

# THE MINERALOGY AND CRYSTALLIZATION PROCESSES OF THE BULANCAK SULPHIDE VEINS

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**ABSTRACT.** — Ore microscopy, X-ray diffraction and electron microprobe studies and chemical analyses of the vein samples collected from the rocks of the lower basic and dacitic series of the Eastern Pontid Volcanism to the south of Bulancak, Giresun led to the discovery and identification of ore minerals such as betekhtinite and aikinite for the first time in Turkey and idaite and tennantite which were not previously reported from the area.

Ore microscopy studies of the gangue and ore minerals, carried out together with fluid inclusion study, showed lateral temperature zonation and mineralization through oxide-sulphate-sulphide and carbonate stages.

History of mineralization along with general characteristics of the veins affected by temperature and physicochemical conditions are discussed and the optical, structural and textural properties of the ore and gangue minerals are given.

## GENERAL GEOLOGY

The investigated area lies within the Eastern Pontid tectonic unit (Ketin, 1969) which is mainly developed in volcanic facies consisting of calc-alkaline and tholeiitic rocks. These rocks are grouped into six distinct series; lower basic series and intermediate intrusions representing the early phases of volcanic activity in the Eastern Black Sea region. The overlying series are dacitic that are followed by upper basic series and acid intrusions, Tertiary granitoids and finally young basic series with late dykes (Schultze-Westrum, 1961; Tugal, 1969; Akinci, 1980). The lowermost two series and Tertiary intrusives are largely represented to the south and southwest of Bulancak where following mappable units were recognized (Fig. 1).

### **Basalt dykes**

### **Quartz-microdiorite**

<b>Dacitic series</b>	{	<b>Agglomerate, sandstone, limestone</b>
		<b>Biotite-andesite</b>
		<b>Rhyodacitic lavas, dykes and pyroclastics</b>
		<b>Tuffaceous breccia and sandstone</b>
		<b>Porphyritic dacite lavas and dykes</b>
		<b>Pelitic limestone and tuff</b>
<b>Lower basic series</b>	{	<b>Andesite, tuff, ignimbrite and breccia</b>
		<b>Basalt, andesite and spilite</b>

The sulphide occurrences, starting almost from the Black Sea Coast between Bulancak and Giresun, where dacitic series are exposed to the south, are confined to an area of approximately 60 km<sup>2</sup> between Boztekke village in the north and Küçükdere village in the south.

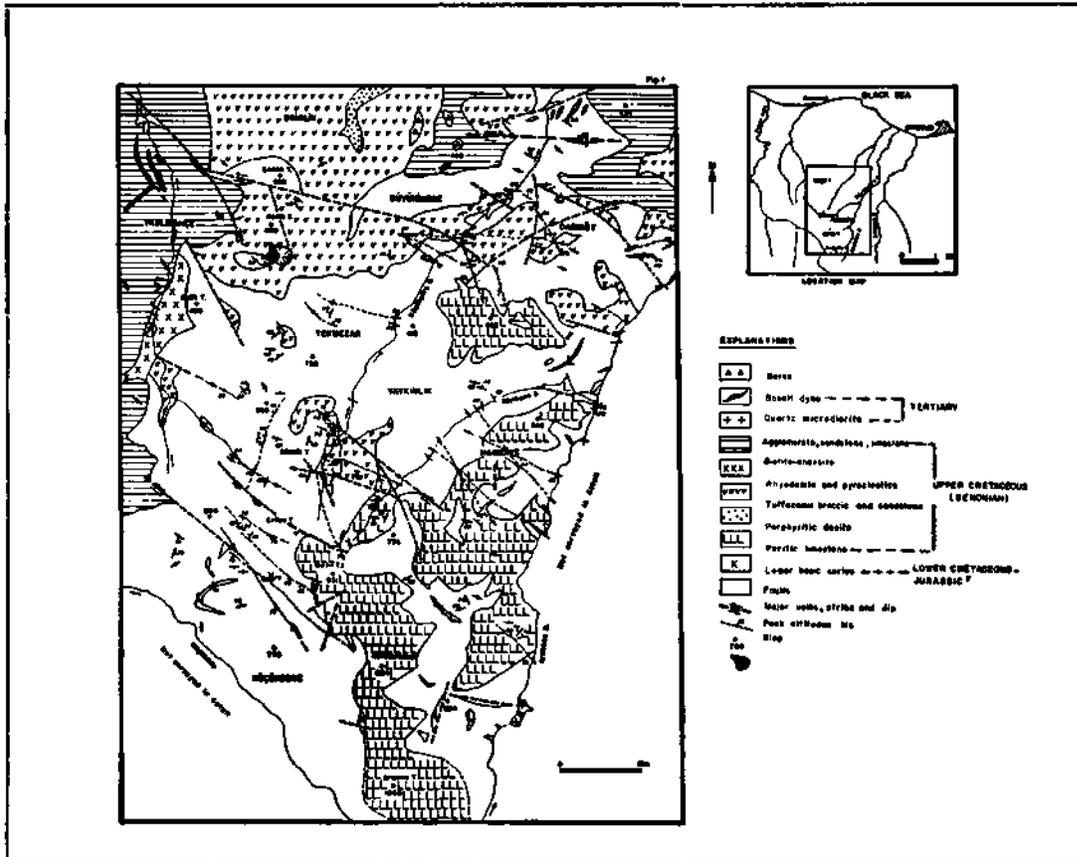


Fig. 1 - Geological map of the vicinity of Darıköy-Tekmezar sulphide veins, SW Giresun, Turkey.

The characteristics and pattern of the veins studied in this area can be outlined as follows:

1. Veins are found in an approximately 3 km wide, 7 to 8 km long mineralization zone-in a broad sense-along an axis marked by Sarıdiken dere- Küçük dere water courses. Along this NE - SW trending zone the dominant direction of the approximately parallel veins is NW - SE (Fig. 1).
2. The mineralization along the entire length of the veins are irregular in terms of attitude, thickness and mineral association. Lens shaped pockets of pinching and swelling character are linked to each other with fine veinlets or barren fractures. Brecciation along the vein zone is the most common feature.
3. Veins exposed in the areas where lower basic series are largely covered with porphyritic dacite formations are chalcopyrite rich. Outside this zone the veins are polymetallic.
4. Fluid inclusion studies have shown a lateral zoning as well as vertical. Highest homogenization temperatures (350°C) and salinities (13.4 wt. % NaCl) were obtained in the southwestern end of the mineralization zone (Akinci, 1976a) as described above.

### PREVIOUS WORK AND INVESTIGATION METHODS

Individual occurrences have long been known and according to Ayışkan (1959) Körüktepe vein was mined along nine adits during the 1870-1880 Russian War. He has given a list of about 50 vein occurrences and the minerals determined in polished specimens around Tekmezar area. Hematite, pyrite, marcasite, sphalerite, chalcopyrite, galena, bornite, tetrahedrite, covellite, chalcocite, malachite, azurite, cerussite, anglesite, ankerite, dolomite, calcite, baryte and quartz are reported from these occurrences. In addition to these, minerals the writer was able to identify and confirm the existence of betekhtinite (Akinci, 1976 *b*) and bismuthinite-aikinite solid solution minerals (Akinci, 1979). Idaite, tennantite and digenite were determined by X-ray diffraction and electron microprobe analysis. The identified minerals are listed in Table 1 below:

**Table 1 - Minerals identified in the Bulancak sulphide veins**

<i>Oxide</i>	<i>Sulphate</i>	<i>Carbonate</i>	<i>Sulphide</i>	<i>Sulphosalt</i>
Hematite	Baryte	Cerussite	Pyrite	Tetrahedrite
Ilmenite	Anglesite	Calcite	Marcasite	Tennantite
Quartz		Dolomite	Sphalerite	Aikinite
Goethite		Ankerite	Galena	
Lepidocrocite		Siderite	Chalcopyrite	
		Malachite	Idaite	
		Azurite	Covellite	
			Digenite	
			Betekhtinite	

The samples studied came from approximately 145 individual sulphide vein exposures within Darköy-Tekmezar area and neighbouring villages. Mineral concentrations were obtained by hand picking or digging by a hand-drill from polished sections for microprobe analysis and X-ray diffraction studies which were mostly employed for the identification of carbonate gangue and barytes as well as for betekhtinite, aikinite and especially for cell-size determinations of sphalerites. Several different minerals were also etched to see the orientation of chalcopyrite exsolution blebs and inclusions in sphalerites and to bring out cleavage, twinning and zoning of sulphides and sulphosalts.

### MINERALOGY OF THE VEIN MINERALS

#### Quartz

It is the principal gangue mineral and present in almost every specimen studied. Skeletal forms, due to replacing sphalerite (Photo 1) and galena and well-preserved idiomorphic crystals are abundant. In general, it is replaced by late stage sulphides, especially by fahlerz.

#### Hematite

In several specimens it has been identified in insignificant amounts in a quartz matrix. At Camideresi locality, at the southern end of the study area a limestone lens at the lower basic and dacitic series contact is almost completely replaced by hematite but the occurrence was not found to be economically viable.

It is found as a bundle of interfingering blades or lath shaped crystals (Photo 2) in different orientations showing strong anisotropy. Crystals were observed altered to and veined by goethite. Pseudomorphs of goethite preserving hematite crystal outlines were also observed.

### **Ilmenite**

This mineral was recognized only as exsolution lamellae in hematite and in accessory magnetites of the basaltic rocks. Its distinct reflection pleochroism and pronounced anisotropy is characteristic.

### **Baryte**

Crystals separated from hand specimens were X-rayed for confirmation. A few veins were found baryte rich along the eastern contact of lower basic and dacitic series (Fig. 1).

### **Anglesite**

Despite the widespread alteration of galena in the studied area, anglesite is less common than cerussite and develops along cleavage planes and grain boundaries of galena together with cerussite and covellite. It is found veining and replacing sphalerite and fahlerz grains enclosed in sphalerite and infiltrates along quartz grain boundaries.

### **Goethite-lepidocrocite**

These alteration products were found associated mainly with altered pyrite and chalcopyrite and to a lesser extent with marcasite, hematite and sphalerite. Cellular-filliform or colloidal banding and rhythmic encrustations around cataclastic or idiomorphic pyrite crystals are the most common textures (Photo 3). A mottled covellite-goethite intergrowth develops after alteration of chalcopyrite while pyrite always gives colloidal goethite bands and less commonly granular lepidocrocite aggregates.

### **Cerussite**

It is found extensively as a weathering product of galena. The cleavage of galena is often preserved in early stages of development but in later stages only relicts or kernels of galena can be seen (Photo 6). Strong bireflection is easily observed in large granular areas but anisotropy usually was obscured due to its transparency.

### **Malachite and azurite**

They are found in fibrous radiating aggregates after chalcopyrite alteration and are not very common.

### **Calcite, dolomite, ankerite and siderite**

Although dolomite is found as the common carbonate gangue in large amounts, calcite and siderite were recognized in single specimens and were confirmed by X-ray diffraction. In some specimens coarse granular calcite was found replaced by bornite grains which cut across a twin lamellae of it.

Siderite is seen as very well-preserved, zoned idiomorphic crystals (Photo- 5) and X-rayed for confirmation. Development of goethite along these-crystals was also observed and in places they are seen as if being replaced by sphalerite and late quartz cutting across the crystal outlines.

Ankerite has not been identified in the polished specimens but X-ray diffraction of a reddish brown carbonate separated from hand specimens has given ferroan dolomite.

### **Pyrite**

By and large this is the most common and earliest formed sulphide mineral. Three forms of pyrite can be identified in the mineralized area:

1. As a result of pyritization of country rock. This tends to form cubic crystals as small as 50 microns or less and in some cases are seen embedded across feldspar crystal outlines.
2. As late stage stringers and crystallites smaller than 100 microns developed along fractures in sphalerites.
3. As early formed, 600 by 700 microns sized aggregates of interlocked grains as well as with individual cubic, octahedral and pentagonal crystal faces.

Zoning is quite common and is usually brought out by gangue minerals or minute chalcopyrite inclusions and limonite alterations. Good examples of cataclasis with matching walls were seen. These fractures and grain boundaries of aggregates are usually filled and replaced by chalcopyrite (Photo 4) which in turn may be replaced by galena, digenite and fahlerz selectively. Poikilitic, sieve-like grains or porphyroblasts may form due to quartz and chalcopyrite replacement. No form of colloidal pyrite was seen. Cell-sizes of six pyrites vary within the range of 5.4172 Å to 5.4182 Å.

### **Marcasite**

It is found only several grains enclosed in chalcopyrite as bladed, pseudo-cubic and lath shaped, 50 by 250 microns size, strongly anisotropic crystals showing indications of transformation after pyrite. In most of the aikinite bearing specimens marcasite was found associated and close to these sulphosalt areas.

### **Sphalerite**

It is the dominant ore mineral of the polymetallic veins and is found in subhedral to xenomorphic grains or corroded, replaced forms with a coarse granular texture. Its grain size averages about 300 to 500 microns. Cleavage and lamellar twinning are common. Displacements were observed across the twin lamellae. Brecciation is common and brecciated grains are seen healed by late stage quartz. Almost all of the sphalerites studied in polished sections have ovoid chalcopyrite exsolution blebs, emulsion type inclusions, exsolution lamellae. Stringers and segregation veins are common, and several specimens show coarse unmixing (Photo 7). Coarse chalcopyrite inclusions are selectively replaced either by galena or fahlerz. It is extensively replaced by galena and contains abundant idiomorphic or skeletal quartz crystals giving rise to atoll and ring textures. Quite often it is seen veined by chalcopyrite, galena, fahlerz, limonite, cerussite or dolomite, covellite and quartz. Deposition overlapping with galena was seen in some specimens.

The structural control of manganese, cadmium and iron bearing sphalerites and the effect of iron content on the colour and cell-size of sphalerites were discussed by the writer previously (Akinci, 1975).

### Galena

It is found in allotriomorphic, poikiloblastic forms in spaces of quartz matrix or replacement forms interstitial to sphalerite. It usually replaces cataclastic pyrite, chalcopyrite and quartz and is also seen as ovoid inclusions in chalcopyrite, ring or atoll textures form when quartz crystals enclosed in galena are replaced centrally. It selectively replaces pyrite crystals enclosed in chalcopyrite. Tiny patches of fahlerz are also common. In places bornite and betekhtinite were found replacing allotriomorphic galena along cleavage planes and grain boundaries. Myrmekitic intergrowths with digenite and bornite are seen.

### Chalcopyrite

In pyrite rich veins it forms the major constituent after pyrite. Narrow, massive chalcopyrite veins were also seen. It replaces early formed pyrites extensively (Photo 4) lamellar and granular nature were brought out by etching. It is selectively replaced by galena and fahlerz.

Paragenetically most of the chalcopyrite grains appear to be formed later than sphalerite but some embayments and large grains of chalcopyrite are found intergrown with sphalerite in mutual boundary textures which may indicate that they are contemporary.

### Bornite

It is found only in a few specimens as a major constituent although in several specimens it was seen replacing chalcopyrite inclusions in sphalerite and pyrite. It shows excellent mosaic texture. It replaces galena grains centrally and is extensively replaced by chalcopyrite which shows rim replacement with exsolution flames in places along the margins of bornite grains or is seen exsolving from bornite with spindle like lamellae along the crystallographic planes of bornite.

Bornite was also found forming rings around quartz crystals enclosed in betekhtinite rimming, veining and replacing betekhtinite, producing rim or atoll textures or exsolution flames in chalcopyrite grains.

### Idaite

The existence of idaite in these sulphide occurrences was first described by the writer (Akıncı, 1974) although it has been recognized by other workers in several occurrences along the Eastern Black Sea Ore Province. It is identified by its strong reflection pleochroism and vivid, greenish polarisation colours by which it is easily distinguished from bornite (Photo 4). Due to chalcopyrite exsolution lamellae which are arranged along crystallographic planes showing a lattice texture and covellite replacement X-ray runs were not made but probe analysis confirmed the idaite composition. Formula derived from this composition is  $\text{Cu}_{4.96} \text{Fe}_{1.0} \text{S}_{5.80}$  which is similar to the original formula given by Frenzel (1959). On the other hand Levy (1967) gave the formula  $\text{Cu}_3 \text{FeS}_4$  based on microprobe analysis.

### Covellite

Two common forms develop either with cerussite and anglesite from dissolving, galena (Photo 4) or with goethite showing intergrowth-like, mottled texture due to the alteration of chalcopyrite and pyrite. Several examples were seen veining and replacing sphalerite grains, in some cases, leaving chalcopyrite inclusions behind. It also replaces chalcopyrite along grain boundaries forming colloidal bands or extensively with characteristic lattice structure and it develops together with veining digenite showing cellular cementation textures in chalcopyrite. Sometimes a covellite band occurs developing outwards around fahlerz grains enclosed in sphalerite.

### Digenite

In several specimens it is seen veining chalcopyrite irregularly showing a cementation structure. It replaces bornite as xenomorphic patches. Digenite has only been found in large quantities coarsely intergrown with galena as the decomposition product of betekhtinite showing atoll textures, mutual boundaries and lamellar development.

### Betekhtinite

Reported and described in detail by the writer previously (Akinci, 1976 *b*). It is found associated with galena, bornite, digenite, fahlerz, pyrite, quartz, dolomite and cerussite. Bornite as described previously was seen, in many cases rimming and veining betekhtinite. Myrmekitic intergrowths of galena-bornite are also enclosed in betekhtinite as well as rare galena-digenite intergrowths. In polished specimens it has a grey to creamy colour with a pinkish tint and a dull red or blue polarization colours. Cleavage is not commonly seen.

### Fahlerz

Since the colour is not a dependable property to differentiate tetrahedrite from tennantite (Htein, 1973) and known compositional zoning (Photo 8) and heterogeneity (Smirnov et al., 1972; Yui, 1972) most of the specimens were analysed by microprobe.

Minerals were found mostly replacing chalcopyrite and sphalerite in places leaving chalcopyrite inclusions in sphalerite behind. Pseudomorphs of hexagonal sections or prismatic projection of quartz crystals are common. Etching with  $\text{HNO}_3$  has revealed excellent zoning (Photo 8).

Tennantites were found to be more common than tetrahedrite. Previously only tetrahedrite was reported from the area (Ayışkan, 1959).

### Aikinite

Together with betekhtinite it was discovered for the first time in Turkey in these occurrences by the writer (Akinci, 1974). Minerals identified as aikinite have compositions which are not in agreement with the theoretical aikinite composition. They are suggested to be the members of aikinite bismuthinite solid solution series and are close to the aikinite and member. Complete solid solution possibilities were discussed by the writer (Akinci, 1979).

Aikinite grains are commonly found enclosed in chalcopyrite as rounded or elongated forms, or as an island in digenite which is also enclosed in chalcopyrite. Digenite always replaces aikinite along cleavage planes or occurs as patches throughout the aikinite grains. Replacement of sphalerite by aikinite was also observed in which tennantite was found replacing, veining or surrounding aikinite. It is usually found in association with pyrite and marcasite.

## PARAGENETIC SEQUENCE

The paragenesis of the ore and gangue minerals is given in Table 2. Although it is based on textural relationships, mineral stabilities and fluid inclusion study results, it is somewhat speculative because of the textural evidence for several ore and gangue minerals is not complete.

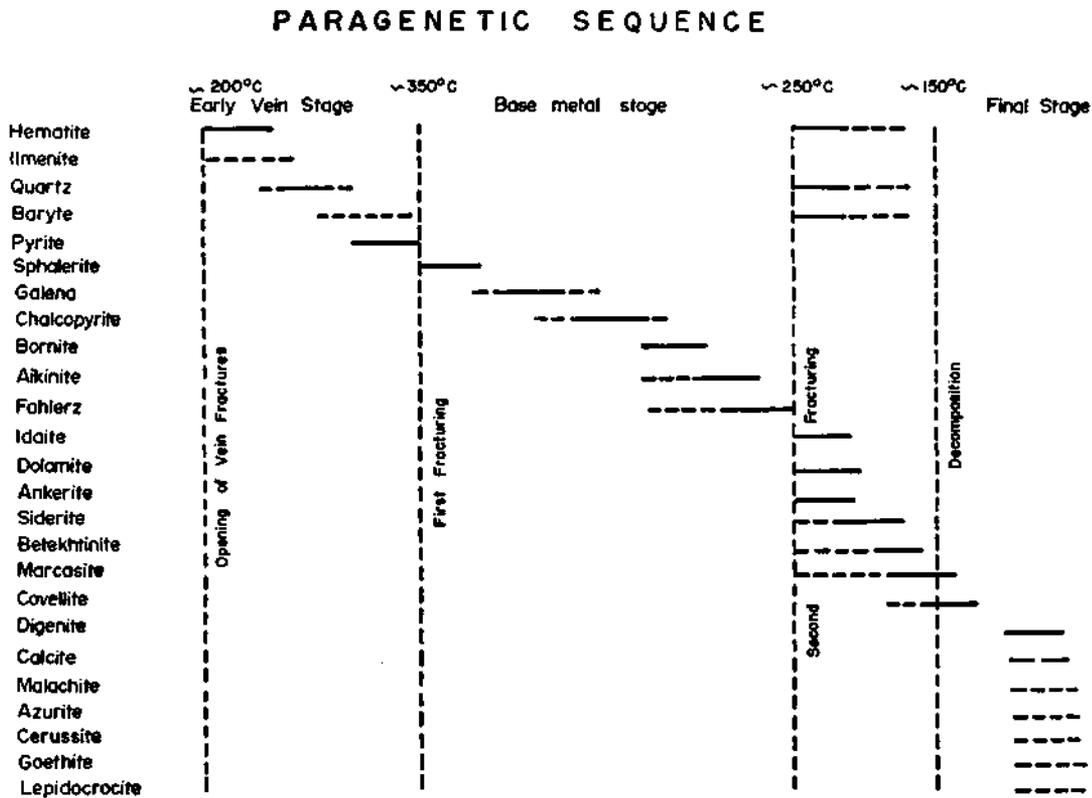
The lamellar form of hematite, from which ilmenite is being exsolved, and its textural relationship with quartz, suggest a formation at the beginning of the early vein stage. During this stage the temperature rises about 100°C with the formation of quartz followed by pyrite (Akinci, 1976a).

Fracturing occurs at the end of the early vein stage after the crystallization of the pyrite. This is indicated by cataclasis in pyrite, and veining in sphalerite.

Most of the ore minerals form in the base metal stage in the order given in Table 2. At the end of this stage a second fracturing occurs which is mostly marked by large scale vein filling by ganque carbonates.

Decomposition and alteration of the early formed ore minerals occur in the last stage of mineralization. These include decomposition of betekhtinite to digenite and galena, covellite and cerussite formation and limonitization.

Table - 2



#### DISCUSSION ON THE CRYSTALLIZATION PROCESSES

Thermal history of the vein mineralization was previously discussed by the writer in part (Akıncı, 1976 a). Since the minerals suitable for geothermometric measurements such as pyrrhotite, arsenopyrite and exsolved phases indicating high temperatures in sphalerite (i.e pyrrhotite) and chalcopyrite (i.e cubanite) are not present, only fluid inclusion study of ore and gangue minerals might help to reveal the time-temperature relations. According to Craig and Kullerud (1973) the association of hypogene idaite, marcasite, and digenite is not stable at temperatures above 430°C. This temperature range possibly indicates the highest temperature at which BulancaK sulphides start to grow from the ore-bearing fluids. Examination of polished specimens proved the existence of the idaite-marcasite-digenite association, but their hypogene origin is not certain.

The time-temperature curve (Akinci, 1976a, Fig. 1) shows a horizontal temperature zoning established by the hot spots and bisulphosalts in the Tekmezar area and the observed higher temperatures at the higher levels of some previously mined veins. This curve also indicates a temperature rise in the early vein stage during the crystallization of the early quartz. A rapid heat exchange between the ore-bearing fluids and wall rocks may cause the temperature of the solution to increase. Most of the wall-rock alteration processes are suspected to be exothermic and the rise of formation temperature in the early vein stage and progressive cooling of the later vein fluids can be explained in reference to Toulmin and Clark's (1967) discussion of the "Thermal Aspects of Ore Formation".

Quartz is seen to form at the beginning of the early vein stage. The early precipitated quartz, although its solubility is not affected by the pH of the solution and amount of the dissolved salts, later becomes more soluble with the increasing temperature which led to boiling. This is indicated by later stage pseudomorphs after quartz and skeletal and rounded, corroded crystals as seen in Photo 1. The earlier formation of quartz in the paragenetic sequence rather than pyrite, other sulphides and pyrrhotite may be explained as being due to the predominance of  $\text{SO}_2$  over  $\text{H}_2\text{S}$  at high temperatures (Meyer and Hemley, 1967; Kelly and Turneaure, 1970).

Destruction of  $\text{SO}_2$  during cooling of hydrothermal solutions can lead to the production of  $\text{H}_2\text{SO}_4$  and hence to the precipitation of sulphate minerals. Baryte is the least soluble and most common of the alkaline earth sulphates in hydrothermal systems (Holland, 1967). Simple cooling must be an important mechanism in the precipitation of barytes as well as other gangue minerals. If the solutions with which the hydrothermal compounds are mixed (e.g. connate waters rich in Ba) they are highly oxidized; the sulphate concentration can be increased at the expense of the bisulphide concentration. But in many epithermal deposits, baryte is associated, as in the Bulancak veins, with sulphide minerals which are unstable in highly oxidized environments.

Holland (op. cit.) suggested that the ratio  $\text{Ca}/\text{Mg}+\text{Fe}$  of unaltered silicate wall rocks is normally sufficiently low such that dolomite or an ankeritic carbonate, rather than calcite, will tend to form in alteration zones around hydrothermal veins. At Bulancak, dolomite is the dominant hydrothermal carbonate unlike the other known lead-zinc deposits elsewhere in the world where calcite is the dominant carbonate. As the solubility product of calcite and dolomite decreases with the increasing temperatures these cannot, therefore be precipitated from hydrothermal solutions by simple cooling. However if, during boiling, the  $\text{CO}_2$  content of the hydrothermal solutions decreases by pressure drops, then the carbonates can be precipitated.

It has been pointed out by Helgeson (1968) that pyrite becomes less soluble than pyrrhotite as the temperature drops below  $300^\circ\text{C}$ . With decreasing temperature, conversion of one sulphide to another and replacements may occur. In the Bulancak deposits the existence of marcasite with pyrite without any trace of pyrrhotite is distinctive.

Based on the experimental evidences, it has been shown that Mn-Zn and Pb are enriched in the aqueous phase during the crystallization of magma and maximum transfer of zinc into the aqueous phase has been shown by Holland (1972) to be very sensitive to the initial chloride content of alkali granite melts. The proportion of zinc removed increased from 78 % to 97 % if the amount present in the chloride content in the magma increases from 0.7 wt % to 1.5 wt %. The presence of pyrite as a common accessory mineral in granitic rocks and the reported sulphurization of iron and nickel in the host rocks adjacent to sulphide orebodies (Naldrett, 1966) and direct precipitation in many hot springs, suggest that there is always enough reduced sulphur to form base metal sulphides even in excess amounts.

These processes tend to precipitate sulphide from solutions in which either  $H_2S$  or more probably  $HS^-$  is dominant, and carbonates. In addition, replacement must have played an important role, e.g., replacement of dolomite by siderite as well as that of early sulphides by late stage sulphides. A plot of homogenization temperatures against paragenetic features support a single extended period of mineralization-in a column of liquid-for the sulphide deposition (Akıncı, 1976a).

### CONCLUSIONS

Bulancak sulphide veins are associated with tholeiitic and calc-alkaline volcanics of the Eastern Pontids. Most of the veins are exposed cutting across the lowermost lower basic series in the area. Veins are also found in the overlying dacitic and rhyodacitic rocks of intrusive and lava character.

The mineralization zone of 7-8 km in length and 3 km in width and lateral temperature zoning extend from Tekmezar to Darıköy in a northeasterly direction in which strike of the steeply dipping veins are perpendicular to that of the mineralization zone.

Polymetallic vein mineralization paragenetically is in the order of oxide, sulphate and sulphide-carbonate.

Fluid inclusion studies combined with ore microscopy support a single extended period of mineralization in a column of liquid.

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Photo 1 - Sphaerulite replacing euhedral quartz with pyrite: X 125, Parallel nichols, air.



Photo 2 - Bladed hematite crystals: X 125, Parallel nichols, air.



Photo 3 - Bladed hematite (white) in quartz (dark grey),  
colloform banding in goethite (light grey):  
X 300, Parallel nichols, air.

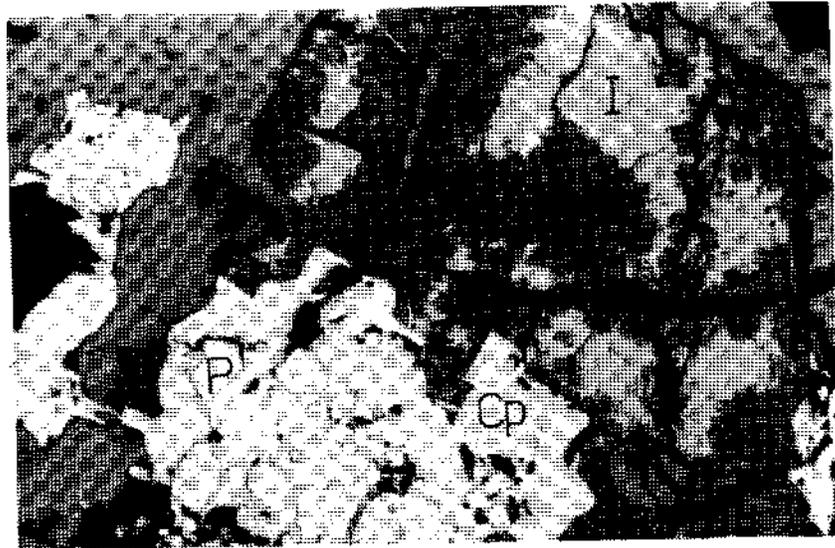


Photo 4 - Idaitite (I), covellite (Cv), chalcopyrite (Cp) and pyrite (P) relationship:  
X 300, Parallel nichols, air.



Photo 5 - Siderite replacing rhombohedral dolomite crystals, euhedral white pyrite crystal on the left: X 45, Parallel nichols, air.



Photo 6 - Galena (white) relicts in cerussite - coyellite intergrowth: X 45, Parallel nichols, air.

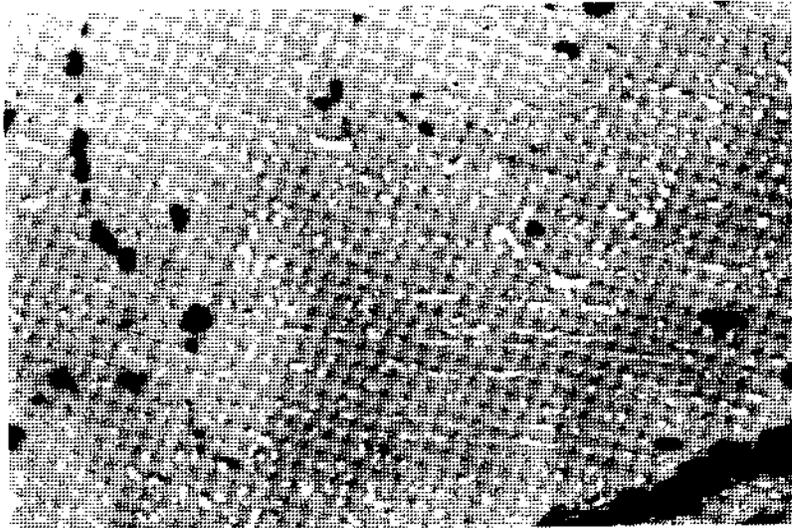


Photo 7 - Chalcopyrite exsolution in sphalerite:  
X 125, Parallel nichols, air.



Photo 8 - Zoning in fahlerz revealed by etching:  
X 125, Parallel nichols, air.