Relationship between Alteration, Rare Earth Element Distribution, and Mineralization of the Murgul Copper Deposit, Northeastern Turkey

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Abstract

The Murgul ore deposit belongs to the East Pontic metallogenetic province and is related to a subvolcanic formation of an Upper Cretaceous island-arc volcanism. The deposit is linked to a 250-m-thick felsic pyroclastic member of Senonian age.

The Murgul deposit consists of (1) a widespread disseminated ore with varying Cu contents ranging from 0.2 to 0.7 percent, (2) a stockworklike ore with average Cu contents between 1.0 and 2.5 percent, and (3) small ore lodes with Cu contents from 5.0 to 10.0 percent.

The mineralization is spatially associated with an intense two-stage alteration of the host rock: an initial stage of phyllic and argillic alteration, and a late stage characterized by silicification. The continuation of the volcanic activity with ascending hydrothermal solutions caused an intense remobilization of the disseminated ore producing the stockworklike and vein mineralizations. They consist of pyrite and chalcopyrite with minor contents of sphalerite, galena, and fahlore.

The study of the rare earth element distribution in the alteration zones supports the distinction between the two stages and reveals a close correlation between increasing wall-rock alteration and depletion of the rare earth elements. Petrographic data indicate that the rare earth element patterns of the altered host rocks are predominantly controlled by the abundance of sericite, illite, montmorillonite, kaolinite, and dickite. The formation of the two main orebodies must have been completed before an intense subaerial or intertidal erosion and weathering took place, This phase is indicated by a relatively thin layer of transgressive kaolinized weathering products which are overlain by barren felsic volcanics.

Introduction

MURGUL is one of the principal copper deposits of Turkey and has been the object of numerous studies, mainly during the past two decades. Even though different authors agree on the age of its formation, its genesis has remained controversial and it has been interpreted as both "submarine-exhalative-sedimentary" and "subvolcanic-hydrothermal" (Wijkerslooth, 1946; Schneiderhöhn, 1955; Kraeff, 1963; Sawa and Sawamura, 1970; Mado, 1972; Buser and Cvetic, 1973; Hilmer et al., 1974; Vujanovic, 1974).

As a supplement to the geochemical results reported by Özgür (1985), we have concentrated our studies especially on the rare earth element distribution in relation to alteration and formation of the ore deposit. Field observations show that the alteration processes, which are related to enrichment of the ore metals, were strictly linked to a distinct level of the volcanic sequence only.

A systematic sampling of the Murgul deposit and its environment has been performed in connection with a project on lithogeochemical indicators for base metal exploration. The combination of alteration and mineralization seems to reveal a strata bound event consistent with many similar deposits of the East Pontic metallogenic province (Dieterle, 1986).

Detailed results of the petrographical investigations on the alteration paragenesis will be reported in a separate paper. The aim of this paper is to elucidate the relationship between increasing alteration, depletion of rare earth elements, and formation of the ore deposit.

Geologic Setting

The Murgul deposit is situated in the northeastern part of Turkey (Fig. 1). There, the East Pontides represent an island arc extending south of the Pontic continent; they date from the Early Jurassic through the Miocene (Akýn, 1979; AkýnCý, 1980; Pengőr et al., 1980). The succession consists of a 2,000- to 3,000-m-thick volcanic sequence with relatively thin
The extrusive series has been divided into three volcanic cycles (Maucher, 1960; Maucher et al., 1962):

1. The first cycle comprises a volcanic pile deposited between the Jurassic and Upper Cretaceous. It is represented by initial basaltic activity (spilites) which changes progressively to felsic lava flows and thick pyroclastics in the middle and top of the cycle. The Senonian age of the mineralization in this pyroclastic sequence is based on paleontological evidence (Buser and Cvetic, 1973).

2. The second cycle starts transgressively with volcanic breccias, tuffs, and marine sediments of minor thickness overlain by andesitic and rhyolitic flows and followed by limestones of uppermost Cretaceous age (Maastrichtian).

3. The last cycle is introduced by a sequence of marine sediments of Paleogene age which are overlain by andesitic and minor basaltic lava flows constituting Tertiary volcanic activity.

The above-noted volcanic cycles represent the development of a late Mesozoic island arc according to various plate tectonic conceptions, which differ with respect to the positions of the continental and oceanic plates and the dipping direction of the subduction zone (Akin, 1979; Akinci, 1980; Sengör et al., 1980). Despite these controversial plate tectonic interpretations, the Jurassic through Lower Cretaceous spilites at the base of the Murgul volcanic pile (first cycle) display island-arc tholeiites, as indicated by trace element discrimination (Fig. 2). More information about the tectonic development of this area is not available due to a lack of geologic mapping.

The Murgul deposit is linked to the upper part of the first volcanic cycle and is associated with a 250-m-thick felsic pyroclastic sequence, the upper contact of which is marked by a thin layer of marine sediments (Sawa and Sawamura, 1970; Mado, 1972; Buser and Cvetic, 1973) and is characterized by intense erosion and weathering (e.g., kaolinization; Özgür, 1985).

The pyroclastic host rock consists of altered breccias and tuffs. Their primary mineral components can be observed only in a few remnants. In less altered samples the fluidal groundmass contains fragments of phenocrysts (plagioclase-An$_{28-3.5}$ and quartz) and plagioclase microlites (An$_{12-36}$), relics of hornblende and biotite, quartz, and minor quantities of apatite, sphene, and hematite.

The strike and dip of the lithologic sequence in the mining area are mainly controlled by an intraformational paleorelief caused by volcano-tectonic phases overprinted by younger block tectonic activities. Outcrops on the steep slopes of the Murgul valley in the vicinity of the deposit show angular contacts and mutual discordances between various pyroclastic members of the first cycle revealing very turbulent and repeated volcanic activity.

FIG 1. Geologic sketch map of the Murgul deposit. 1= andesitic lava flows of the uppermost Cretaceous, 2= hanging-wall felsic volcanics, 3= pyroclastic host rocks, 4= main faults, generally vertical movements, and 5= limits of the open pits (1983).
FIG 2. Ti/Cr-Ni discrimination diagram after Beccelova et al. (1979) for spilites of the first volcanic cycle from Murgul. A = ocean-floor tholeiites, B = island-arc tholeiites, C = CaO-enriched ocean-floor tholeiites, D = picritic lava. Black circles = spilites from Murgul.

The mineralized pyroclastics are overlain by 200 to 500-m-thick and barren felsic volcanics. In contrast to the mineralized host rock, the hanging-wall volcanics do not show either characteristic strong alteration patterns or any mineralization. Therefore, between the alteration of the host rock with its ore mineralization and the transgression of the barren volcanics, a temporal hiatus with a short period of erosion has to be assumed (Özgür, 1985). Corroboration for this assumption is represented by the clastic part of the Bognari orebody, interpreted by Mado (1972) to be an erosional product of the upper part of the Anayatak deposit (Fig. 1). Furthermore, the mineralized pyroclastics are topped by a strongly kaolinized layer of reworked tuffs, limestone, and sandstone (max. 10m) forming a marker bed. This level can be interpreted as the result of the intense weathering and superficial reworking of the pyroclastics and sediment lenses which were exposed subaerially during a relatively short period.

In contrast, the pyroclastics outside the mineralized area as well as the transgressive lavas show evidence of only weak alteration on a regional scale. The typical features of this regional alteration are chloritization of hornblende and biotite, and sericitization-albitization of feldspars (propylitic alteration). This differs significantly from the stronger alteration related to the orebodies, which is indicated by a concentric pattern of silicic, phyllic, and argillic zones (Figs. 3 and 4). With the silicic alteration a complete kaolinization of the feldspars is effected. This process does not correspond to the formation of the strongly kaolinized layer at the top of the sequence (the marker bed).
Ore Deposit

The Murgul deposit contains predominantly pyrite and lesser chalcopyrite. Minor quantities of galena, sphalerite, and fahlore occur locally only. The deposit is mainly mined in two open pits (Anayatak and Cakmakkaya). A third minor orebody, Bognari, came into production recently (Fig. 1). During the past ten years the average crude ore production has risen to about 1.8 million metric tons yearly. The recoverable reserves are estimated at 40 million metric tons with an average content of 1.25 percent Cu, 0.1 percent Zn, 0.05 percent Pb, 25 ppm Ag, and 0.2 ppm Au.

The mineralized zones are funnel shaped, with oval surface sections oriented north-northwest. The thickness of the mineralized zones has been estimated from results of drilling programs to be about 2,50 m. In general, the following textural types of ore mineralization can be recognized.

Type 1

A disseminated ore type occurs commonly over the entire deposit. It shows varying Cu contents ranging from 0.2 to 0.7 percent. This type seems to be the oldest and primary ore mineralization because it is crosscut by all the younger vein types. Ore minerals of this type are generally fine grained (about 1-2 mm in diam) and intergrown with quartz, sericite, and clay minerals; locally it shows a layered structure of pyroclastic material in combination with fine lenses of jasperoid matter (Fig. 5).

A peculiar problem arises with the fabrics of disseminated primary pyrite. A genetic contradiction exists with regard to the appearance of pyrite framboids in vast quantities; these were formerly interpreted to be products of a very low thermal (Schneiderhohn, 1923; Love, 1964; Love and Amstutz, 1969) environment (e.g., 100°C). However, in this case they must have been precipitated in a pyroclastic sequence during the late stage of volcanic activity in a hydrothermal range of about 280°C, as indicated by fluid inclusion measurements (Özgür, 1985; Özgür and Schneider, 1988).

FIG 5. Fine-grained and disseminated ore minerals (type 1) (black) with intergrowths of quartz (white), sericite, and clay minerals (gray), locally depicting the layered structure of the pyroclastic matter. Anayatak open pit.

Type 2

This stockworklike mineralization with average Cu contents between 1.0 and 2.5 percent indicates a younger phase of hydrothermal remobilization during an interval of volcano-tectonic fracturing of the pyroclastic pile. The mineralization crosscut type 1 as well as the altered host rocks, mainly in network structures (Fig. 6). The size (about 2-3 mm in diam) of the ore minerals is noticeably larger than of type 1. Thus, the sphalerite and galena crystals are occasionally visible in addition to the predominating pyrite and chalcopyrite.

FIG 6. Network ore (type 2) in altered pyroclastic host rocks (gray, white) from the Anayatak open pit.
Type 3

Small ore lodes with Cu contents from 5.0 to 10.0 percent are developed mainly as relatively short veins and are essentially concentrated in the central part of the mineralized zones. The ore paragenesis indicates occasional open-space fillings with coarse euhedral crystals (about 10 mm in diam) of pyrite, chalcopyrite, and quartz, locally covered by a thin skin of (secondary) covelline (Fig. 7). They are interpreted to be the latest open-space fillings which occur near the surface and were generated by discharging hydrothermal solutions.

FIG 7. Open-space filling (type 3) of a short vein from the Çakmakkaya open pit with euhedral crystals of pyrite, chalcopyrite, and quartz (white).

TABLE 1. Mineral Assemblages of the Altered Pyroclastics

<table>
<thead>
<tr>
<th>Sample</th>
<th>No. 1</th>
<th>No. 2</th>
<th>No. 3</th>
<th>No. 4</th>
<th>No. 5</th>
<th>No. 6</th>
<th>No. 7</th>
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<th>No. 9</th>
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<th>No. 14</th>
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<tr>
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<tr>
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<tr>
<td>Montmorillonite</td>
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<td>15</td>
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<td>9</td>
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Mineral abundances are expressed in vol percent; samples 4 to 6 (phyllic alteration) are from the Anayatal open pit, samples 7 to 10 (argillic alteration) are from the area between both open pits, samples 11 to 15 (silicic alteration) are from the central part of both open pits (for locations see Fig. 3). — = not observed.

Hydrothermal Alteration and Mineralization

In order to determine the quantitative ratio of the secondary minerals, 89 thin sections were analyzed by means of a point counter. About 4,000 points were counted in each thin section. The results are briefly given in Table 1.

The intense host-rock alteration of both orebodies may be divided into an initial stage represented by a central phyllic zone surrounded by a peripheral argillic zone and a late stage of pervasive silicification (Figs. 3 and 4). No potassic zone has been observed.

The first stage of alteration led to destruction of the primary paragenesis of the pyroclastics and to the replacement of the host rock by quartz and pale green to greasy sericite. During this stage an extensive but poor mineralization of disseminated pyrite and chalcopyrite took place. In some thin sections, small veinlets of calcite crosscut the phyllic assemblage, indicating a late stage of calcite crystallization. The phyllic zone together with the early mineralization shows relatively sharp contacts with the argillic zone containing 1- to 3-m-wide transition spheres. The surrounding zone is characterized by pervasive argillitization in which the alteration assemblage consists of quartz, montmorillonite, dickite, illite, and pyrite.

The late stage of hydrothermal activity is represented by silicic alteration, which consists of quartz replacement of the volcanic host rock, as cryptocrystalline jasper. Kaolinite occurs occasionally as a relict component. The available petrographic and field data indicate that the mineral assemblage of the silicic alteration was superimposed on the earlier phyllic alteration paragenesis because patches of it are included in the silicic zones (Fig. 3). The mineralized veins and veinlets (types 2 and 3) crosscut the altered host rock. Consequently, from geochemical observations the mineralization of the vein ore types can be interpreted to be a younger phase of ore remobilization (Özgür and Schneider, 1988) which was generated by a last episode of volcanic activity. During this time interval mechanical disintegration (volcano-tectonic or hydrofracturing) took place,
opening faults and fissures for ascending ore-bearing solutions which resulted in the formation of the stockworklike mineralization (type 2). The source of the ore components is considered to be the deeper part of the altered volcanic pile with its disseminated ore content, as indicated by geochemical trends (Özgür and Schneider, 1988).

Finally, the latest open-space fillings (type 3) suggest ore mineralization closely below the terrestrial surface; they indicate a further uplift of the volcano-thermal field. Presumably, during this final stage the various episodes of intensive erosion processes had already started.

According to Beane (1982), a mineral assemblage reflects changing pH-fs₂ conditions of the hydrothermal solutions. From a low pH and intermediate sulfur fugacity regime under which phyllic and argillic alteration took place, the physicochemical environment varied to a low pH and high sulfur fugacity system which upgraded mineralization during silicic alteration.

**Behavior of the Rare Earth Elements**

In 12 samples of altered volcanics and in three samples which are relatively free of alteration, seven rare earth elements were determined by instrumental neutron activation at the Hahn-Meitner Institut, Berlin. The routine precision was estimated at better than 9 percent for most elements (Dulski and Möller, 1975) using GSP-1 of the U.S. Geological Survey as the reference standard. The rare earth element data were normalized to their abundance in chondrites according to Boynton (1984).

The less altered samples (Table 2) were taken from pyroclastics located between 500 and 700 m west of the mineralized and altered area. In these samples the alteration on a regional scale is generally weak. The abundance of secondary minerals ranges between 3 and 7 vol percent only and the plagioclases maintain their original character. Thus, the rare earth element patterns of these samples should nearly preserve their original geochemical characteristics. A comparison of Tables 1 and 2 reveals a plausible correlation of the mineral assemblages and their geochemical data for both altered and less altered sample groups. The rare earth element plots are summarized in Figures 8 and 9. Examination of the rare earth element distribution in samples from the different alteration zones shows the response of rare earth elements to the alteration processes.

**Initial stage of hydrothermal alteration**

Compared with the less altered pyroclastics the samples from phyllic alteration zones show a moderate depletion from La through Lu (Fig. 8a, Table 2). This suggests that sericite could have fixed the light rare earth elements preferentially.
According to Alderton et al. (1980), white mica can take up a considerable amount of trivalent rare earth elements.

The pyroclastics with argillic alteration show a general rare earth element depletion, which seems to be more intensive than in the rocks affected by phyllic alteration only (Fig. 8b). Generally the heavy rare earth elements were preferentially released from the rocks during argillic alteration.

**Late stage of hydrothermal alteration**

Since silicic alteration was superimposed on the phyllic zones, the rare earth element patterns are compared to those of rocks with phyllic alteration. The silicic altered rocks exhibit a strong decrease in rare earth elements, which emphasizes the shape and slope of the samples, with phyllic alteration at a lower concentration level (Fig. 9). The greater depletion of the rare earth elements in argillitic compared with phyllic alteration reactions may be correlated with the decrease of suitable cation positions in the clay mineral-rich facies.

The secondary paragenesis (Table 1) suggests that sericite, montmorillonite, illite, dickite, and kaolinite are the principal minerals fixing rare earth elements during alteration. It is implied in Figure 10 that rare earth elements of the altered rocks are predominantly controlled by these secondary minerals. The potential of phyllosilicates for rare earth element adsorption has been discussed by Roaldset (1973, 1975) and Alderton et al. (1980). The example presented from the Murgul deposit (Fig. 10) may further support this apparent correlation.

In summary, the data indicate a progressive rare earth element depletion in the host rock together with an increase of intensity in phyllic, argillic, and silicic alteration. These observations can partly be explained by: dilution effects caused by the modal increase of quartz, the decrease in density that accompanies generally pervasive argillitization, the inability of the secondary minerals to fix all the released rare earth elements during alteration, and the general leaching effect of the recurring circulation of hydrothermal solutions. Furthermore, the original Eu anomaly
tends to disappear with increasing grade of alteration.

FIG 10. Correlation between rare earth element concentration and the abundance of sericite and clay minerals in the altered rocks samples. Open circles = phyllic alteration, black circles = argillic alteration, black triangles = silicic alteration.

Discussion

In the last decades the genetic interpretation of the Murgul deposit has been the subject of a distinctive controversy. Various results obtained by this study necessitate a new consideration of the genesis of the deposit. It can be related to a subvolcanic process associated with an Upper Cretaceous island-arc volcanism developed under temporally terrestrial conditions. The subvolcanic character of the deposit can be inferred from the following observations:

1. The sparsely and only locally intercalated marine sediments in the thick volcanic host-rock sequence indicate a shallow-water depositional environment at least for the upper part of the (mineralized) first cycle. The violent volcanic activity during this time is documented by various outcrops extending over more than 160 km in an east-west direction, which suggest an island chain. In Murgul, stratiform mineralizations are not known either in the sedimentary lenses intercalated into the pyroclastic sequence or in the adjacent stratified tuffs of the same stratigraphical level. Thus, the mineralization is endogenous in respect to the development of the volcanic pile. However, farther to the west, from Trabzon through Giresun, the paleogeographic features change within the East Pontides to real submarine tuff depositions with synsedimentary ore enrichments (Maucher, 1960; Maucher et al., 1962).

2. The thick pyroclastic sequence has been altered and mineralized in its upper part during a late stage in the volcanic activity by ascending hydrothermal solutions.

3. The formation of the two orebodies must have been completed before a short time interval of intense subaerial or intertidal erosion and weathering took place. Furthermore, this interval is represented by a strongly kaolinized layer of reworked pyroclastics and sediments (max 10m) like a regional marker bed (Özgür, 1985; Dieterle, 1986) which is taken as evidence for a temporal terrestrial environment. This marker bed can be interpreted to be the result of a short period of intense weathering and superficial reworking of the pyroclastics together with their ore content.

4. The mineralized and intensely altered host rocks are overlain by a series of relatively low-grade altered and barren felsic volcanics. It should be noted that the mineralization nowhere traverses the marker bed. This fact emphasizes that the deposits were formed prior to the erosional interval and the later eruption of the hanging felsic volcanics.

5. The mineralized host rock shows structures similar to the "ore-related-breccias" described by Sillitoe (1985).
They suggest a local subsurface brecciation that might have been generated by repeated volcanic activities which reheated the system, because there is evidence for several nearly contemporaneous eruption centers at a distance of a few hundred meters. Although the features of the concentric alteration-mineralization patterns (Fig. 11) show some similarities to porphyry ore deposits described by Lowell and Guilbert (1970), there are some remarkable differences: (1) the high-grade ore is mainly concentrated in the center, (2) there is no potassic alteration zone observable, and (3) the mineralization must have taken place relatively close to the surface.

The two stages of hydrothermal alteration have been separated into an initial stage characterized by phyllic and argillic alteration, and a late stage associated with silicification. The mineral assemblage of the initial stage is well preserved in the distal zones of the deposit whereas the late stage marks the center. This observation is supported by an at least two-stage depletion of the rare earth elements in the host rock during alteration. Furthermore, the data provide unequivocal evidences of rare earth element mobility: the rare earth elements were progressively released from the rocks in good correlation with the increase in the hydrothermal effect, which reached its maximum in the silicified center with the strongest ore enrichment.

Finally, the Murgul deposit mineralization resulted from at least three periods of increasing concentration of the ore material (Fig. 11). After the deposition of the thick dacitic pyroclastic sequence, subsequent volcanic activity caused the disintegration of the pile at isolated small centers, thus producing optimal permeability. During this stage hydrothermal ore solutions spread upward forming the first phase of a disseminated mineralization (type 1). The continuation of hydrothermal activity, combined with a further breakup of the host rock, led to an additional concentration of the ore matter into two younger generations of vein mineralization (types 2 and 3). They have been recognized close to the surface of the volcanic sequence due to the open-space mineralization of the youngest ore lodes (type 3).

This observation indicates an increasing uplift of the area leading to its emergence above sea level for a short period. During this time, the uppermost parts of the deposit would have been eroded and weathered. The products of such processes are concentrated locally in the elastic part of the minor Bognari orebody which reveals the typical mineral assemblages of the Anayatak ore. The violent weathering in this relatively short period could have been intensified by a discharge of hydrothermal fluids during a fumarolic stage. These processes led finally to the formation of a transgressive, psammitic, and intensely kaolinized layer which occurs in isolated lenses as a marker bed. This fact points to a temporal hiatus between the formation of the deposit and the later transgression of the hanging felsic volcanics. The characteristic layer at the top of the deposit marks the end of its formation because the transgressive hanging felsic volcanics are barren and without any geochemical anomalies which would be indicative of a mineralized host rock beneath the marker bed (Fig. 11). The above observations and interpretations suggest that the Murgul deposit resulted from a subvolcanic hydrothermal process and not an exhalative-sedimentary one. Therefore, comparison with kuroko-type deposits is only possible with respect to the volcanic-arc position of the Murgul deposit, since the geochemical data seem to make it more related to a Fiji-type deposit (Colley and Rice, 1975; Özgür and Schneider, 1988).

Reviewing the entire East Pontic metallogenetic province, a genetic correlation appears between Cu/(Pb + Zn) ratios of the various ore deposits and the
intensity of volcanic activity, both of which increase from west to east and culminate at Murgul (Möller et al., 1983). In the western part the Upper Cretaceous deposits are predominantly stratiform (Maucher, 1960; Maucher et al., 1962) whereas in the eastern part they are generally strata bound in a subvolcanic position as stockworks, veins, and disseminations, Both types are confined to dacitic and andesitic volcanics of Upper Cretaceous age.

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