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On the occurence of iron-magnesium carbonates in hydrothermal systems

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INTRODUCTION

Mount Amiata is in the south of Tuscan region (Italy). It is an extinct volcano that hosts in its roots one of the two geothermal systems exploited in Italy, the other one being Larderello (Pisa province). The Amiata geothermal system is liquid dominated with two reservoirs at different depths, in different temperature conditions: 150-220° the shallower and 300-350°C the deepest. Geothermal wells are drilled up to 4500m TVD (Barelli et al., 2010).

At present, geothermal production comes only from the second reservoir, the Farma formation (Upper Palaeozoic – Middle Trias), that comprises the Verrucano Group (Triassic metapelites, metasandstones and metaconglomerates), graphitic phyllites and metasandstones, with dolostones and late Permian limestones. The geothermal fluid is near neutral in reservoir conditions, can be classified as a sodium chloride facies with a CO_2 content of 2% w/w on total fluid (Minissale et al., 1997).

The deep reservoir of Mount Amiata Geothermal system (Italy) is made of schists (Barelli et al., 2010) with a content of Ca ranging 7-1.5%, Mg 0.8-3%, Fe 7-9%, and a CO_2 content in the geothermal fluid of about 2% w/w. In this work, using XRD, XRF and SEM analyses, we recognize an hydrothermal alteration consisting of a mineral assemblage of quartz, muscovite, clinochlore, Illite, albite, paragonite, carbonates (dolomite, ankerite, ferroan magnesite and magnesian siderite), chlorite (Mg, Fe) and pyrite, where CO_2 plays a role in alteration products of the upper portion of the reservoir.

METHODS

XRF analyses with X-rays Rigaku ZSX Primus II
XRD powder diffraction with D8 "Da Vinci" (Bruker) diffractometer
SEM EDS analysis with ZEISS EVO MA15 with EDS-EBSD OXFORD AZTEC 5.0 PS 1 (EDS Oxford ULTIM MAX 40mm2 e EBSD Oxford Symmetry)

AMIATA GEOTHERMAL FIELD DEEP CORES

The drilled core samples come from increasing depth: at the top (1114m), middle (1340m) and bottom (2872m) of the deep geothermal reservoir at Amiata geothermal field.
Within the 1114m depth sample, we are able to clearly identify many ankerites domain (fig. 1, 2, 3 and 4, tab. 1).



Figure 1 1114m depth sample

Table 1 sample 1114m depth, chemical composition by XRF

		1114
SiO2	mass%	57.869
Al2O3	mass%	17.812
Fe2O3	mass%	7.238
MnO	mass%	0.091
MgO	mass%	0.88
CaO	mass%	1.722
Na2O	mass%	1.233
K2O	mass%	2.874
TiO2	mass%	0.795
P2O5	mass%	0.138
Ba	ppm	447
Cr	ppm	120
Ni	ppm	27
Rb	ppm	124
Sr	ppm	103
Y	ppm	37
Zn	ppm	70
Zr	ppm	324

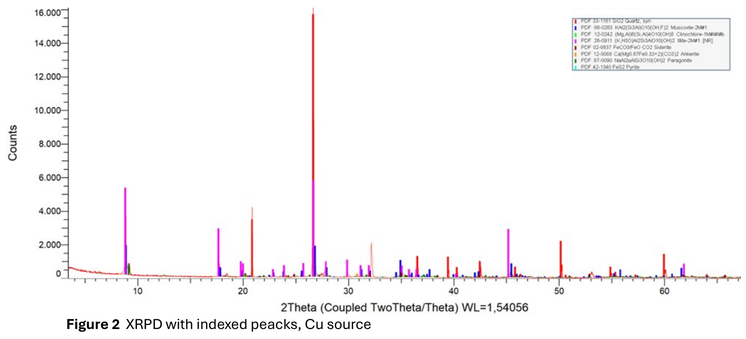


Figure 2 XRPD with indexed peacks, Cu source

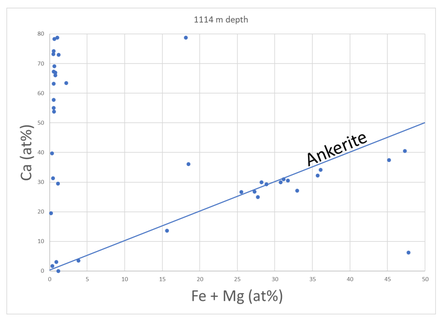


Figure 3 SEM point analysis of ankerite and host rocks

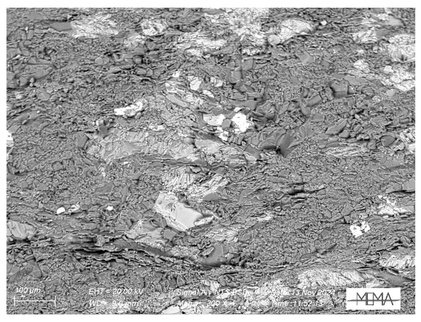


Figure 4 Backscattered electron image

The 1340m deep sample is dominated by the presence of pyrite and pyrrhotite, with trace of carbonation (fig. 5,6,7,8 and tab. 2).



Figure 5 1340m depth sample

Table 2 sample 1340m depth, chemical composition by XRF

		1340
SiO2	mass%	54.578
Al2O3	mass%	16.468
Fe2O3	mass%	8.701
MnO	mass%	0.107
MgO	mass%	1.459
CaO	mass%	1.578
Na2O	mass%	0.693
K2O	mass%	3.114
TiO2	mass%	1.047
P2O5	mass%	0.18
Ba	ppm	540
Cr	ppm	191
Ni	ppm	33
Rb	ppm	139
Sr	ppm	75
Y	ppm	37
Zn	ppm	83
Zr	ppm	239

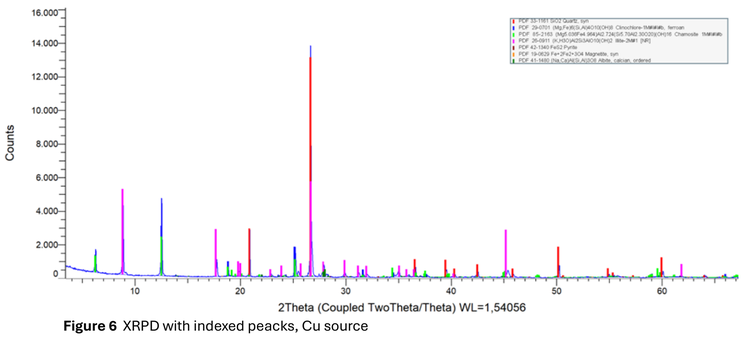


Figure 6 XRPD with indexed peacks, Cu source

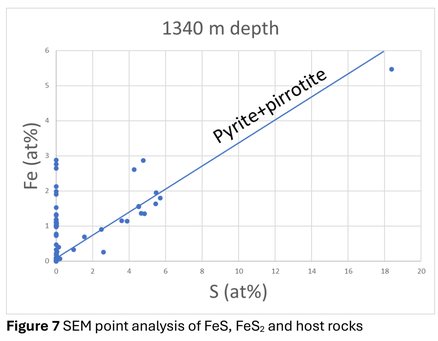


Figure 7 SEM point analysis of FeS, FeS_2 and host rocks

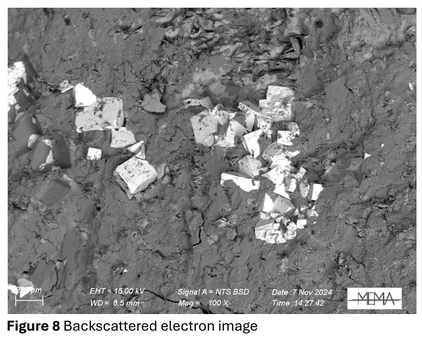


Figure 8 Backscattered electron image

The 2872m deep sample show intact chlorites, with the presence of calcite, dolomite and plagioclase, while the other samples have only traces of albite (fig. 9, 10, 11, 12 and tab. 3).



Figure 9 2872m depth sample

Table 3 sample 2872m depth, chemical composition by XRF

		2872
SiO2	mass%	49.926
Al2O3	mass%	10.616
Fe2O3	mass%	8.72
MnO	mass%	0.222
MgO	mass%	3.092
CaO	mass%	7.078
Na2O	mass%	0.599
K2O	mass%	2.168
TiO2	mass%	0.544
P2O5	mass%	0.12
Ba	ppm	315
Cr	ppm	133
Ni	ppm	27
Rb	ppm	89
Sr	ppm	191
Y	ppm	23
Zn	ppm	68
Zr	ppm	120

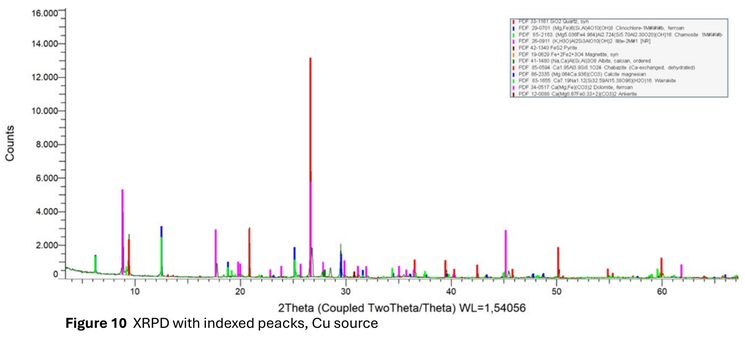


Figure 10 XRPD with indexed peacks, Cu source

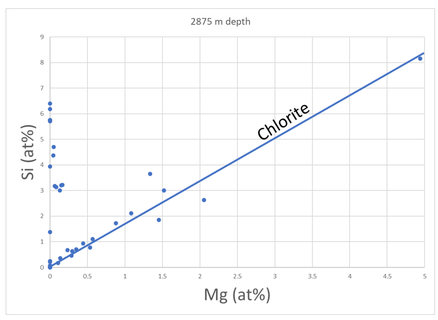


Figure 11 SEM point analysis of Chlorite and host rocks

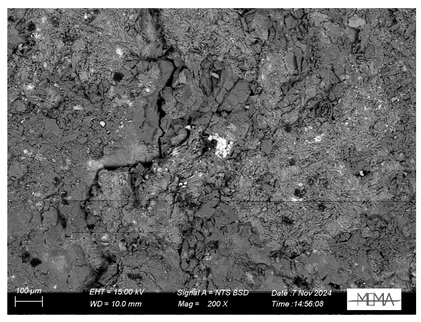


Figure 12 Backscattered electron image

DISCUSSION AND CONCLUSION

In the cored material from deep geothermal reservoir, hydrothermal and metamorphic chlorites are replaced by magnesian siderite and other carbonates, mainly siderite and ankerite, after the CO_2 – driven alteration given that iron is the dominant cation.

The Mg, Fe chlorites show an hydrothermal alteration process toward carbonates. Plagioclase are found only as traces, mostly albite, and rarely k-feldspar.

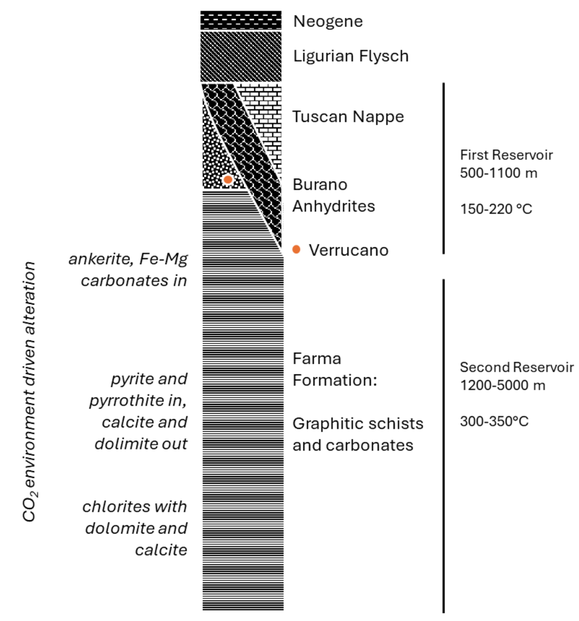
Of the three samples, the 2872 m depth is located in the deep reservoir and shows a large compositional range of Fe-Mg chlorites, illite and dolomite; going upward, at 1340 m, dolomite disappears and a large amount of pyrite can be found. Finally, in the 1114 m depth sample, located at reservoir top, a relevant amount of ankerite is found with quartz and muscovite.

Thus, a progressive increasing alteration from bottom to top of the deep reservoir seems to be present:

- chlorites with dolomite and calcite are dominant;
- iron from chlorites forms pyrite and pyrrothite in the midlevel, where calcite and dolomite disappear;
- in the top reservoir precipitation of ankerite and other carbonates in the solid solution siderite-magnesite with a reduction of pyrite amounts occurs.

We hypothesize that the primary $(Mg,Fe)CO_3$ solids undergo a dissolution–recrystallization process driven by two forces: the preferential partition of Fe(II) toward the $(Mg,Fe)CO_3$ solid solution and the oxidation of aqueous Fe(II) species to form Fe(III)-bearing solids; limited amount of magnetite are found in the mid and top level samples. Under limited oxygen supply, the oxidation of Fe(II) is a slow process, particularly in the presence of chlorine and inorganic carbon. In absence of oxidation, the initial solid solution should become increasingly Fe-rich as the supersaturation decreases and the system approaches equilibrium.

Concluding, the progressive alteration in the second reservoir at Amiata is likely related to the CO_2 (and H_2S) circulation in the upper part of the reservoir providing an increasingly alteration grade of the original host rocks.



Acknowledgements
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References

Barelli, A., Ceccarelli, A., Dini, I., Fiordelisi, A., Giorgi, N., Lovari, F., Romagnoli, P. (2010): A review of Mount Amiata geothermal system (Italy). Proceedings World Geothermal Congress.
Minissale, A., Magro, G., Vaselli, O., Verrucchi, C., Perticone, I. (1997): Geochemistry of water and gas discharges from the Mt. Amiata silicic complex and surrounding areas (central Italy). Journal of Vulcanology and Geothermal Research 79; pp 223-251.