

MINERALOGICAL/PETROLOGICAL AND GEOCHEMICAL SURVEY OF HISTORIC MINING SITES IN THE TYROL AND ADJACENT AREAS IN THE COURSE OF THE SONDERFORSCHUNGSBEREICH HIMAT (HISTORY OF MINING ACTIVITIES IN THE TYROL AND ADJACENT AREAS)

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With 4 figures

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Zusammenfassung

Grundlage einer gezielten Auswahl und erfolgreichen archäologischen Bearbeitung historischer Kupferbergbaugebiete in Tirol und seinen Nachbargebieten ist zunächst eine mineralogisch/petrologisch-geochemische und montanarchäologische Prospektion bereits bekannter sowie neuer Fundstellen bergbaulicher Aktivitäten. Diese Prospektion stellt die Grundlage für die archäometallurgischen Untersuchungen dar. Um die in vorgeschichtlichen und historischen Zeiten genutzten Metallsorten regional zuordnen zu können, ist die Erstellung eines Lagerstättenkataloges unerlässlich. Dieser soll neben einer mineralparagenetischen Kennzeichnung (Haupt- und Nebengemengteile, Akzessorien) insbesondere eine genaue geochemische Charakteristik (Spurenelementgehalte, Verteilung der Seltene Erde Elemente, Isotopenverhältnisse u.a.) der Erz- und Nebengesteine sowie der Gangarten in den jeweiligen Lagerstätten beinhalten.

Die mineralogisch-petrologische Charakterisierung beinhaltet 1) die Erfassung der Mineralparagenesen sowie die Texturen in den Proben, 2) die chemische Analyse (Haupt- und Nebenelemente) der Erzphasen, der Gangminerale als auch der Gesamtgesteine und der anfallenden Schlacken und 3) die Quantifikation der *P-T-X* Bedingungen der Erzgenese an ausgewählten Lagerstätten. Ein wesentlicher Bestandteil der geochemischen Charakterisierung ist auch die Messung der Spurenelemente und Pb-Isotopenverhältnisse der Erzphasen, Gangartminerale und Schlacken, damit Aussagen bezüglich der Herkunft von Kupferartefakten getroffen werden können.

Abstract

The basis of a thorough archaeometrical/archaeometallurgical investigation of historical copper-mining sites in Tyrol and adjacent areas is the comprehensive mineralogical/petrological-geochemical and mining archaeological investigation of new all known mineralizations in view of their potential for the exploitation in prehistoric and historic periods. These prospection campaigns provide the basis for the subsequent archaeometallurgical research. In order to perform provenance studies of the metals used and thus trace prehistoric trade routes, it is a prerequisite to obtain mineralogical-petrological and geochemical data of the ore minerals, gangue minerals, host rocks and metallurgical remains such as slags and parts of furnace assemblages, and systematically collect such information for every ore mineralization and mining (smelting) site considered in this project.

The mineralogical-petrological characterization involves 1) determination of mineral assemblages and their textures, 2) determination of their chemical compositions, the major and minor element composition of the ore and gangue minerals as well as the host rocks and the slags and 3) quantification of the *P-T-X* conditions of ore-forming processes of selected ore deposits. In addition, an essential part of the geochemical characterization will focus on the analysis of the trace elements as well as Pb-isotope ratios of the ore minerals and the gangue and the slags in order to obtain geochemical proxies for subsequent provenance studies.

1. Introduction

The introduction of metallurgy into prehistoric Europe led to substantial changes in human culture and the environment. Due to the location of profitable ore deposits in the Eastern Alps, in particular in the provinces of Tyrol and Salzburg, this region experienced marked economic revivals especially during the Bronze Age and the late Middle Ages and early Modern Times. Although archaeological studies from these areas exist (Herdits & Löcker, 2004; Goldenberg, 2004; Goldenberg & Rieser, 2004) there are still many questions concerning the beginnings of metallurgy in the Eastern Alps as well as concerning the spatial and temporal extent of the resulting social, economic and environmental impacts. Therefore it is the objective of the Sonderforschungsbereich (SFB) HiMAT ("History of Mining in the Tyrol and Adjacent Areas"), which was established in 2007, to contribute to the elucidation of these open questions by using a multidisciplinary research approach.

The basis of a thorough archaeometrical/archaeometallurgical investigation of historical mining sites is the comprehensive mineralogical/ore-geological and mining archaeological investigation of all known mineralizations in view of their potential for the exploitation in prehistoric and historic periods (e.g. Niederschlag et al., 2003). These data then provide the basis for the subsequent archaeometallurgical research to perform provenance studies of the metals used and thus trace prehistoric trade routes as has been shown for instance in a comprehensive study for the Erzgebirge (Niederschlag et al., 2003). It is therefore prerequisite to obtain mineralogical, petrological and geochemical data of the ore minerals, gangue minerals, and metallurgical remains such as slags, and systematically collect such information for each ore mineralization and mining (smelting) site considered in this project.

Due to their historical importance for metallurgy and metal trade in the Eastern Alps, most geochemical, mineralogical and mining archaeological investigations focussed on the mining areas of Schwaz-Brixlegg (Schulz 1972a, 1974, 1979, 1986; Gstrein 1979, 1981, 1983, 1984; 1988; Krischker 1990; Arlt 1994; Grundmann & Martinek 1994; Arlt & Diamond 1996, 1998; Goldenberg & Rieser 2004; Höppner et al. 2004; Huijsmans et al. 2004; Martinek & Sydow

2004) and Mitterberg/Salzburg (Herdits & Löckner 2004; Paar 1997, Paar & Mrazek 1994; Stöllner et al. 2004). Contrary to these areas, almost no modern analytical data (EMPA, ICP-MS) exist from other important mining areas in the Tyrol and adjacent areas such as Kitzbühel-Röhrerbühel and Montafon.

In addition, the ore deposits of the Kitzbühel region in North-Tyrol were investigated by members of the Economic Mineralogy group of the Institute of Mineralogy and Petrography at the University of Innsbruck in terms of their ore petrography and ore genesis in the course of the Research Project S21 (Ilkhani 1971; Schulz 1972b, 1979, 1986, 1997; Vavtar 1977; Donati 2003). Subsequently, the data were compiled in the „Austrian Handbook of ores, ore deposits, industrial minerals and energy resources“ (Weber 1997).

The Cu-ore deposits in Montafon/Vorarlberg were investigated by Haditsch & Mostler (1986) and Haditsch et al. (1978) and also lack mineral chemical data so far.

Thus geochemical (EMPA, laser ICP-MS) investigations are, except for Schwaz-Brixlegg and Mitterberg, completely lacking. Therefore, it is the main goal of this project to characterize the Cu-ore deposits of this investigation in the Tyrol (North-Tyrol) and adjacent areas (Montafon/Vorarlberg, Mitterberg/Salzburg) in terms of their mineralogy, petrology and chemical composition of the ore minerals. This will also affect samples (parts of furnace assemblages, slags) from the ongoing mining archaeological prospection campaigns as well, which will also be investigated in terms of their mineralogical and chemical composition.

Although mineral phase relations and qualitative information concerning ore genesis of the specified mining sites has been largely established, quantitative data concerning the *P-T-X* conditions of ore formation as well as the relations between the ore-forming processes and the surrounding metamorphic overprint of the host rocks in these areas are still lacking. This is especially important since all the ore deposits of this investigation occur within the poly-metamorphic basement units of the Eastern- and the monometamorphic basement units of the Southern Alps and thus *P-T* conditions of ore formation could provide clues about the attribution of these ore-forming processes to the metamorphic evolution of the Eastern and Southern Alps.

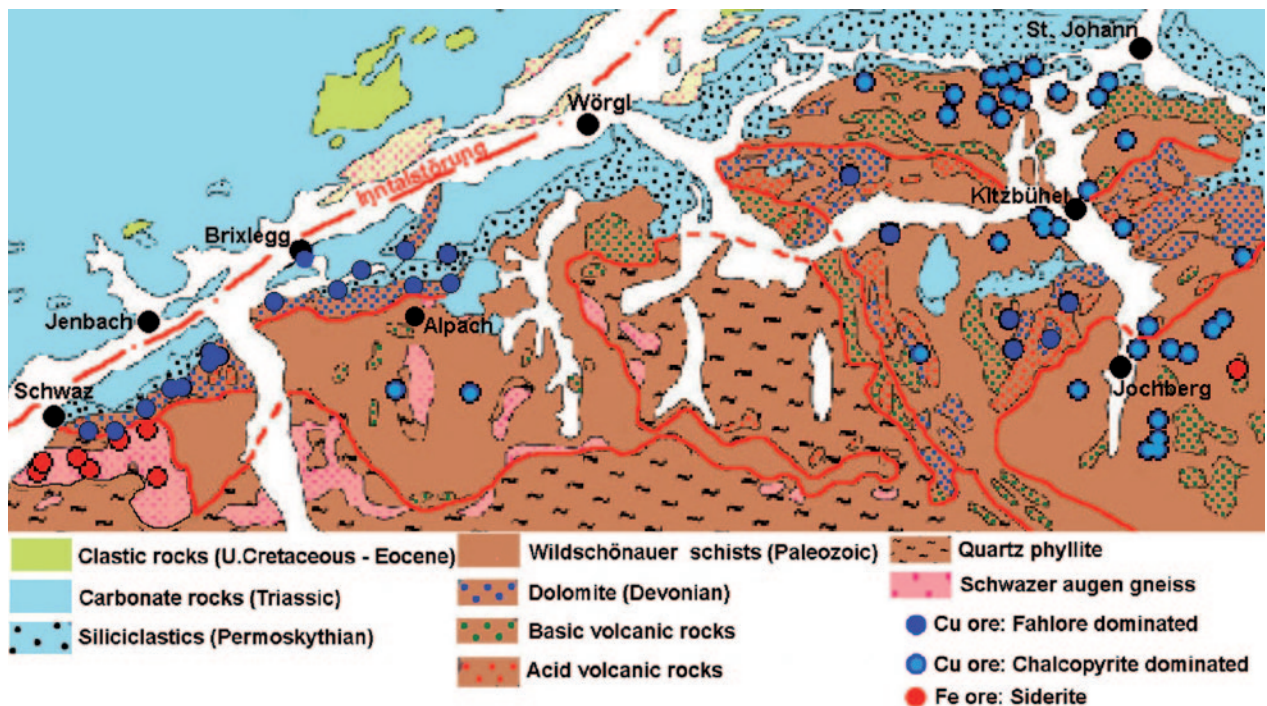


Figure 1: Excerpt from the metallogenetic map of Austria showing the distribution of ore deposits in the western Greywacke Zone between Schwaz and Kitzbühel (modified after Weber, 1997).

2. Goals and methods

Archaeometrical and archaeometallurgical studies of ancient mining sites are based on a large geochemical database which form the basis of provenance studies of artefacts and hence the reconstruction of ancient trade routes. Therefore, the focus of the first phase (2007 – 2011) of this sub-project of the Sonderforschungsbereich (SFB) – Special Research Area, will be on geochemical, mineralogical and petrological investigations concerning selected important copper mining sites in Tyrol and adjacent areas (Salzburg, Vorarlberg) where prehistoric mining activities have been documented so far. For the areas Schwaz, Brixlegg/Radfeld and Mitterberg only minor prospection campaigns are planned to complete the already existing large database on ancient and mediaeval mining activities. For the areas Montafon, Kitzbühel-Röhrebühel, fieldwork (mapping) is planned in addition to sampling. Emphasis will be laid on broadening the geochemical database of this important mining district. In addition to newly obtained ore samples from the filed work, these investigations will also be conducted on i) an already existing large number of ore samples in the mineral collection of the Institute of Mineralogy and Petrography at the University of Innsbruck and ii) on parts of furnace assemblages and slags from the ongoing mining archaeological prospectations.

Mineralogical and geochemical survey

Based on the results of previous comprehensive (mining archaeological, mineralogical, geochemical) investigations of the Bronze Age copper mining sites from Schwaz-Brixlegg-Rattenberg and Jochberg by Goldenberg (2004), similar prospectations are planned for a number of selected copper deposits in Vorarlberg as well as in North-Tyrol. In a first step, it is planned to document and map already existing mining sites, galleries, dumps, slag deposits, smelting installations, etc. in cooperation with the archaeological and geodetic survey project parts, based on already existing maps (metallogenetic map of Austria 1:500.000, map of mining sites and slag deposits of Austria 1:500.000, metallogenetic map of the Italian Eastern Alps, maps of the mining sites). This research will enable to select specific sites suitable for a detailed mining archaeological investigation as shown in Figure 1.

In addition to Schwaz, Brixlegg and Mitterberg, important Cu-(Pb-Zn-Ag-Fe)-mining areas in Vorarlberg and North-Tyrol, were selected for ore petrography and geochemical analyses in close co-operation with archaeometallurgical investigations during the first project phase (4 years):

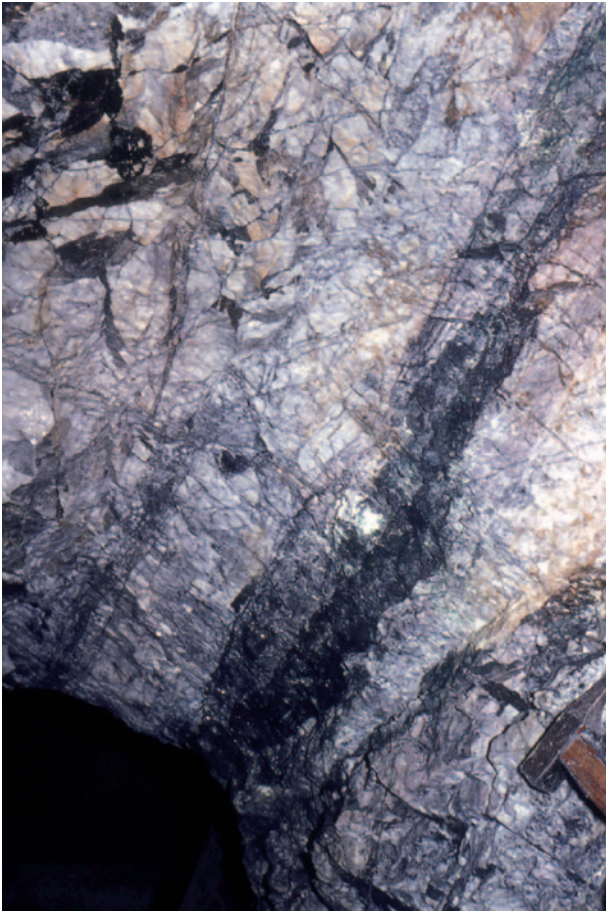


Figure 2: Syngenetic, concordant fahlore within the Lower Devonian "Schwazer Dolomit" from Groß Kogl/Brixlegg.

Schwaz–Brixlegg (North–Tyrol)

Syngenetic, concordant Cu-ore deposits (the main ore mineral is Ag- and partly Hg-containing tetrahedrite $\text{Cu}_{12}\text{Sb}_4\text{S}_{13}$) occur within the Lower Devonian "Schwazer Dolomit" (Fig. 2). During the Variscan and Alpine orogenies remobilisation frequently occurred leading to As-enrichment in tetrahedrite, the formation of discordant ore bodies, and transport into the overlying Mesozoic strata. Gangue minerals are dolomite $\text{CaMg}(\text{CO}_3)_2$, quartz SiO_2 and baryte BaSO_4 (Kleinkogl/ Brixlegg). Typical mining sites are: Burgstall, Falkenstein, Gratlspitz, Hösel-Alpe, Lerchkopf, Mehrerkopf, Pfaffen, Kleinkogl, Roggland, Rotenstein, Schrofen.

Mitterberg (Salzburg)

Mineralizations are also mostly syngenetic, concordant Cu-ore deposits (main ore minerals: chalcop-

pyrite CuFeS_2 , pyrite FeS_2 , \pm tetrahedrite $\text{Cu}_{12}\text{Sb}_4\text{S}_{13}$ partly with Au-content and gersdorffite NiAsS with Co-content) in Lower Palaeozoic metasediments and metavolcanics as well as discordant Upper Cretaceous Cu-ore bodies (main ore minerals: chalcopyrite CuFeS_2 , \pm tetrahedrite $\text{Cu}_{12}\text{Sb}_4\text{S}_{13}$ partly with Au-content, gersdorffite NiAsS and millerite NiS) which occur as remobilised veins crosscutting Palaeozoic metasediments. Gangue minerals are dolomite $\text{CaMg}(\text{CO}_3)_2$, siderite FeCO_3 , ankerite $\text{Ca}(\text{Fe,Mg,Mn})(\text{CO}_3)_2$ and quartz SiO_2 . Typical mining sites are: Buchberg, Filzmoos, Gielach, Igelsbach, Mitterberg, Mitterberg-Südrevier, Mitterberg-Nordrevier, Naglgraben.

Montafon (Vorarlberg)

The ore enrichment occurs 1) as primary Cu ore bodies in Permocarboneous clastic rocks and Lower Permian Alpine Verrucano respectively "the Kristberg Schichten" in close association with rhyodacites and 2) as detrital, secondary siderite-rich Cu ore enrichments in Scythian clastic rocks (Alpine Buntsandstein) (Angerer et al. 1976, Haditsch et al. 1978, Bertle 1979, Haditsch & Mostler 1986). The main ore minerals are: chalcopyrite CuFeS_2 , pyrite FeS_2 , fahlores $\text{Cu}_{12}(\text{Sb,As})_4\text{S}_{13}$, siderite FeCO_3 and ankerite $\text{Ca}(\text{Fe,Mg,Mn})(\text{CO}_3)_2$. The most important mining sites are: Bartholomäberg, Silbertal, Ganeu, Gollmerbach (Rellstal), Kristbergsattel. In addition, Krause et al. (2004) reported a Bronze-age fortification from the Montafon.

Kitzbühel–Röhrerbühel (North–Tyrol)

Abundant Cu-(Ag)-ore bodies occur in Early-Palaeozoic metasediments (main ore minerals: chalcopyrite CuFeS_2 , pyrite FeS_2 , pyrrhotite FeS , \pm fahlores $\text{Cu}_{12}(\text{Sb,As})_4\text{S}_{13}$) as shown in Figure 3 and in Devonian dolomites (ore minerals: fahlores $\text{Cu}_{12}(\text{Sb,As})_4\text{S}_{13}$, \pm chalcopyrite CuFeS_2) within the Northern Greywacke Zone. Well known historical mining sites are: Göttschen (Brixen im Thale), Brunnalm (Kirchberg), Röhrerbühel (Oberndorf), Sinnwell (Kitzbühel), Schattberg (Kitzbühel), Kelchalpe (Jochberg), Kupferplatte (Jochberg). At some of these sites prehistoric mining has been documented (Mutschlechner 1968; Neuninger et al. 1970; Goldenberg 2004).

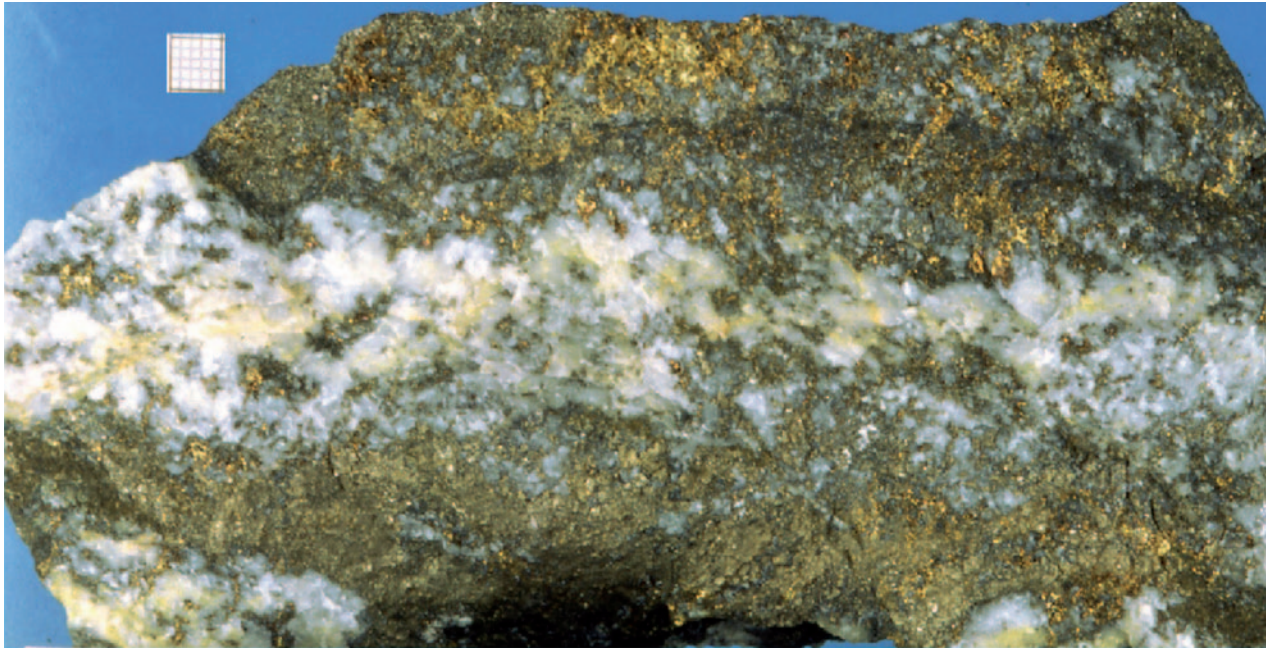


Figure 3: Hand specimen showing the mineral assemblage chalcopyrite + pyrite + quartz + ankerite in a sample from the Kelchalm.

Mineralogical investigations

The mineralogical investigations require the preparation of polished thin sections of ore, gangue, and slag samples for transmitted and reflected light microscopy. The microscopic investigations allow determination of the mineral assemblage and provide textural information about growth stages and mineral reactions. Subsequently, the mineral assemblages will be analyzed with the electron microprobe and/or the micro-Raman spectroscopy at the Institute of Mineralogy and Petrography at the University of Innsbruck to obtain quantitative chemical compositions of minerals on a micron-scale. The mineralogical and geochemical investigations will focus not only on new samples and research targets resulting from the mining archaeological prospection campaigns but also to a large degree on existing sample materials from the large mineral collection of the Institute of Mineralogy and Petrography.

Geochemical investigations

For a complete catalogue of the selected ore deposits in Tyrol and adjacent areas it is not only necessary to investigate the samples of the ore deposits in terms of their mineral assemblage (main, minor, accessory minerals) but to determine their geochemical characteristics and assess their possible exploitation

in early periods (major, minor, and trace elements, lead isotope ratios).

In addition to the geochemical analyses of the ore minerals, the gangue material, which is intergrown with the ore minerals, will also be investigated, since its composition has not only a strong influence on the smelting techniques to be applied but also on the resulting slags. The geochemical investigations will, furthermore, accompany the archaeological field campaigns since geochemical analyses of soil samples, metallurgical objects, slags, parts of furnace assemblages, etc. will help to estimate the extents of metallurgical sites and will also help to interpret remains from various technical processes such as ore beneficiation, roasting of the ore etc.

Whole rock analyses will be conducted with a wavelength-dispersive XRF. XRF analysis of ores require certified reference materials which will have to be purchased as well as special corrections for matrix-interference corrections which will be developed in the course of this project. Trace element analyses (Pernicka 1999) and Pb-, Cu-, Sn- (Begemann et al. 1999, Niederschlag et al. 2003) and Os- and Pb- (Junk & Pernicka 2003) isotope analyses of ore minerals and slags will be conducted with laser-ICP-MS at the Institute of Early History and Archaeology of the Middle Ages, University of Tübingen at the Curt-Engelhol-Center for Archaeometry in Mannheim, which is associated with the University of Tübingen.

Petrological investigations

The petrological investigations will focus on deriving quantitative estimates of P - T - X conditions of the formation of selected ore bodies and the relation between ore-forming processes and metamorphism. Due to the usually small number of suitable thermobarometric phases present in the ore bodies, estimation of P - T - X conditions is a difficult task and one has to carefully investigate the textural relationship between the silicate-bearing host rocks and the ore mineralization, since the silicate-bearing host rocks may allow thermobarometric estimates of the P - T conditions of metamorphism (and hence re-mobilization) in an area. These investigations also require the preparation of polished thin sections of ore, gangue, and slag samples for transmitted and reflected light microscopy. The microscopic investigations of the ore bodies allow determination of phase relations, which then will be analyzed with the electron microprobe analyzer and/or the micro-Raman spectroscopy to obtain quantitative chemical compositions of minerals on a micron-scale. These chemical (electron microprobe) and structural (micro-Raman spectroscopy) data then will provide the basis for calculations of the P - T conditions of the ore-forming processes by using structural changes (T -dependent inversion of chalcopyrite etc., e.g. Mücke 1989) and distribution of ions between minerals within certain phase assemblages (e.g. P -dependent Fe incorporation in sphalerite in assemblage with pyrite and pyrrhotite, Scott 1976), where possible. Overall, there are only extremely few P - T data available from these deposits and no mineralogical-based thermobarometry exists yet. The only quantitative data available are fluid inclusion data, which yielded ca. 200°C and 2 kbar for fahlore formation in Schwaz (Arlt & Diamond, 1996).

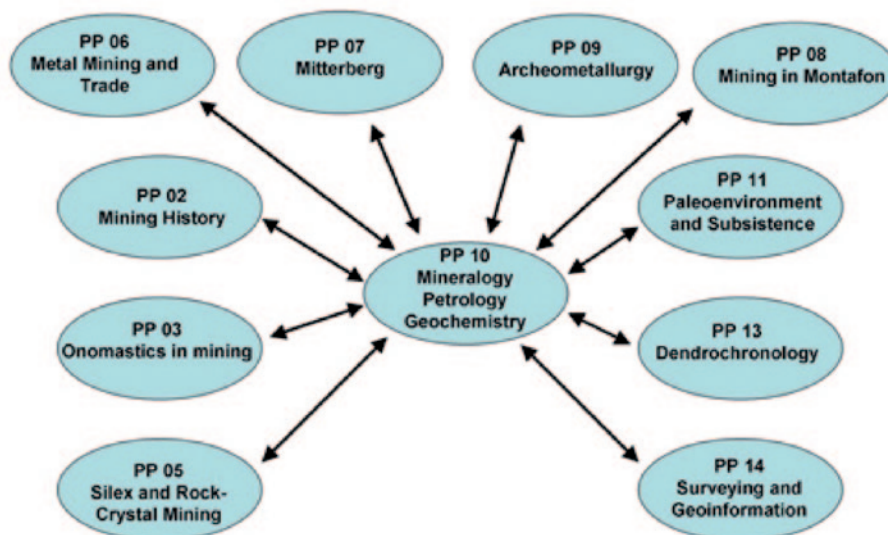
If the textural relation between ore minerals and metamorphic minerals from the host rocks indicate synmetamorphic growth, and it has been shown that Variscan and/or Eo-Alpine orogeny led to re-mobilizations as in Schwaz and Mitterberg. For instance, the P - T conditions from the adjacent silicate-bearing host rocks of the Greywacke Zone can be used as P - T constraints for the re-mobilization. The P - T conditions for the Greywacke Zone, which includes Schwaz/Brixlegg and Mitterberg, were recently calculated by Piber (2005) and Piber & Tropper (2004) to be 320 – 340°C and 4.4 – 4.6 kbar by applying thermodynamic databases and programs such as THERMOCALC

(Holland, 2004 written comm.), and the database of Holland & Powell (1998). Knowing the P - T conditions, one can then put constraints on additional parameters such as fluid composition (X_{CO_2}) and/or fugacity (f_{O_2} , f_{S_2}) by applying appropriate thermodynamic databases and programs, which involve sulfide and oxide minerals (SUPCRT92, Johnson et al. 1992).

Petrological investigations concerning slags and furnace samples will provide information about the firing temperatures and/or oxygen fugacity (f_{O_2}) during the smelting process since abundant newly formed silicate (clinopyroxene, olivine, melilite) and oxide (spinel, magnetite) phases are present (e.g. Hauptmann et al., 1984; Martinek, 1995; Heimann et al., 1998, 2001; Tropper et al., 2004) and the breakdown of silicate precursor phases (e.g. biotite, Tropper et al., 2004) also provides limiting temperature estimates. These T - f_{O_2} estimates will then be obtained by calculating appropriate phase equilibria at 1 bar with the thermodynamic database of Holland and Powell (1998) and the compilation of high-temperature phase diagrams for material scientists and ceramicists (National Institute of Standards and Technology, 2004, written comm.). Similar investigations are very rare from this area and for instance, Martinek (1995) has shown that certain phase assemblages (clinopyroxene + spinel + melilite) from Cu-bearing slags from Buchberg near Wiesing are thermodynamically stable above 1200°C and relatively high f_{O_2} conditions. This important information in turn assists archaeometallurgists and montanarchaeologists when investigating the nature of the smelting processes applied at these mining sites.

3. Position within the SFB HiMAT and internal collaborations

The closest cooperation will be with the archaeological project parts (PP) PP 06, PP 07, PP 08 due to the prospection of prehistoric mining sites, which forms the primary source of archaeometallurgical samples for this investigation, and with project part PP 09 concerning trace element analysis and measurement of Pb-isotope ratios as shown in Figure 4. A close cooperation is also planned with the historical project area PP 02 to identify additional Pb sources from outside Tyrol. Our analytical facilities will also be used by project area PP 11, which deals with palaeo-environmental issues of historical mining, to analyze soil samples from various prospection sites.



Schematic illustration showing the collaborations between the mineralogical/petrological project part (PP10) and the other relevant project parts in the course of the SFB HiMAT.

Co-operations with project areas PP 13 and especially PP 14 during the mining archaeological inspections are also anticipated.

4. Conclusions

The prerequisite of an archaeometrical/archaeometallurgical investigation of historical copper-mining sites in the Eastern Alps is the comprehensive mineralogical/geological and mining archaeological investigation of all well known Cu-mineralizations of the area of Tyrol, Vorarlberg and Salzburg, in view of their potential for the exploitation in prehistoric and historic periods. In order to elucidate the provenance of the metal used and, thus, trace prehistoric trade routes, it is a prerequisite to obtain mineralogical and geochemical data of the ore and gangue minerals as well as of slags, parts of furnace assemblages, and systematically collect such information for every ore mineralization and mining and smelting sites considered in this SFB.

Acknowledgements

The authors wish to thank the Austrian Science Foundation (FWF) for financial support in the course of the SFB HiMAT project part F3110-G02.

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Manuscript submitted: 2.12.2008

Revised manuscript accepted: 7.1.2009