffite and, less frequently, by ullmannite (Figures 3 and 4). Rare, rounded grains of native Au in gersdorffite have been also attributed to these early ore stages [8]. Local breithauptite intergrowths in niccolite and late bismuth-rich infillings along microcracks in the arsenides-sulfarsenides are also observed, with frequent substitution of native Bi by late bismuthinite; löllingite crystals are overgrown by arsenopyrite. Every major ore stage in the five-element veins is marked by strong brecciation and fragmentation of the previously crystallized assemblages, providing the evidence of a remarkably dynamic mineralizing process. Hence, the native Bi-bearing Ni-Co arsenide-sulfarsenide ore occur throughout the veins as irregular fragments and masses enveloped by later base metal sulfides and quartz/carbonate gangue minerals; the Bi-Ni-Co aggregates are more abundant in oreshoot zones that were the main targets of former mining operations.

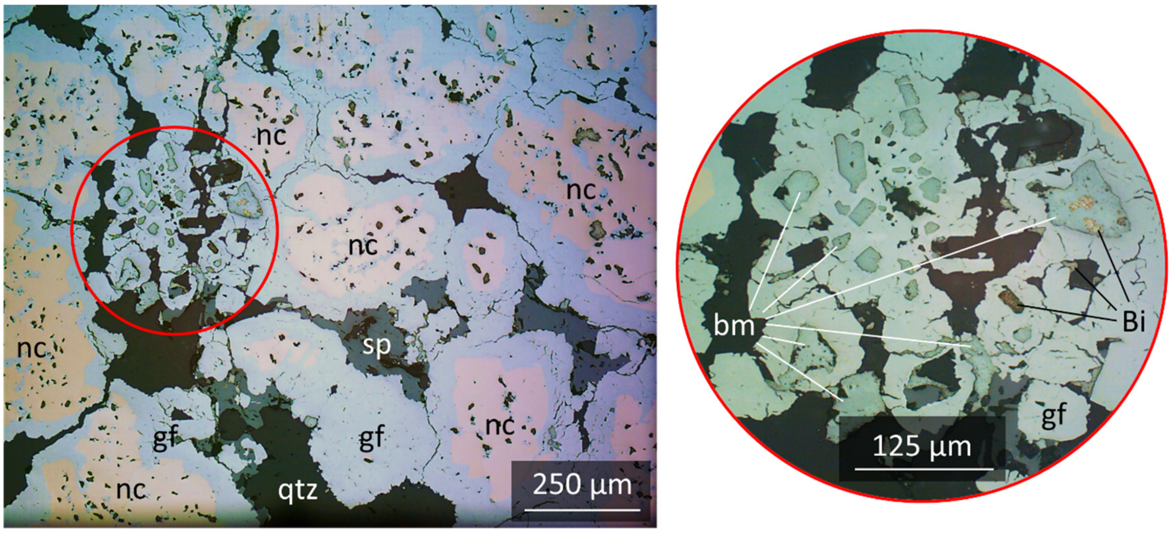

Figure 3. RL microscope images showing the textural relationships between native elements and Ni-Co arsenides-sulfarsenides of the five-element vein of Pira Inferida: native Bi grains with bismuthinite (bis) overgrowths are enclosed in the niccolite and gesdorffite aggregates (PI-2, 10–20×). nc = niccolite; gf = gersdorffite; Bi = bismuth; bm = bismuthinite; sp = sphalerite qtz = quartz.

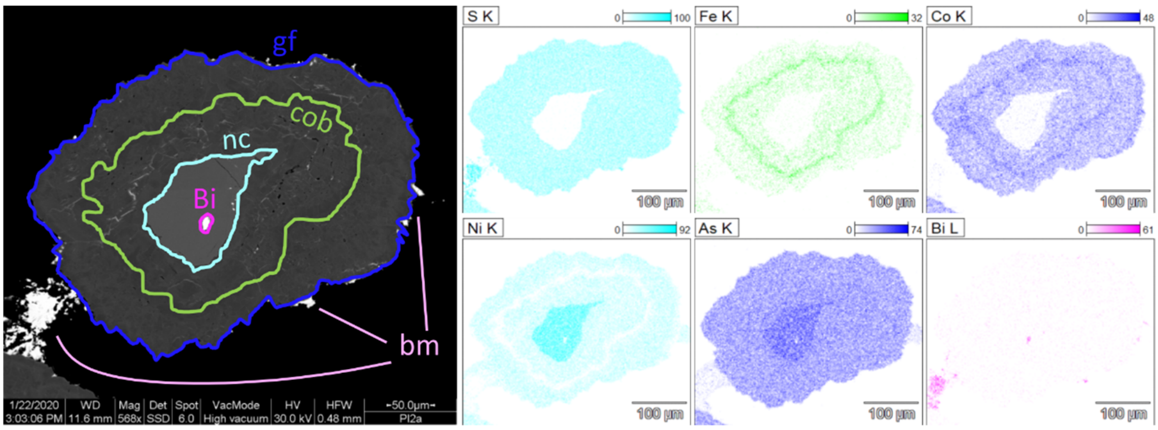

Figure 4. SEM-EDS elemental maps of Ni-Co arsenides-sulfarsenides of Pira Inferida: niccolite with native Bi inclusions surrounded by gersdorffite and cobaltite; fine-grained bismuthinite occurs on the edges of the Ni-Co ore minerals (PI-2). nc = niccolite; gf = gersdorffite; cob = cobaltite; Bi = bismuth; bm = bismuthinite.

3.3. The Wolframite Ore

The mineralogical association of the Togoro wolframite-bearing veins comprises wolframite in a granoblastic quartz and white mica gangue. Radial aggregates of mica are common both interstitial to quartz and along the vein selvages. Wolframite crystals are idiomorphic and tabular-shaped; their dimensions vary from 100 µm up to 6 cm in length. The lack of internal reflections and the weak anisotropy, coupled with the Fe-rich and Mn-low composition provided by SEM-EDS analyses, suggest a ferberite term. Scheelite pseudomorphs with relict wolframite are frequent; thin cross-cutting scheelite veins extend in both wolframite and quartz. Late-stage goethite-hematite alteration is also frequent, occasionally forming perfect pseudomorphs on wolframite and probable arsenopyrite. Numerous zoned Bi phases were observed as microinclusions both in the goethite-hematite pseudomorphs on wolframite and in the quartz gangue. SEM-EDS spot analyses and elemental mapping on Bi-phases in the quartz gangue showed Bi-rich cores with complex associations of bismuth tungstate (probable russellite), up to 500 µm in size, and sulfotellurides, up to 100 µm surrounded by bismite alteration (Figure 5a). Occasionally, native bismuth occurs in the quartz gangue. The presence of native Au/electrum grains, previously observed under OM (Figure 5b), was confirmed by SEM-EDS analyses. Small (10–15 µm) Au+/−Ag blebs occur in close association with native Bi, Bi-W (Figure 5c) and Bi-Te-S phases included in wolframite and in the quartz matrix. Sulfides are rare and are represented only by pyrite and bismuthinite. Accordingly, a paragenetic succession of the W-Bi-Te-Au ore may be proposed with an initial precipitation of native Au, Bi-tellurides and wolframite in a quartz gangue with subordinate white micas, followed by scheelite and Bi-tungstate substituting wolframite and Bi-tellurides, respectively.

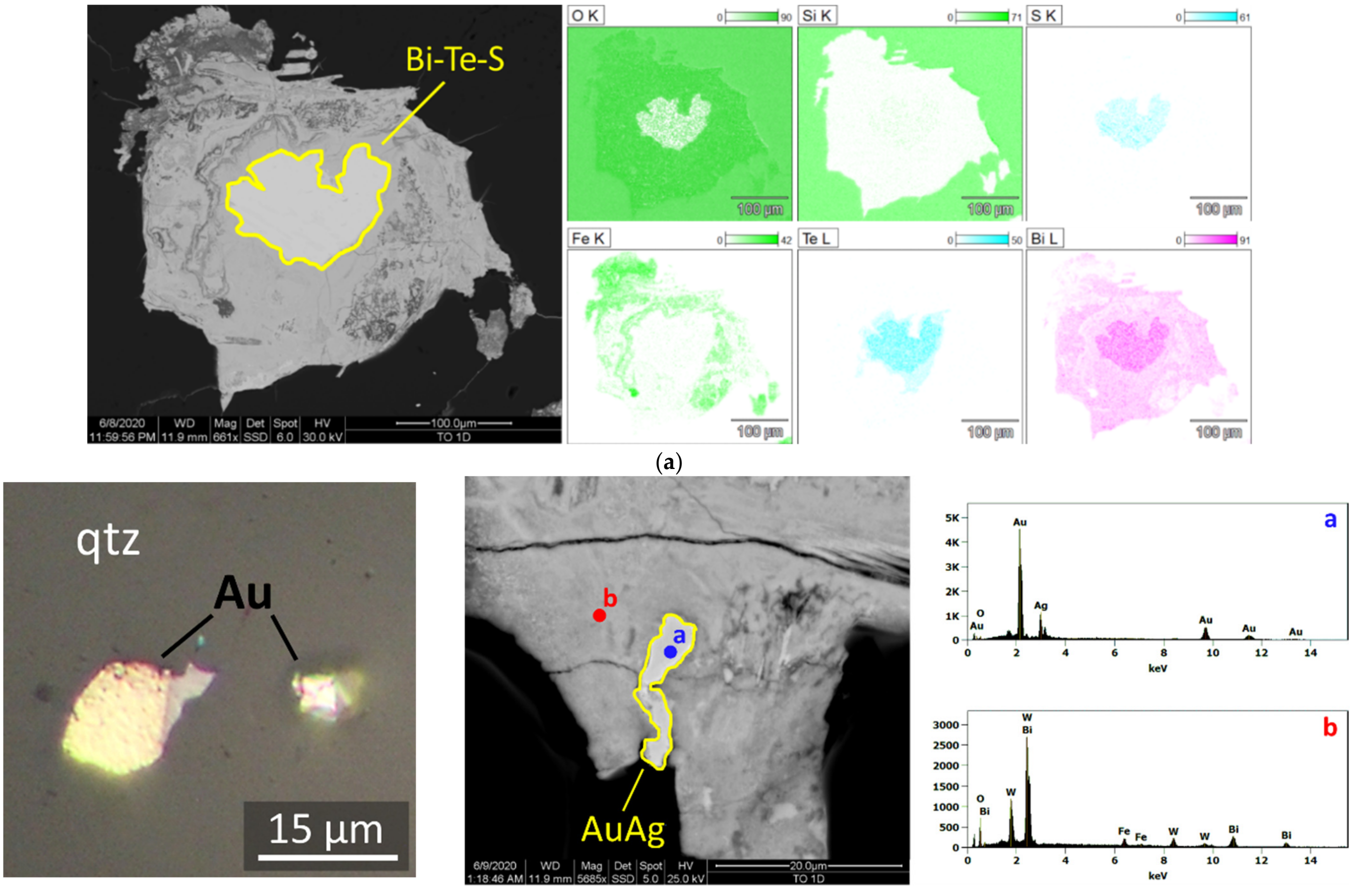

Figure 5. Bi-Te-Au mineralization from the Togoro wolframite veins: (**a**) Bi-sulfotelluride grain surrounded by bismite alteration (TO-1); (**b**) visible µm-scale gold grains enclosed in quartz (TO-1, 50×); (**c**) SEM-EDS spot analysis showing electrum (AuAg) grains, point a, enclosed in a Bi-tungstate, point b (TO-1).

4. Discussion and Conclusions

In this study, new mapping and sampling, OM and SEM-EDS analyses allow us to envisage a relationship between the wolframite-bearing veins and the polymetallic five-element vein system of the Pira Inferida mine sector and to locate these vein systems within a framework of successive mineralizing events that occurred in the Southern Arburèse district during and after the emplacement of the Monte Linas granite. The wolframite-bearing veins are part of the magmatic hydrothermal systems of Monte Linas pluton, which belongs to a suite of early Permian (289 ± 1 Ma) F-bearing, ilmenite-series granites widely represented in SW Sardinia and associated with several Mo-W-Sn skarn, greisen and vein deposits [10]. Although fluid inclusion data on the Togoro vein system are not yet available, cassiterite and quartz microthermometry performed in other veins related to the Monte Linas granite (Perdu Cara in Figure 1b, and Perd'e Pibera deposits) assessed hypothermal to mesothermal conditions (410–320 °C) for their main Sn-(W) and

Mo-(W) ore stages, at a depth corresponding to the shallow emplacement of the pluton (1 kbar) [10]. The detection of previously unrecognized Bi-Te-Au mineralization associated with the granite-related wolframite ore expands the knowledge on the metallogenic endowment of the area. More detailed mineral chemistry studies on Bi-Te phases will evaluate composition and distribution of Bi-tellurides and their relationship with Au enrichment in the vein system [12], the latter being a feature of high economic relevance and worthy of further investigation. Moreover, the presence of abundant Bi phases in the wolframite veins can contribute to defining their metallogenic relationships with the contiguous Ni-Co-As-Bi- Ag ± Au, five-element vein system. The paragenetic succession established in the five-element ore at Pira Inferida approaches the general models [3–5], and broadly matches the depositional sequence in many similar deposits in alpine Italy and in Europe [6–8]. Few data reported for fluid inclusions in quartz [8] document low-temperature (<110 °C) and highly saline (20%NaCleq) fluids, presumably related to late mineralizing stages. The first two stages of mineralization likely represent rapidly changing physico-chemical conditions, causing the fast and abundant precipitation of native Bi "droplets", followed by sudden precipitation of arsenides and, in turn, sulfarsenides, thereby forming zoned aggregates. Diffuse brecciation and cockade textures testify tectonic-driven fluid flow and hydrofracturing of host rocks. The absence of a pre-native element sulfide stage [4] may be ascribed to initial very low S contents in mineralizing fluids [5]; only after the As-dominated stage and precipitation of abundant siderite/ankerite gangue, do sulfides became dominant in the ore. The absence of early siderite, the high Ni/Co ratios and the low Ag contents reflect local peculiarities distinguishing the five-element veins of the Southern Arburèse district from many analogous systems worldwide [3]. Moreover, according to the most recent studies, the interaction between hydrothermal fluids and methane-, graphite- or Fe^{2+}-rich rocks cause abrupt redox changes and triggers metal precipitation [4–6]. Methane has not been detected so far in fluid inclusions from the studied system; however, the late Ordovician metasediments and Silurian carbonaceous black shales, both pyrite-bearing and deposited in oxygen-poor environments [11], may be potential sources of methane and other reducing agents. In the Southern Arburèse district, the polymetallic five-element vein system might have cut across the granite-related veins. At Pira Inferida the crosscutting spatial relationships between the two vein systems are inferred by geometry of the orebodies and are documented by vein breccia fragments enveloped in the gangue of five-element veins. The latter feature is actually reported in other parts of the district, such as the Acqua is Prunas mine area, 4 km W from Pira Inferida, where small fragments of a quartz-molybdenite ore was observed in the five-element vein breccia [8]. These new evidences may have metallogenic implications in tracing the sources of metals of the five-element system. Such sources remain largely hypothetical, particularly for Ni and Co, tentatively ascribed to mafic rocks related to the older Arbus pluton [8]; however, the discovery of a notable Bi and Au mineralization in the wolframite veins suggests that the latter may have had a role as local sources of these metals. Bi and, to some extent, Au, may have been selectively remobilized from wolframite-bearing veins and re-precipitated in the native element stage of the five-element ores. This idea is strongly supported by the high Bi (native Bi + bismuthinite) contents of five-element ores in the highly mineralized intersection zone between the two systems within the mine; Bi contents decrease rapidly moving away from this sector. There is a final issue regarding the timing of the metallogenic events in the district. No geochronological data are so far available for the five-element veins, whereas the age of wolframite-bearing veins may be close to the 289 ± 1 Ma age of the Monte Linas granite. The diffuse presence of cockades in five-element vein textures support the idea of mineralization along a regional-scale fault network during repeated seismic cycles at very low depths (<2 km) [13], indicating that, at the time of the five-element metallogenic event, the Monte Linas pluton and related deposits underwent an exhumation of almost 0.5 kbar. This supports the idea that the five-element vein system may instead belong to a late or possibly post-Variscan metallogenic event similar to the large-scale ones recorded in various mining poles across

Europe [14–17]. Such an event might have been developed at a regional scale and involved hypersaline fluids like those which caused the formation of the giant hydrothermal vein system of the Montevecchio district located just north of the five-element veins [18].

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