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SEA-FLOOR SULFIDE MINERALIZATIONS OF MUNELLA DEPOSITS, CENTRAL MIRDITA, ALBANIA

A. Sinojmeri

ABSTRACT

Munella deposits are related with a bimodal volcanism represented by rhyodacites and basalts. Galena and sphalerite concentration at the upper part and at the rim, and pyrite-chalcopyrite concentration in the central part of stratiform orebody delineate a scarce mineralogical zonality. Baryte and tennantite-tetrahedrite appears frequent in polymetallic mineralizations while visible gold inclusions are found in the bornitic and in the disseminated ones. Thermochemical calculations led in temperature forming up to 350°C, while $\delta^{34}S$ values ($\approx +3.5^{o}/oo$) v.s. C.D.T. suggest limited participation of sea-water in the mineral formation processes.

INTRODUCTION

The Munella deposits represent a sulfide volcanogenic mineralization developed in the upper part of volcanic sequence of Albanian ophiolites. The presence of polymetallic mineralizations with Cu, Pb, Zn, Sb, Au as an additional of pyrite-chalcopyrite one increases the interest for this deposit.

The samples carried out by drilling constitutes several vertical orebody cuts and represent different types of mineralizations.

Results of studies of actual sea-floor mineralizations are used to appreciate the geological conditions of mineral formation.

METHODS OF ANALYSES

The geochemistry of lavas is based on bulk analyses of the rocks made in CRPG-Nancy, France using x-ray florescence and atomic absorption, while the measurements of REE contents and Y are made using plasma emission. The data of macro-element content are recalculated after the subtraction of loose weight on ignition, where the REE contents are normalised vice chondrites. Bulk chemical analyses of ores made at the Analytical Department of Institute of Geological Researches of Tirana, Albania are used for the programation of microprobe WDS analyses in Analytical Department of BRGM-Orléans, France. Prolongated account times are applied for the dosage of low-content elements as Au in arsenopyrite and pyrite, In in sphalerite etc. Minimum detection levels (MDL) are calculated for trace-elements, where for the macro-elements are calculated the relative errors. Scanning electron microscopy is applied on polished sections to observe the oscillatory zonation of arsenopyrite, sphalerite, tennantite-tetrahedrite, gold etc or to definite small inclusions as proustite, bournonite etc.

Sulfur isotopic analyses are also made in Analytical Department of BRGM-

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Polytechnic University, Faculty of Geology and Mining, Tirana, Albania

Orléans. Only microscopic observations are made in fluid inclusions from disseminated mineralizations. Salt cubes within these inclusions indicate approximative temperature forming, where aren't made microthermometric measurements.

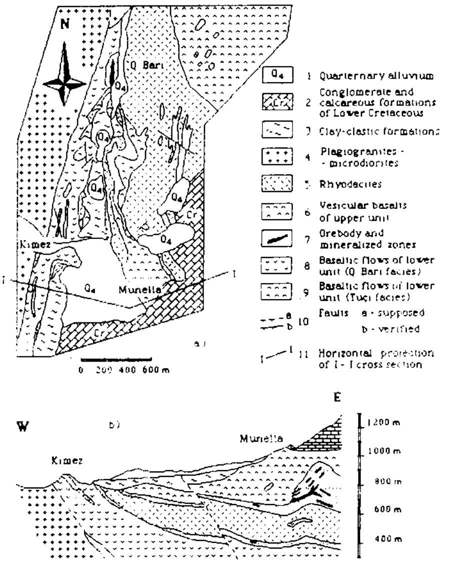

Fig. 1: Geologic map and cross section of Munella region.

GEOLOGICAL SETTING

The Albanian ophiolites are included in Mediterranean ophiolites and take a full development in Mirdita zone. There are two important moments of the ophiolitic volcanism development in Albania (Kodra and Gjata, 1989): the first during the slow-spreading of Mirdita Sea at Lower Malm where the tholeitic volcanites (the border of Mirdita zone) were generated, and the second during the fast-spreading of Mirdita Sea at the Upper Malm where the calcalkalin volcanites (Central Mirdita) were generated.

Two important units are distinguished in Central Mirdita volcanites (Shallo et al., 1987): the lower unit compiled by massive basaltic flows intersected by granodiorite dykes and the upper unit composed by rhyodacitic formations with subordinated andesitic flows, where the black vesicular basalts constitute the top of the sequence.

Sea-floor mineralizations of Munella (fig.1) is located between rhyodacites and vesicular basalts of upper unit (Bezhani et al., 1989; Sinojmeri, 1990) and were developed during a tectonic stress change from strongly extensional to weakly extensional (Sinojmeri, 1992). One North-Sud trending synform deformation lead to overturned folds and frequently sheared zones in the Eastern limb.

Underlying rhyodacites, up to 300m thick are composed mainly by massive ryhodacitic flows with any intercalation of volcanic glaces. The ryhodacites are affected by hydrothermal alteration giving kaolinization and zeolitization which increase hardly near the stockworks.

The black vesicular basalts overlie the stratiform mineralization. The vesicles more than 5 mm in diameter which are filled with fluorite and calcite besides silica, sulfides and silicate minerals characterise the lavas at the end of the volcanic activity. Several mineralized levels with disseminated sulfides are developed within these basalts. The black colour of basalts is due to the chloritization.

The zeoliferous (up to 80% zeolite) rocks (up to 3m thick) divide uncontinously the basalts and the rhyodacites. The principal mineral of these rocks is heulandite. Between the phenocrystals of heulandite are found quartz and albite spherolites.

CHEMISTRY OF THE SURROUNDING ROCKS

The Central Mirdita volcanites are Ti and K poor and their chemical composition is similar to those from island arcs volcanites (Shallo et al. 1989).

Table 1: Chemical composition of surrounding rocks.

	87	88	89	90	91	93	94
				wt%			
SiO_2	45.57	52.84	70.34	73.02	74.59	73.39	77.05
$Al2O_3$	17.07	12.64	11.05	10.75	9.64	10.03	9.33
$Fe2O_3$	11.91	6.34	5.30	4.87	6.95	7.79	4.47
MnO	0.20	0.08	0.05	0.05	traces	0.06	0.11
MgO	5.24	1.31	1.43	2.83	0.17	1.63	1.92
CaO	8.24	5.44	3.24	0.73	0.19	0.29	1.27
Na_2O	2.75	0.56	2.70	2.59	0.12	0.11	2.77
K2O	0.28	0.72	0.24	0.28	2.34	1.41	0.11
TiO_2	0.64	0.34	0.32	0.34	0.32	0.32	0.29
$P2O_5$	0.17	0.19	0.17	0.17	0.17	0.24	0.17
L.I.	7.81	12.34	4.52	4.28	5.32	3.97	2.95
Total	99.88	99.80	99.36	99.91	99.88	99.23	100.44
				ppm			
Ba	96.0	300.0	60.0	44.0	609.0	133.0	76.0
Be	1.2	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Co	44.0	7.0	7.0	6.0	102.0	<5	6.0
Cr	25.0	7.0	6.0	18.0	6.0	8.0	9.0
Cu	193.0	52.0	19.0	5.0	5.0	>1000	8.0
Ga	22.0	6.0	5.0	15.0	<5	9.0	6.0
Nb	<5	<5	<5	<5	<5	<5	<5
Ni	24.0	6.0	<5	14.0	<5	6.0	<5
Rb	9.0	9.0	5.0	7.0	33.0	23.0	<5
Sc	56.2	19.2	19.3	16.0	14.7	15.1	13.0
Sr	74.0	24.0	34.0	24.0	<5	<5	48.0
Th	<5	<5	<5	<5	<5	<5	<5
V	461.0	5.0	8.0	13.0	12.0	7.0	11.0
Y	11.0	32.0	32.0	32.0	24.0	29.0	25.0
Zn	104.0	323.0	187.0	40.0	8.0	41.0	170.0
Zr	5.0	34.0	31.0	37.0	29.0	36.0	22.0

The chemical analyses from surrounding rocks of sulfide mineralizations (tab.1) reveal important variations of SiO_2 content which after the correction for the loss on ignition result from 48.79% to 78.63%, low TiO_2-contents (0.30 to 0.68%) and an average content of REE fluctuating around 12ppm (fig.3). The Si-content of rhyodacites underlying stratiform sulfide mineralization increases (SiO_2 from 73.71 to 78.63%) where the REE content generally decreases dipping in rhyodacitic sequence. Light REE are leached away during the hydrothermal alteration around the stockworks, nevertheless in their distribution is preserved the negative anomaly of Eu (fig.3).

The zeolitized rocks are richer in Ca and Al and poorer in Si than rhyodacites (tab.1), presenting a chemical composition which correspond to the altered andesites. The absence of the Eu negative anomaly distinguish the REE distribution of andesites from the other Munella lavas.

The vesicular basalts at the top of Munella volcanic sequence is characterized by a low Ti content and high Al content (fig.2). Such a type of basalts delineate the upper part of volcanic sequence in the island arcs (upper lavas of Troodos or Oman), testing the impoverishment of the source

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with incompatible elements at the end of the magmatic evolution (Beurrier et al., 1989; Ohnenstetter and Sider, 1988).

The REE contents are evidently lower in basalts than in rhyodacites and the Eu negative anomaly is smoothed in comparison with those in rhyodacites.

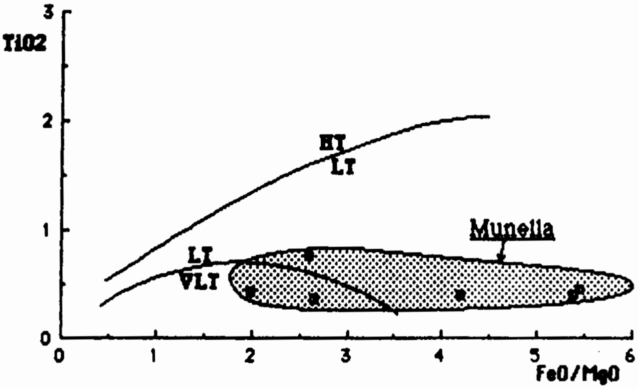

Fig. 2: Projection of (FeO_{total}/MgO)-(TiO_2) compositions of Munella lavas on the fields of Myashiro (1973).

SULFIDE MINERALIZATION

Three different mineralization facies are distinguished in Munella deposits:

1. Massive stratiform mineralization situated mainly between rhyodacites and vesicular basalts.
2. Stockwork mineralization developed in rhyodacites.
3. Disseminated mineralization mainly developed within vesicular basalts.

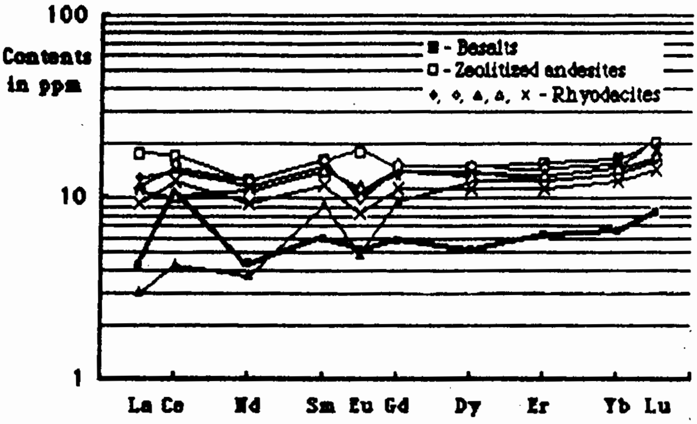

Fig. 3: Distribution of rare earth elements in the Munella lavas.

1. The massive stratiform mineralization dip to west by the inclination angles from 60° (eastern part) to 10° (western part) following the contact between rhyodacited and basalts. The orebody, up to 50 m thick, including interlayerd thin basaltic flows, is covered here and there by jasper thin beds (up to 30 cm thick). The hydrothermal alteration of underlain rhyodacites is not important and become scarce dipping in.

Three types of mineralizations constitute the massive stratiform orebody; the pyrite mineralization, pyrite-chalcopyrite mineralization (yellow ores) and polymetallic mineralization (black ores). The pyrite mineralization

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composed mainly by pyrite (≈ 90%) generally occupies the central part of massive mineralization. Chalcopyrite, sphalerite and quartz are present in small quantities. Increasing degree of crystallinity (cockade textures) of pyrite indicate the change from high to normal oversaturation of hydrothermal solutions, reflecting abrupt to slow cooling conditions.

Polymetallic mineralizations are concentrated at the western and eastern extremities of massive orebody. Chalcopyrite, sphalerite and bornite predominate on pyrite in yellow ores while sphalerite galena and tennantite-tetrahedrite predominate on chalcopyrite and pyrite in black ores. Besides Cu, Fe, Zn, Pb, As, Sb, S which are the main elements of these minerals there are found Ag, Au, Cd, Tl, Ge, Ga, Ba. The galena and sphalerite concentration at the upper part and the chalcopyrite concentration at the lower part delineate a scarce mineralogical zonation of massive stratiform orebody. High temperature baryte appears frequently in polymetallic mineralizations and visible grains (up to 50 μm) of native gold are also found.

High contents of Bi (up to 1.3%), Te (up to 1.9%), Ag (up to 1.2%) and Ga (up to 0.7%) (tab. 2) are found in tennantite-tetrahedrite, but there are also Hg-rich (up to 0.3%) chalcopyrite and Bi and Sb rich (1.1 and 4.9% respectively) galena (tab. 3). Sb-rich galena is associated with geochronite $Pb_{14.2}Cu_{0.1}(As_{2.4}Sb_{3.5})S_{22.9}$, tennantite - tetrahedrite $Cu_{9.8}(Fe_{0.1}Zn_{1.8})(As_{2.2}Sb_{1.8})S_{12.9}$, baryte, calcite and native gold at the orebody top. Always in the high level mineralizations other Sb-rich associations like galena, sphalerite, tetrahedrite (Sb/As =3/1) and bournonite (Sb/As =4/1) are found.

Table 2: Microprobe analyses of tennantite-tetrahedrite from polymetallic ores.

	Cu	Ag	Fe	Zn	Pb	As	Sb	S	Se	Other Elements	Total
12	40.8	-	1.07	8.14	-	10.89	13.82	26.3	0.18	Ga=0.72	101.92
15	42.5	-	1.01	7.23	0.26	15.46	2.70	27.4	-	Bi=1.06 Te=1.92	99.54
126	28.3	1.20	0.62	7.03	-	3.30	25.59	25.7	-		101.74
129	40.3	-	0.59	7.76	-	10.61	13.54	26.5	-		99.30
130	42.1	-	0.58	8.38	-	19.69	-	27.4	-	Hg=0.13	98.28

Colloform textures and successive replacements (ghost baryte textures) indicate the quenching effect in these mineralizations. Only at the final mineralforming stages it is possible to distinguish crystallisation in equilibrium conditions.

The Fe-content (≈ 0.15 at %) in sphalerite equilibrated with chalcopyrite-pyrite-bornite association indicate a temperature of about 250°C and oxygen activity 10-9 of the mineralforming solutions (Czamanske, 1974; Hannington and Scott, 1989). The gold presence in bornite associations confirm the conclusions of Hannington and Scott (1989) about the high solubility of gold as $Au(HS)_2^-$ complex in such conditions. The hematite crystallisation at the end of the sequence suggests that cooling the system attempt to

Table 3: Microprobe analyses from Sb and Bi rich galena.

	Pb	Ag	Bi	As	Sb	S	Other Elements	Total
47	80.6	-	1.07	-	-	14.0	Cu=1.42 Fe=0.92	98.01
126	84.4	-	-	0.17	0.42	13.6	Cu=0.07 Ti=0.18	98.84
127	81.1	-	-	-	3.95	14.2		99.25
128	80.8	0.15	-	-	3.37	14.1	Cu=0.29	98.71

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increases the oxygen activity favouring bornite to chalcopyrite crystallisation (Sinojmeri et al., 1988). The precipitation of native gold seems to be closely related with this increase of the oxygen activity.

The chemical composition of tennantite-tetrahedrite confirm the conclusions of Sack and Loucks (1985) about the thermochemical affinity between As → Sb and Fe → Zn substitutions. The thermodynamic relations considering the Fe and Zn distribution in tetrahedral sites of tennantite-tetrahedrite equilibrated with Fe-poor sphalerite lead to a mineralforming temperature of about 300°C (Sack and Loucks, 1985).

Marcasite presence in some colloform pyrite-sphalerite associations suggest temperatures lower than 250°C and pH values smaller than 5. Small pH values (≈ 2) are found in several sites of active back-arc structures (Fouquet et al., 1991).

Characteristic textures composed by euhedral non jointed chalcopyrite crystals, replaced successively by sphalerite, apatite and chlorite, very similar to those observed in black chimneys from modern see-floor mineralizations (Juan de Fuga, 21°N etc) and fossil ones from Cyprus (Oudin and Constantinou, 1984) are found in black and yellow ores of Munella. Brecciated ores, composed by massive pyrite-chalcopyrite associations indicate the impulsive activity of black smokers during the massive ore formation. Traces of vital activity of worms are present in these mineralization.

The isotopic analyses of sulfur point out a $\delta^{34}S$ of about 3.5^{o}/oo, that indicate a profound origin of sulfur and a less participation of the sea-water. The isotopic desequilibria between pyrite chalcopyrite and sphalerite confirm the quenching effect suggested by the textural study.

2. The stockwork mineralizations developed in rhyodacites characterise the feeder zone of stratiform massive mineralization. Dipping to East with 30° angle, their thickness and their metallogenic potential hardly decrease. Their principal mineral association is quartz, pyrite, chalcopyrite and rarely sphalerite, though small bornite inclusions appears occasionally in pyrite. The grained textures and the absence of colloform textures characterise the stockwork mineralization.

The combined chalcopyrite-sphalerite crystals analogous to stannite-sphalerite crystals described by Oen et al. (1980) represent a special facies of mineral forming in annealing conditions. This sphalerite presents Ga and In enriched zones (0.5 and 0.3% respectively; tab. 4), accordingly to the heterovalency substitutions :

$Cu^{+} + In^{3+} \Leftrightarrow 2Zn$ and $Cu^{+} + Ga^{3+} \Leftrightarrow 2Zn$

presented by Johan (1988).

The marcasite missing in stockworks indicates temperatures higher than 250°C while the $\delta^{34}S$ in these mineralizations is similar to those from stratiform one.

Table 4: Microprobe analyses from In and Ga rich zones of sphalerite.

	Cu	Fe	Zn	Cd	Ga	In	As	S	Other Elements	Total
13	0.81	0.89	65.8	0.18	-	0.18	0.13	32.4	Sb=0.09 Ag=0.12	100.60
24	0.93	0.59	65.4	0.21	0.62	0.28	-	33.1		101.13
24/1	2.13	1.26	63.0	0.20	0.50	0.26	0.08	32.6	Sb=0.19	100.22
24/2	0.68	0.51	64.8	0.20	-	0.22	-	32.7		99.11

In_{MDL} and Ga_{MDL} for counting times 30" are 0.08%.

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The arsenopyrite-sphalerite-galena association is a special case of stockwork mineralizations. The Sb-rich zones (up to 2.5%) in arsenopyrite and the presence of pyrargyrite (Ag_3SbS_3) indicate the elevated presence of Sb in hydrothermal solutions. The oscillatory zoning of arsenopyrite and sphalerite crystals as well as the presence of pyrite, marcasite and ghost pyrrhotite textures suggest important changing of physico-chemical conditions during the mineralforming processes. Nevertheless, the edges of arsenopyrite crystals seems to be homogenous and equilibrated with sphalerite (tab. 5, anal. 5, 6).

The chemical composition of these large homogenous zones in arsenopyrite and Fe-content of sphalerite in log aS_2-temperature diagram (Scott, 1983) point out a crystallization temperature of about 400°C and a sulfur activity of 10^{-8}.

Table 5: Microprobe analyses from a zonated arsenopyrite crystal.

	Fe	Co	As	Sb	S	Total
1	34.1	-	44.4	2.47	20.0	100.97
2	32.5	0.11	52.8	0.25	15.9	101.56
3	34.7	-	44.6	0.99	20.6	100.89
4	34.0	0.10	47.4	0.22	18.7	100.42
5	34.7	0.04	43.0	0.16	21.3	99.20
6	34.6	-	43.5	0.11	20.3	98.51
7	34.0	-	47.0	0.64	18.8	100.44

3. Disseminated mineralizations are developed in rhyodacites and in basalts. The disseminated mineralizations in rhyodacites under the massive orebody and around stockworks are the product of hydrothermal alteration and reflect the composition of massive and stockwork mineralizations.

Networks of sulfide veinlets up to a few millimetres across, are well developed in basalts overlying massive sulfide mineralizations. Two types of mineral associations are found in basalts; the first one pyrite, marcasite, sphalerite and baryte, and the second one pyrite, chalcopyrite, marcasite. Fine grained gold inclusions are occasionally found in both mineral associations. The presence of marcasite witness low pH values (<5) and the solubility of gold was controlled by the complexes $Au(HS)^{\circ}$. The study of fluid inclusions in baryte and chalcedony suggests the minerals were formed from Cl-rich fluids at about 250°C. The formation of fluids with 25 wt % equiv. NaCl without evaporite participation is a process, energetically very expensive, and result from boiling (Sinojmeri, 1990).

The abundance of oxygenous minerals as baryte, calcite, and quartz, indicate a high oxygen activity, while Fe-rich sphalerite witness a lower sulfur activity than in massive mineralizations. The abundant presence of marcasite suggest low pH values, low crystallisation temperature (<250°C) and equilibration temperatures lower than 160°C (Murowchick and Barnes, 1986).

CONCLUSIONS

Geological context in which Munella deposits is developed is characteristic to a back-arc spreading centre. The chemical composition of surrounding lavas confirm this tectonic setting. Modern sea-floor sulfide mineralizations related to submarine volcanism of bimodal suite back-arc basins are found at West Pacific Ocean (Fouquet et al., 1991; Taylor et al., 1990).

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The volcanic depression consequent of caldera forming in Mirdita basin becomes the locus of discharging hydrothermal fluids. By the way in high-permeability rhyodacites took place the water-rock interaction, and consequently the kaolinization and pyritization.

The forming temperature, calculated in massive stratiform mineralizations is about 250 to 350°C, and mineral associations suggest high sulfur activity, low oxygen activity and acid pH in the hydrothermal fluids.

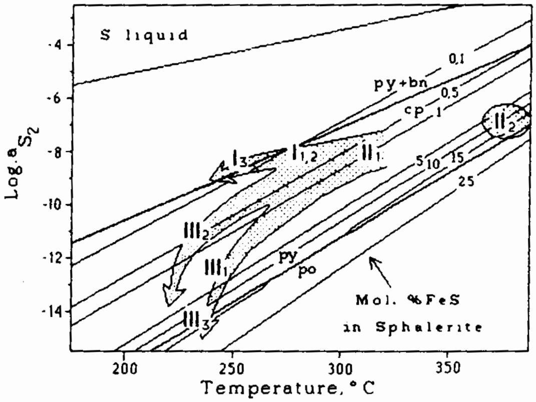

Fig. 4: Schematic evolution of Munella mineralizations projected on the Fe-Zn-S system at 1 bar compiled by Scott (1983).
po : pyrrhotite ; cp : chalcopyrite ; bn : bornite ; py : pyrite.
Bold lines : py/po and (py+bn)/cp equilibria.
I - Stratiform mineralizations
1 - Massive pyritic mineralization
2 - Massive pyrite-sphalerite-chalcopyritic mineralization
3 - Bornitic mineralization
II - Stockwork mineralizations
1 - Pyrite-chalcopyritic mineralization
2 - Arsenopyritic mineralization
III - Disseminated mineralizations
1 - Sphalerite-galena mineralization
2 - Pyrite-chalcopyrite mineralization
3 - Vessicular mineralization

The massive mineralization is developed in a black smokers field and is characterised by a roughly horizontal and vertical zonality delineated by ZnS and PbS concentration at the rim and on the top of massive orebody. The thin silicic bed overlaying discontinuously the stratiform orebody translates a break in volcanic activity before the generation of basaltic flows. The presence of fluorine and calcite besides zeolite, chlorite, epidote, quartz and sulfides in vesicle fillings indicate the presence of volatiles in basaltic flows, that make possible the vesiculation of submarine basalts even in deep (>1000m) marine conditions (Dudas, 1983).

The disseminated mineralization developed in overlaid basalts witness the continuity of hydrothermal activity. Fluid inclusions in baryte and chalcedony indicate boiling conditions and temperatures up to 250°c, while the abundant presence of marcasite suggests acid pH (<5).

The $\delta^{34}S$ values (+1.4 to +4.4 in pyrite, +3.5 to +4.8 in chalcopyrite and +3.3 to +5.3 in sphalerite) from different types of sulfide mineralizations

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similar to those found in Canyon Diablo Troilite indicate a weak miscibility of mineral forming fluids with sea-water (Ohmoto, 1986; Ohmoto and Rye, 1979). Similar values are found in fossil and modern sea-floor deposits. Consequently the H2S hydrothermal oxidations is madden loosing H_2 by the reaction:

$$2H_2S \rightarrow H_2S_2 + \uparrow H_2$$

The abundant H_2- content in hydrothermal solutions can explain the high-concentration of H^+, that make possible marcasite formation.

The arsenopyrite mineralization remains a problematic one in Munella deposit. It is noticeable that arsenopyrite is an uncommon mineral in a submarine environment.

The development of sulfide mineralization in Munella deposit is projected in log aS_2-Temperature diagram (fig.4).

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