Magnesite Veins from Ultramafic Massif of Beni Bousera (Internal Rif, Morocco)

Zaineb Hajjar[1*], Amina Wafik[1], Marc Constantin[2]

1- DLGR Laboratory (associated Unit to CNRST, URAC 43), Department of Geology, Faculty of Sciences Semlalia, Cadi Ayyad University, Marrakech, Morocco.

2- Department of Geology and Genie Geologic, Faculty of Science, University of Laval, Quebec, Canada.

* Corresponding Author: zaineb.hajjar@edu.uca.ma

Abstract

The magnesite veins are located in the Beni Bousera massif (internal Rif, Morocco), especially in Tafidest region about 103 km SW of the Tetouan city. They are hosted by perridotite and serpentinite formations. The aim of this study is to clarify the origin of this type of orre genesis. Geochimical study of magnesite ore reveals that is a cryptocrystalline magnesite, formed by the CO_2-rich hydrothermal fluids filling factures. These solutions were brought from serpentinized area following tectonic fractures. The magnesite precipitation is due to the decrease of partial pressure of supersaturated carbon dioxide hydrothermal fluids, most likely at temperature below 300 ºC.

Keywords: Magnesite, Cryptocristalline, Peridotite, Beni Bousera, Rif, Morocco.

1- Introduction

Magnesite ($MgCO_3$) is formed in a sedimentary and/or magmatic environment (Pohl 1990; Abou-Jaber and Kimberley 1992; Schroll. 2002; Kadir *et al*., 2013). At the ultramafic rocks the magnesite results from hydrothermal and supergene alteration of ultramafic or hosted sediment rocks by a carbonic fluid (Griffis 1972; Dulski and Morteani. 1989). The ultramafic rocks are metamorphosed by a carbonic fluids which can gives rise to (i) a typical association of magnesite with serpentine, talc, quartz, enstatite or anthophyllite like in Bou Azzer inlier (Anti Atlas, Morocco; Bhilisse *et al*., 2014), or (ii) only a magnesite veins croscatting serpentinized peridotites as from Mount El-Rubshi and Mount El Maiyit at Egypt (Ghoneim *et al*. 2003) and in Eskişehir area in Turkey (Kahya and Kuşcu 2014). So a stage of serpentinisation of the ultramafic rocks is essential before the carbonation and the formation of magnesite (Zedef *et al*. 2000;

Schulze *et al*. 2004; Teir *et al*. 2007), following this reaction:

$$Mg_3Si_2O_5(OH)_{4(s)} + 3CO_{2(g)} + 2H_2O \rightarrow 3MgCO_3 + 2H_4SiO_4$$
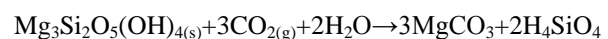
Serpentine   carbonic fluid   magnesite

The experimental study of carbonatation phenomenon indicates that the transformation of serpentinized peridotites to magnesite took place after a basic and carbonic fluid is rich in NaCl, at fluid temperature around 200 °C and CO_2 pressure between 120 and 180 bar) (Teir *et al*., 2007; Hovelman *et al*., 2011).

The model of magnesite formation can be distinguished of listwinite formation (Aftabi and Zarrinkoub, 2013) by the mechanism of formation and fluids involved. The listwinite formation occurs in the late stage of ophiolite serpentinization (Zouita, 1986; Bhilisse *et al*., 2014), whereas magnesite formation is due to fluid circulation in ultramafic rock. These fluids may have different origins: atmospheric, metamorphic, sedimentary or magmatic origin (Kahya and Kuşcu, 2014). The formation of

magnesite in ultramafic rock involves two models: (i) the first process is the filling of fractures by precipitation of magnesite from a magnesium-rich carbonic fluid due to the pressure decrease (Abu Jaber and Kimberley 1992); (ii) the second process is metasomatic replacement 'in situ' of serpentinized peridotite by magnesite and silica under the supergene and/or hydrothermal conditions. In this case, silica can be transported by the fluid (Griffis. 1972 ; Dabitzias, 1980; Pohl, 1990).

The Tafidest magnesite veins were studied for the first time by Agard *et al.* (1959). However, until now, there are no academic or technical Studies are established. In this contribution, we attempt to address these problems in our study of the magnesite veins in the Beni-Bousera massif. We provide (i) a detailed geological features and petrographic description of the peridotite and serpentintes, (ii) in detail on the relationship among paridotites, serpentintes and magnesite. Finally we summarize the formation conditions of the magnesite veins.

2- Geological setting

The ultramafic massif of Beni Bousera outcrops as a NW-SE, 70 km² peridotite body between Bouh'med and Amter Villages. It belongs to the alpine Betic-Rif orogeny (Fig. 1), and exactly to the lower Sebtides unit (internal zones of Rif), where it constitutes a cristallophyllien terrain surrounded by metamorphic series (Kinzigites, Filali and Ghomaride unit).
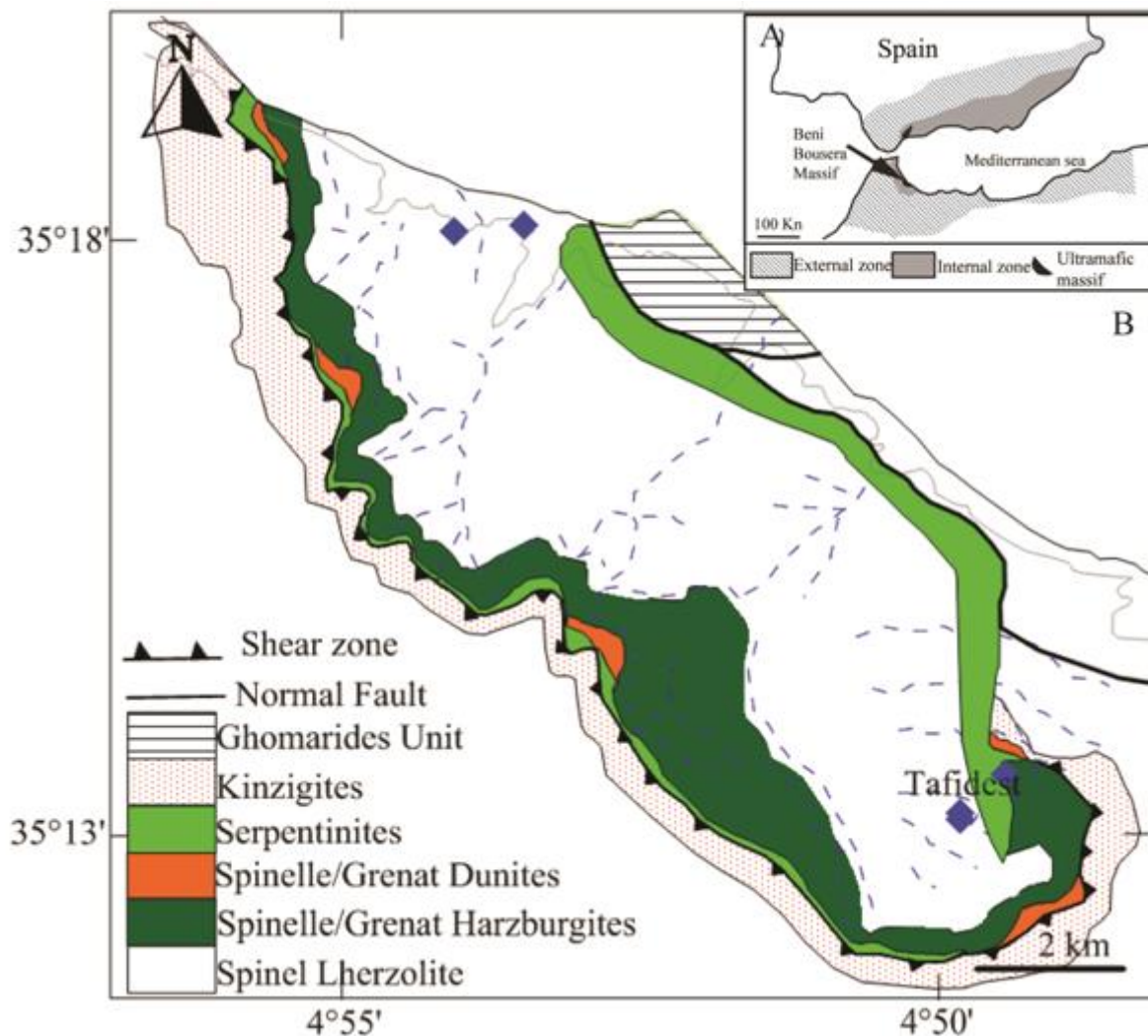

Figure 1) A: Simplified map of the Rif-Betic Cordillera; B: Geological map of Beni Bousera massif (Afiri et al. 2011), Blue square: sample location.

The massif is mainly composed of peridotites and minor pyroxenites layers (Kornprobst, 1974; Gysi, 2007, 2011; Frets *et al*., 2011). In detail, the massif presents a petrographic zonation from Spinel lherzolites in the bottom, towards spinel/garnet dunites and harzburgites in the top (Kornprobst, 1974, Reuber *et al*., 1982; Fig.1).

The exhumation of sub-continental peridotites of Beni Bousera massif in the Miocene occurred into two stages, toward shear zone and normal fault bording the massif (Michard *et al*., 1983; Saddiqi, 1995; Chalouan *et al*., 1995; Afiri *et al*. 2011). These anormal contacts between peridotites and upper crust are underlined by serpentinized area with thickness ranging from 100 to 500 m (Hajjar *et al*., 2015).

3- Analytical method

To establish any structural, petrographic, and geochemical relationships among magnesite, peridotites and serpentinites hosted rocks in the ultramafic massif of Beni Bousera, we sampled in magnesite structures and their host peridotites and serpentinites in both Tafidest (SE of the massif) and serpentinized area in the NE of massif near Sidi Yahya Aaraben Fault.

In total, ten samples were subject to a petrographic study by optical microscope in DLGR laboratory (in the department of Geology, Faculty of Sciences Semlalia, Marrakech). In addition, these samples were subjected to physical characterization by X-Ray technique with Cu anode (in the department of Chemistry at the Faculty of Science Semlalia (Marrakech), and by Raman spectroscopy on magnesite. The Raman spectra were recorded at Lille 1 University, with a LabRam HR800 Jobin-Yvon- microspectrometer equipped with 1800 g/mm gratings and using 532.28 nm (green) laser excitation. Acquisition time span varied from 10 to 20 s during five accumulating cycles. The spectral regions investigated is from 200 to 1200 cm-1.

Geochemical characterization of whole rock samples reported in Table 1, represents seven magnesites analyzed by ICP-MA and ICP-BF in laboratory "Stewart geochimical and Essay" in Russia and by ICP-AES (jobinYvon ULTMA 2c) with mono- and polychromators in Reminex research center and laboratory (Managem group). These data were used firstly to determine the type of magnesite and secondly to establish a global mass balance of the transformation of peridotites to serpentinites so as to evaluate the mobility of MgO, during serpentinization.

Two samples of peridotites and serpentinites from Beni Bousera massif (Table. 2) were analyzed in Reminex research center and laboratory (Managem group) using ICP-AES (jobin Yvon ULTMA 2C) with mono-and polychromators. These data was used to study the principal of mass balance. This later is based on the immobility of chemical elements during metasomatism (Gresens, 1967; Grant. 1986; Kranidiotis and Maclean, 1987). In this paper only the method of Grant (1986) is used.

4- Results

4.1- Structure

Our results show that the normal fault of Sidi Yahya Aaraben, parallel to fault system used at the opening of the Alboran Sea in the Burdigalian-Tortonian and exhumation of Beni Bousera massif is highlighted by the presence of a serpentinites area (500m). These serpentinites resulted from hydrothermal alteration of spinal lherzolite. These serpentinites are affected by magnesite filling the N100 to N130° and N50 to N70° trending fractures (Fig. 2).

A strong carbonatation is observed at the Tafidest region, 500 m from the left side of the Amter River, and 4- 5 km from its embouchure. In this region we observe the large monomineral structure of magnesite, with the same direction as the N100°-trending fractures which affect the serpentinized zone (Fig. 3). The Tafidest zone is

a half-graben parallel to the normal fault used for the opening of the Alboran Sea. The magnesite veins range in length from 1 to 20 m, and in thickness from a few decimeters to a few meters. These structures contain fragments of peridotite slightly serpentinized, angular to sub-angular, with variable size. The microtextural study reveals the presence of magnesite veins crosscutting the peridotites, however there are no signs of replacement of peridotites by magnesite.
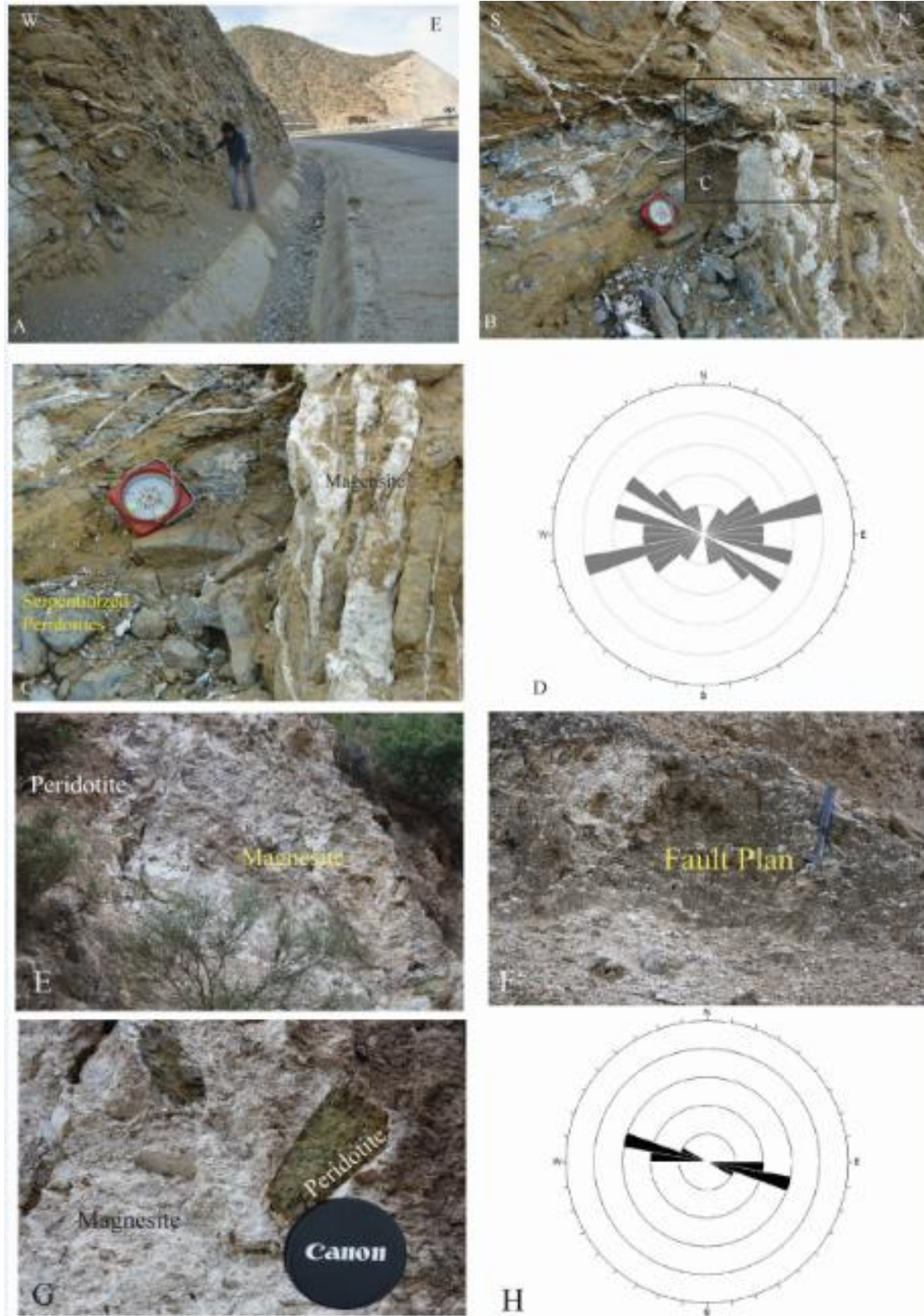

Figure 2) Magnesite vein in serpentinized area from Beni Bousera massif: A to C: magnesite vein croscatting serpentinized peridotites in serpentinized area from Beni Bousera massif; D: stereographic

representation of magnesite veins in serpentinized area from Beni Bousera massif; E: magnesite vein in peridotite hosted from Tafidest region; F: fault plan affecting magnesite structure in Tafidest; G: fragments of peridotites in magnesite structure in Tafidest; H: stereographic representation of magnesite veins structure.
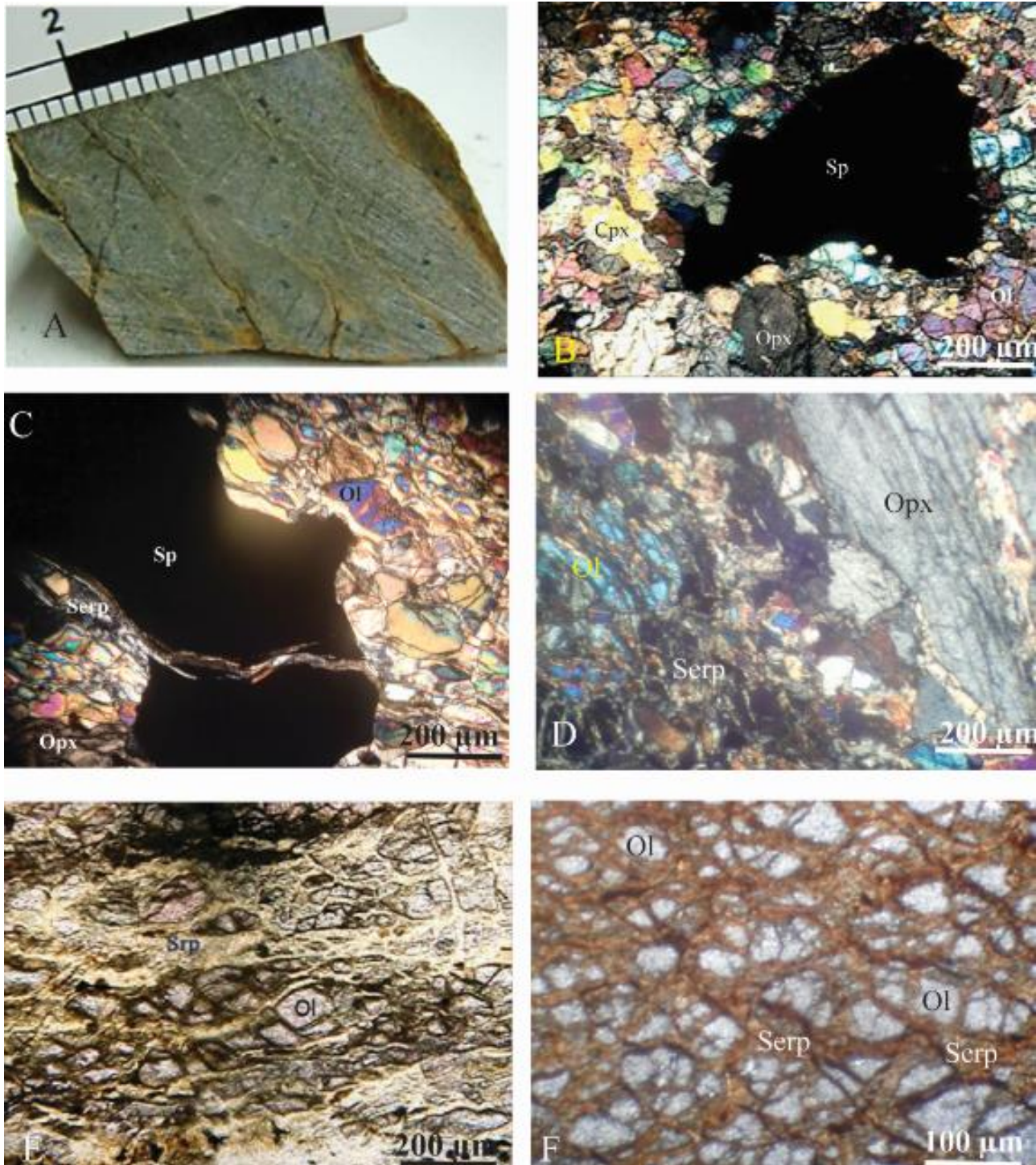

Figure 3) Photographies and microphotographies of fresh peridotites and serpentinized peridotites in serpentinized area from Beni Bousera massif; A: macroscopic sample of fresh peridotite; B: Spinel lherzolite Porphyroblastic texture (LPA); C: serpentinization affecting spinel lherzolite facies, where serpentine affect olivine, and fractures (LPA); D: serpentinized olivine, with intact pyroxene in serpentinites in the NE of massif (LPA); E & F: massifmesh texture of serpentine with olivine in the heart (LP).

4.2- Petrographic study

4.2.1- Peridotites

Petrographically, the Tafidest peridotites can be classified as spinel-lherzolite. They contain olivine (50-65%), orthopyroxene (15-25%),

clinopyroxyne (15-20%), and spinel (5-15%). These peridotites have a porphyritic texture (Fig. 4). We note the presence of two assemblages: (i) a primary paragenesis formed by porphyroclast of olivine (6-5 mm), orthopyroxene (4-6 mm), and clinopyroxene (2.5 mm).
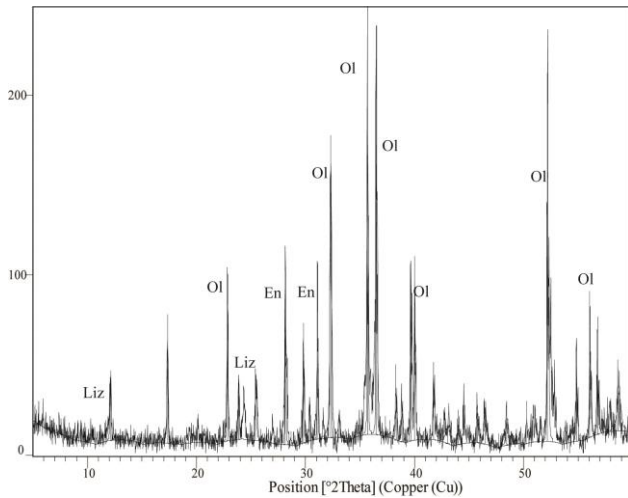

Figure 4) X-Ray diagram from serpentinzed peridotite.

The orthopyroxene porphyroclasts contain fines exsolutions of clinopyroxene. The aluminous

phase is represented by brownish spinel (1-5 mm); and (ii) a secondary paragenesis formed by serpentine which outlines weak plans and fractures affecting peridotite. Indeed, serpentine filled cracks of olivine, joints minerals and fractures. Whole rock characterization by X-Ray analyses indicates that the serpentine replacing the peridotites is a lizardite type.

4.2.2- Serpentinized Peridotites (Serpentinites)

Peridotites, especially spinel lherzolite, are strongly serpentinized in the NE contact of the massif; this stage of serpentinization is marked by the development of serpentine in the expense of olivine. The Orthopyroxenes and clinopyroxenes remain trace of serpentinization in the cleavage plans without complete transformation of orthopyroxène to serpentine or talc (Fig. 5). Like serpentine from the peridotites rocks, the serpentine from serpentinite rocks is a lizardite type.

Tabel 1) whole rocks analysis of magnesite veins in Beni Bousera by ICP-MA and ICP-BF in laboratory "Stewart geochimical and Essay" in Russia, and (*) analyzed by ICP-AES in Reminex laboratory (Managem Group).

	Tafidest					Serpentinized area	
	Mag 1	Mag 2	Mag 3	Mag 4 *	Mag 5 *	Mag 6	Mag 7 *
Al_2O_3	0,1	0,1	0,3	0,5	0,5	0,7	0,6
CaO	0,9	1,2	0,9	1,0	1,0	1,2	0,7
FeO	0,3	0,3	0,4	0,1	0,1	0,6	0,7
MgO	45,2	42,4	45,4	43,2	41,1	40,8	41,1
MnO	0,0	0,0	0,0	0.01	0,0	0,0	0,0
Na_2O	0,0	0,0	0,0	0,5	0,5	0,0	0,5
SiO_2	7,8	10,0	2,2	5,3	9,7	13,0	10,1
SrO	0,0	0,0	0,0	0,0	0,0	0,0	0,0
CO_2	45,8	46,0	50,3	49,3	47,1	43,5	46,4
Total	**100,0**	**100,0**	**99,6**	**100,0**	**100,0**	**99,8**	**100,0**
Calcite ($CaCO_3$)	0,70	1,08	0,71	0,86	0,84	1,04	0,56
Siderite ($FeCO_3$	0,42	0,63	0,41	0,12	0,18	0,84	0,98
Rhodochrosite ($MnCO_3$)	0,01	0,00	0,00	0,01	0,01	0,01	0,01
Magnesite ($MgCO_3$)	98,87	98,29	98,89	99,01	98,96	98,12	98,45
Strontianite ($SrCO_3$)	0,00	0,00	0,00	0,00	0,00	0,00	0,00
TOTAL	**100,00**	**100,00**	**100,00**	**100,00**	**100,00**	**100,00**	**100,00**
	Elements traces						
Cr	26,0	20,0	30,0	58,0	385,0	50,0	246,0
Ni	190,0	200,0	100,0	73,0	408,0	500,0	309,0

4.2.3- Magnesite

Structure of magnesite in Tafidest and serpentinized area mainly consist of magnesite,

that appears in white color with porcelain luster; and has massive, or cauliflower texture. It occurs as microcrystalline and interlocked aggregates forming granular and mosaic textures. In the serpentinzed area, the vein of magnesite cut serpentine, indicating that formation of magnesite is posterior to serpentinization of peridotites. The X-Ray technique used for determination of characteristics of magnesite; confirms that magnesite is the unique mineral constituting magnesite ore structures at Tafidest. The Raman spectra of magnesite proves that this one has two characteristic peaks in 330 and 1099 cm^{-1}.
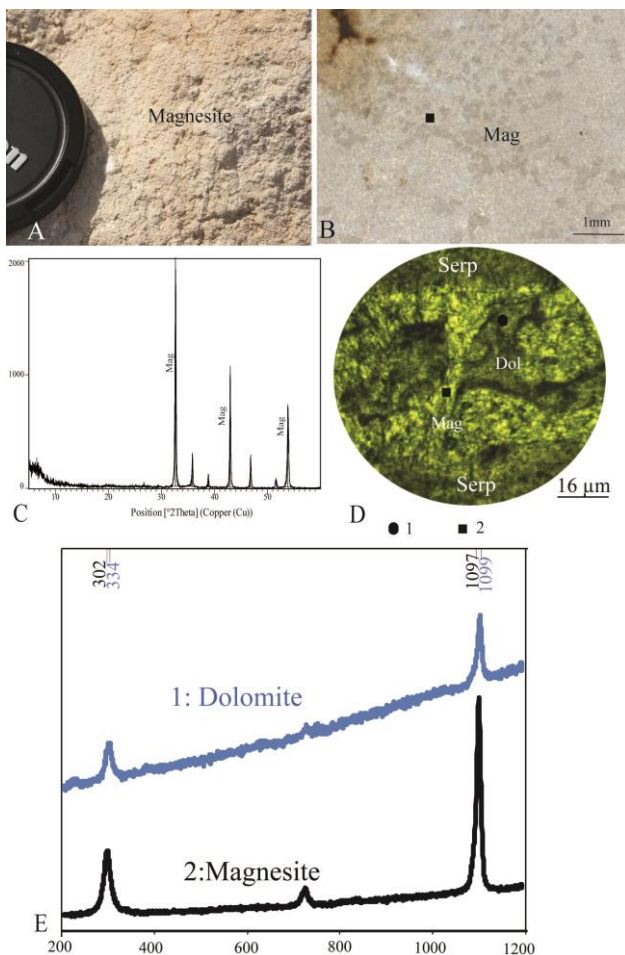

Figure 5) A: macroscopic sample of magnesite; B photomicrograph of magnesite in polarized light; C: X -Ray diagram from magnesite; D: magnesite vein coruscating serpentine in serpentinized peridotites in the NE of the massif; E: Raman Shift of magnesite from serpentinized area; (Black Square in B and D is a magnesite; and Black circle is a dolomite.

4.3- Geochemistry

4.3.1- Magnesite

The average structural formulae of magnesite in Beni Bousera veins is $(Mg_{0.989}, Ca_{0.007}, Fe_{0.004})CO_3$ and they reveal the substitution of magnesium by mainly calcium and ferrous iron. The Cr and Ni enrichment in magnesite is due to mixture of other trace mineral phases derived from parent ultramafic rocks. The projection of studied magnesite data in diagram with those of cryptocrystalline, sparry and evaporate magnesite, as shown in Figure 6, indicates that the studied magnesite belongs to cryptocrystalline type. Moreover, the Cr/Mg, Mn/Mg, Fe/Mg ratio in the magnesite structures, compared to those of the metamorphic and cryptocrystalline magnesite from Rösler and Lange (1972), indicate that the discussed magnesite approaches rather to the context of the cryptocrystalline magnesite than the one related to metamorphic processes (Table 3).

Tabel 2) Whole rocks analysis of peridotite and serpentinites in Beni Bousera massif by ICP- AES in Reminex Research center in Morocco.

	serpentinized peridotite	peridotite
Al2O3 (%)	4,07	3,60
CaO (%)	1,22	3,51
Fe2O3 (%)	22,97	8,70
K2O (%)	0,01	0,01
MgO (%)	23,77	37,06
MnO (%)	0,87	0,13
NaO (%)	0,49	0,53
P2O5 (%)	0,23	0,01
SiO2 (%)	44,29	43,88
TiO2 (%)	0,12	0,14
As (ppm)	38,00	40,00
B (ppm)	255,00	68,00
Ba (ppm)	14,00	1,00
Be (ppm)	0,10	0,10
Bi (ppm)	10,00	10,00
Co (ppm)	232,00	149,00
Cr (ppm)	48,00	1946,00
Cu (ppm)	1,00	29,00
Nb (ppm)	0,50	5,00
Ni (ppm)	51,00	2181,00
Pb (ppm)	29,00	13,00
S (%)	0,07	0,02
Sb (ppm)	16,00	16,00
Sn (ppm)	80,00	42,00
Sr (ppm)	38,00	16,00
Y (ppm)	14,00	9,00
Zn (ppm)	78,00	47,00

4.3.2- Mass balance of transformation of peridotite to serpentinites

Characterization of gain and loss elements during the transformation of periodotite to serpentinized peridotite was evaluated by Grant (1986) method (Fig. 7); through this study we can distinguish three groups of elements:
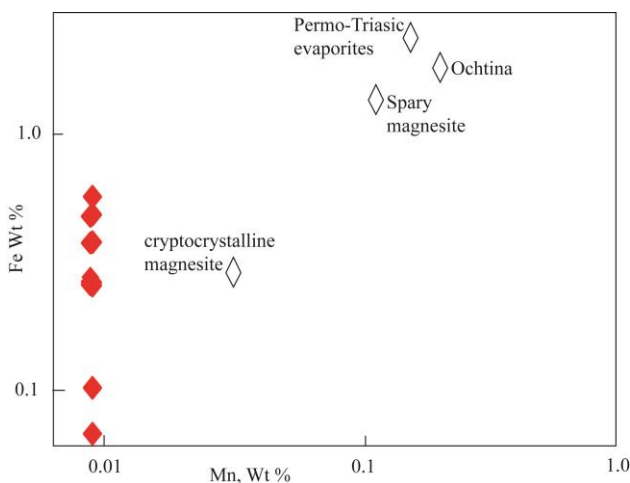

Figure 6) The Fe vs Mn contents in the studied magnesites, compared to cryptocrystalline magnesite (Rösler and Lange, 1972) and sparry

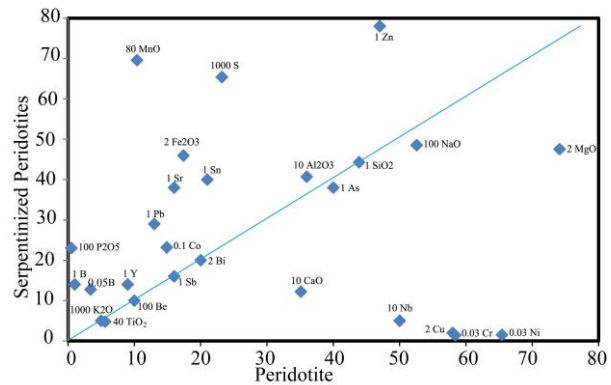magnesite (Martiny and Rojković, 1977) from various localities in the area of Slovakia.

Figure 7) Diagram of mass balance from peridotite to serpentinite (method of Grant, 1986)

- Group of Al_2O_3, K_2O, Na_2O, SiO_2, TiO_2, As, and Sb: these elements constitute an isochronous and may be considered immobile during the transformation of peridotite to serpentinite.
- Group of CaO, MgO, Cr, Cu, Nb, and Ni below the isochronous, suggesting a strong leaching of these elements, mainly MgO.
- Group of Fe_2O_3, MnO, P_2O_5, S, B, Ba, Co, Pb, Sn, Sr, Y, and Zn: in the top of the isochronous, indicating an enrichment of these elements in serpentinized peridotite.

Thus, we can suggest that the serpentinization of peridotite has provided the magnesium concentration, which served to the formation of magnesite.

5- Discussion

The serpentinization degree increases toward the Sidi Yahya Aaraben fault, which is marked by development of serpentine (lizardite) in the weak plans. The percentage of serpentine in these rocks is up to (60-80%). That reveals the highest degree of serpentinization.

When discussing the genesis of Beni Bousera magnesite veins, the following observations must be considered:

Pure magnesite veins occur within both peridotites and serpentinites rocks.

- Magnesite is predominantly cryptocrystalline in texture.

- Talc is absent in magnesite veins whereas silicate minerals are present in some magnesite veins in the serpentinites area.

- Rare angular to subangular fragments of host-rocks are encountered within magnesite veins, in Tafidest; however, they are observed restrictedly in margins, involving fracture filling.

- Spinel which is common in the host-rocks occurs in magnesite veins only within the host rock fragments.

The above mentioned results of field and laboratory investigations (petrological and geochemical features), may unravel the following origin of magnesite:

- - The cryptocrystalline texture of magnesite indicates that the adding of the CO_2 rich fluids must have been fairly rapid causing the abundant precipitation of magnesite from solutions under near-surface conditions (Abu-Jaber and Kimberley, 1992) at temperature below 300 °C as indicated by absence of talc in magnesite veins (Johannes, 1970).

- The presence of angular fragments of host-rocks and no other relics of spinel in magnesite veins suggest that magnesite was not formed by metasomatism of serpentinites rocks. Nevertheless, it precipitated from ascending solutions rich in Mg^{2+} (Dulski and Morteani 1989; Pohl, 1990). This is also confirmed by the comparison of Cr/Mg, Fe/Mg, and Mn/Mg ratios with those reported by Rosler and Lange. (1972).

- Most likely the serpentinization of peridotite form serpentinized peridotite and serpentinites in the NE of massif was the source of magnesium in the vein filling. This is also confirmed by the leaching of MgO during serpentinization event.

Tabel 3) selected element ratios for magnesite veins.

| | This study | | | | | | | Rösler, Lange, 1972 | |
| | Tafidest | | | | | Serpentinized area | | | |
	Mag 1	Mag 2	Mag 3	Mag 4 *	Mag 5 *	Mag 6	Mag 7 *	cryptocristaline	Metamorphic
Cr/Mg	1E-04	8E-05	1E-04	2E-04	2E-03	2E-04	1E-03	1E-04	2E-03
Fe/Mg	1E-02	1E-02	9E-03	3E-03	2E-02	2E-02	4E-03	2E-04	
Mn/Mg	3E-04	3E-04	3E-04	3E-04	3E-04	3E-04	3E-04	4E-04	2E-03
Ni/Mg	7E-04	8E-04	4E-04	3E-04	2E-03	2E-03	1E-03	2E-04	2E-04

6- Conclusion

The precipitation of magnesite in the Beni Bousera massif probably occurred after serpentinization of the peridotites. The temperature of magnesite precipitation was less than 300°C as indicated by cryptocrystalline textures and the absence of talc in magnesite veins. Indeed, the serpentinization yielded significant amounts of available magnesium; but the suggestion of carbon source needs further confirmation, e.g. from the studies of stable carbon isotopes.

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