FAULTING, FLUID-ROCK INTERACTION AND HYDROTHERMAL MINERALISATION IN ULTRAMAFIC ROCKS (VOLTRI MASSIF, LIGURIAN ALPS)

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INTRODUCTION

Faults are complex and compound structures found in many geological settings in the upper crust. They are in general mechanically weak, prone to be reactivated during stress build-up, though with rare documented exceptions (Tavernelli, 1997). Faults systems control a wide range of crustal processes. Although faults occupy only a small volume of the crust, they have a controlling influence on the crust's mechanical and fluid flow properties (Sibson, 1977; Sibson *et al.*, 1988; Sibson, 1992, 2000, 2001; Faulkner *et al.*, 2010). Fault zones are lithologically heterogeneous, anisotropic and discontinuous. Individual fault zones commonly show significant variation in complexity along strike or down dip, even over relatively short distances (Faulkner *et al.*, 2010). In particular, it is well known that the damage zones, consisting of subsidiary structures through relatively large volume of rock surrounding the fault core, are associated with fault initiation, propagation and termination as well as its long-term evolution (Faulkner *et al.*, 2010). Damage zones are regarded as a key factor in a variety of geologic fields, such as the deformation processes associated with faulting (Schulz & Evans, 1998, 2000; Wilson *et al.*, 2003), strain distribution and deformation history in a region, earthquake rupture propagation and related seismic hazards (Sibson *et al.*, 1988; Sibson, 2001; Choi *et al.*, 2016), and fluid permeability in the crust (Caine *et al.*, 2010; Faulkner *et al.*, 2010; Choi *et al.*, 2016). The last one is particularly important as it is used in practical applications to ground water (Lopez & Smith, 1995; Welch & Allen, 2014), hydrocarbon reservoirs and ore-deposits, and the underground storage of CO_2 (Matter & Kelemen, 2009; Dockrill & Shipton, 2010; Kelemen *et al.*, 2011; Van Noort *et al.*, 2013).

Hydrothermal/metasomatic alterations developed along faults are examples of fluid-rock interactions, and can control the location, emplacement, and evolution of economic mineral deposits and geothermal systems (Groves *et al.*, 2000; Sibson, 2001; Micklethwaite, 2009; Caine *et al.*, 2010; Garofalo *et al.*, 2014). These alterations are mainly related to the: lithology, fault scale, fault type, deformation style and history, fluid chemistry, and P-T metamorphic conditions (Reed, 1997; Caine *et al.*, 2010; Schandl & Gorton, 2012). Metasomatic processes, linked to the circulation of CO_2-rich fluids along faults into ultramafic rocks (*e.g.*, peridotites and serpentinites), are exothermal processes (Matter & Kelemen, 2009; Kelemen *et al.*, 2011; Van Noort *et al.*, 2013), which leads to carbonation of the host rocks. For example the hydrothermal/metasomatic process occurring along fault inside serpentinites produces variously carbonated rocks. Listvenites (quartz-carbonate-chromium mica association), quartz-carbonate rocks (silica- carbonate association), carbonate-rich rocks, serpentine- talc- (chlorite)- carbonate rocks, talc-carbonate rocks and talc-silica-carbonate rocks are examples of products of hydrothermal/metasomatic alteration (Hansen *et al.*, 2005; Akbulut *et al*., 2006; Ulrich *et al.*, 2010; Van Noort *et al.*, 2013). These kind of carbonated rocks, and in particular listvenites are genetically linked to gold mineralisation. Moreover, particularly in the last decades, exhumed fault zones represent an engaging geological topic, since they may offer, among others, insights into earthquake- related deformational processes, the main issue being the distinction between seismic and aseismic slip (Boutareaud *et al.*, 2008; Smith *et al.*, 2011; Vigano *et al.*, 2011; Fondriest *et al*., 2012; Rowe & Griffith, 2015; Viti *et al.*, 2016; Chen *et al.*, 2017). The aim of this PhD thesis was to study reverse faults within intensely carbonated meta- lherzolites, with related gold mineralisations, from the Voltri Massif and, in particular, within the Lavagnina Lakes area, in the Gorzente Valley. Moreover these faults display meso- and micro-structures possibly related to fault seismicity. In particular the focus of this work was the study of peculiar dolomitic spherulites that are exceptionally large, have round shape and concentric texture. These structures associated with microtextures related to recrystallization over silica gel, carbonates nano-grains along the slip surfaces and carbonate mirror-like surfaces suggest that these spherulites should record cycles of seismic slips.

GEOLOGICAL BACKGROUND

The investigated area (Fig. 1) is the Lavagnina Lakes zone (GPS coordinates of the centre of the area 44.600685° N, 8.784286° E; WGS84), located near Casaleggio Boiro (Alessandria, Italy); at the North-Eastern boundary of the Voltri Unit (Ligurian Western Alps) and at the border with the Tertiary Piedmont Basin.

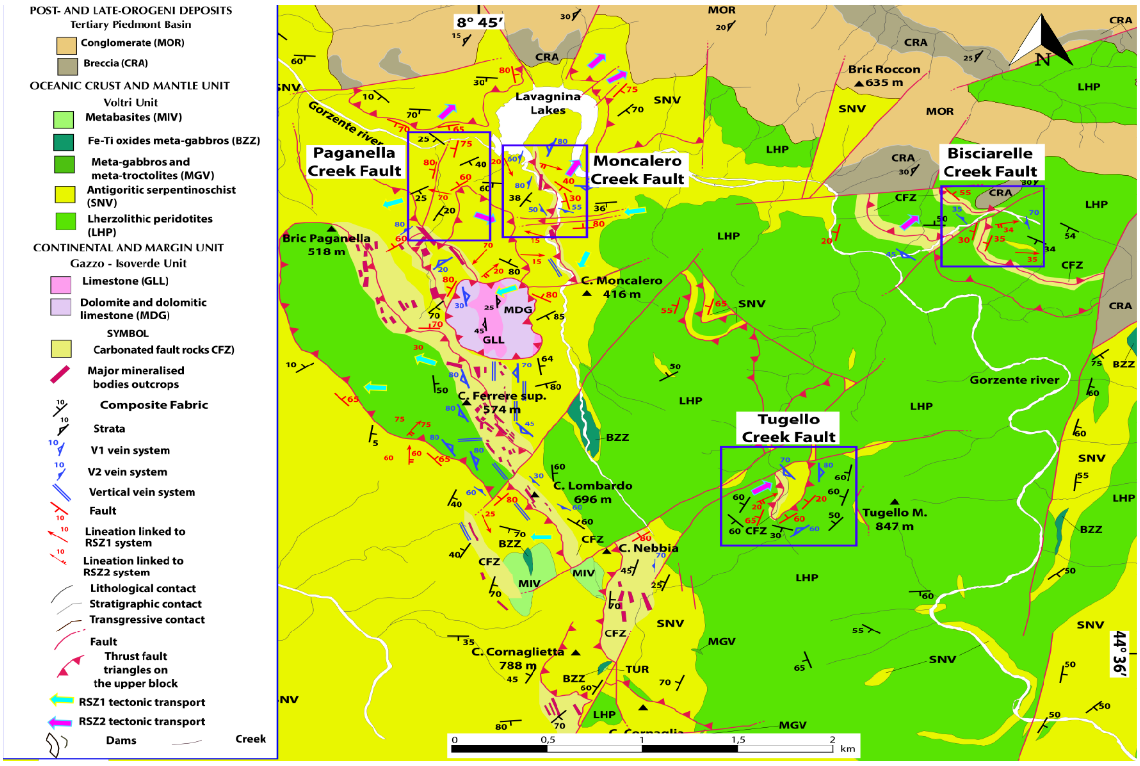

Fig. 1 - Structural sketch of the Lavagnina Lakes Area, redrawn with updates and georeferenced with Qgis open source software (modified after Spagnolo *et al.*, 2007). In this sketch, the four structures analysed in detail are highlighted with the blue squares.

Within the area, the gold occurrences were known since the Roman age (Pipino, 2003) and the area was site of mining exploitation with the presence of a metallurgical plant, dedicated to the manufacturing of gold ingots, and a mining village (from 1589 until the end of 1800).

The Voltri Massif is located at the southernmost termination of the Western Alps. It is a meta-ophiolitic complex, consisting of serpentinites with metagabbros, metabasites, metasediments and lherzolites with minor pyroxenites and dunite bodies. It experienced a complex Alpine tectono-metamorphic history from ductile- to brittle-regime (Capponi & Crispini, 1997, 2002) and from high-pressure conditions to variable retrogressive overprints (Cimmino & Messiga, 1979; Capponi & Crispini, 1997; Desmons *et al.*, 1999; Scarsi *et al.*, 2017). Further details on the geological framework of the area can be found in Capponi *et al.* (1998), Capponi & Crispini (2002), Pipino (2003), and Spagnolo *et al.* (2007).

The Lavagnina Lakes area is characterised by meta-peridotites of the Voltri Massif (Erro-Tobbio Unit, (Chiesa *et al.*, 1975) consisting mainly of lherzolite with various degree of serpentinization, metasediments, and outcrops of continental breccia and conglomerate of the Tertiary Piedmont Basin, limited by vertical faults.

The area also comprises, at Case Ferrere locality, a carbonatic klippe, bordered by a thrust zone with a W-SW sense of shear (Capponi *et al*., 1998).

The structural evolution of the Lavagnina Lakes area is witnessed by several superimposed deformations, which occurred under tectonic conditions evolving from brittle-ductile to pure brittle regime.

The oldest Alpine structures of the area can be referred to the D1/D2 deformation phases described in Capponi & Crispini (2002), characterised by folds and by a composite fabric that is the most pervasive foliation in the field that controls the contacts between lithologies.

The post-D1/D2 deformation events are characterised by a complex superposition of ductile, brittle-ductile and brittle structures that develop at higher structural levels and are represented by different set of folds (comparable with the D3 and D4 folds described by Capponi & Crispini, 2002), reverse shear zones, and fault systems. As described by Spagnolo *et al.* (2007) brittle-ductile upper crustal deformations (UCD) are widespread in the entire area, and are mainly represented by systems of Reverse Shear Zones (RSZs) with associated vein network, wall rock metasomatic alterations, and locally gold mineralisation (Pipino, 2003). The RSZs, according to their structural features and opposite kinematics, can be clustered into two systems.

RSZ1 (Fig. 2) are ductile to brittle-ductile structures syntectonic with greenschist to sub-greenschist metamorphic conditions. RSZ1 damage zone developed the older set of veins recognizable (V1), hydraulic and cockade breccias, and metasomatic alteration (ALT-1) that causes carbonation and hydration of the bedrocks.

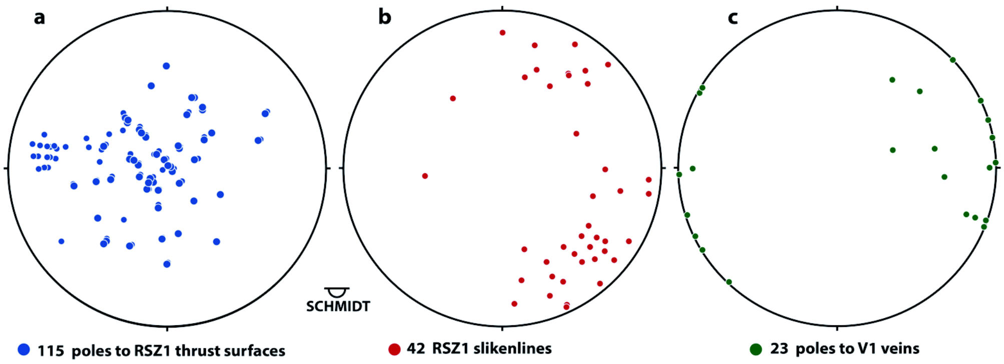

Fig. 2 - Stereographic projections. (a) Poles to RSZ1 thrust surfaces (115 points). (b) Slickenlines (42 points). (c) Poles to V1 veins (23 points).

RSZ2 (Fig. 3) are brittle-ductile to pure brittle structures developed from sub-greenschist to zeolite facies. RSZ2 structures show two main sets of superposed shear fibre lineations: the first mainly dips to W and minor to E with a main top to E, the second dips both to SW and NE with top to NE. RSZ2 damage zone developed two main sets of veins (V2-V3 Auct.), hydraulic and cockade breccias, and two kind of metasomatic alteration (ALT-2, ALT-3).

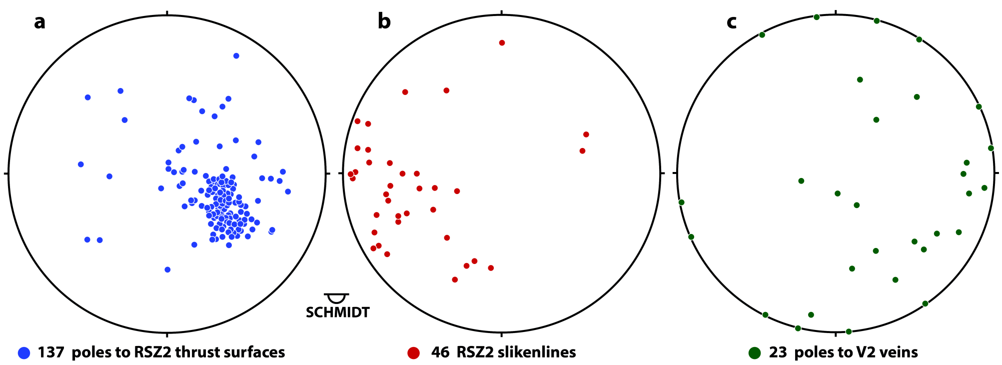

Fig. 3 - Stereographic projections. (a) Poles to RSZ2 thrust surfaces (137 points). (b) Slickenlines (46 points). (c) Poles to V2 veins (23 points). The data are from the field survey for this work.

Strike-slip and oblique slip systems are common in the Lavagnina Lakes. These structures are characterised by kilometric scale, nearly vertical N-S striking faults, which are strictly related to positive flower structures. These structures are associated to minor Riedel faults NNE-SSW and NNW-SSE striking that can be defined as R and P shear faults, and are characterised by dm to m thick fault rocks levels.

On the basis of the geological survey and of the field evidences of intense syn-tectonic fluid circulation and consequent fluid-rock interaction, it was decided to investigate with more detail four main structures (Fig. 1). The fluid-rock interaction triggered the development, along these structures, of several types of metasomatic rocks and peculiar textures. Based on the geological survey these four structures belong to the brittle-ductile structures that affect the investigated area. The four structures are: the Moncalero (Fig. 4), the Tugello (Fig. 5), the Bisciarelle (Fig. 6a), and the Paganella Creek Fault (Fig. 7).

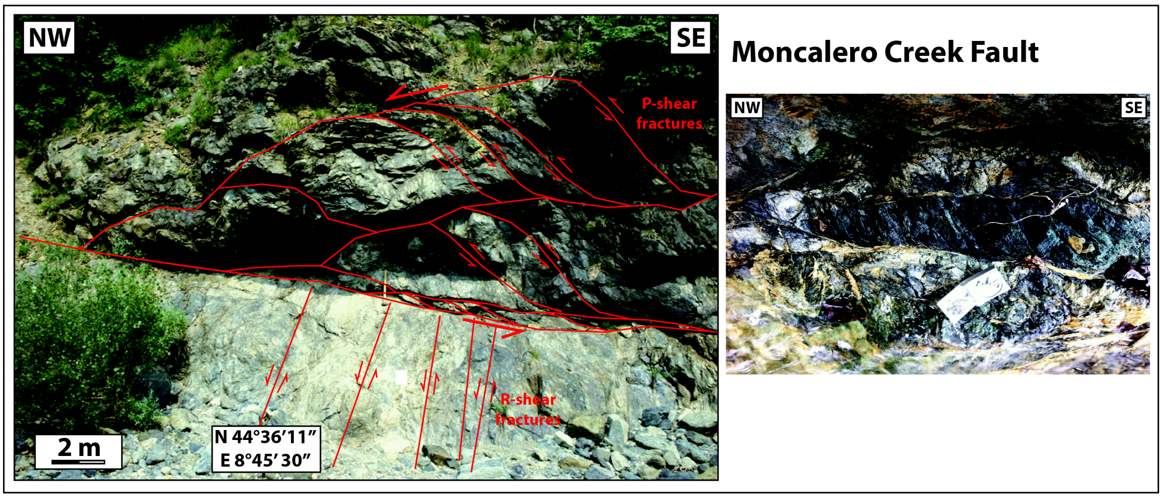

Fig. 4 - Moncalero Creek Fault, with detail of the fault core.

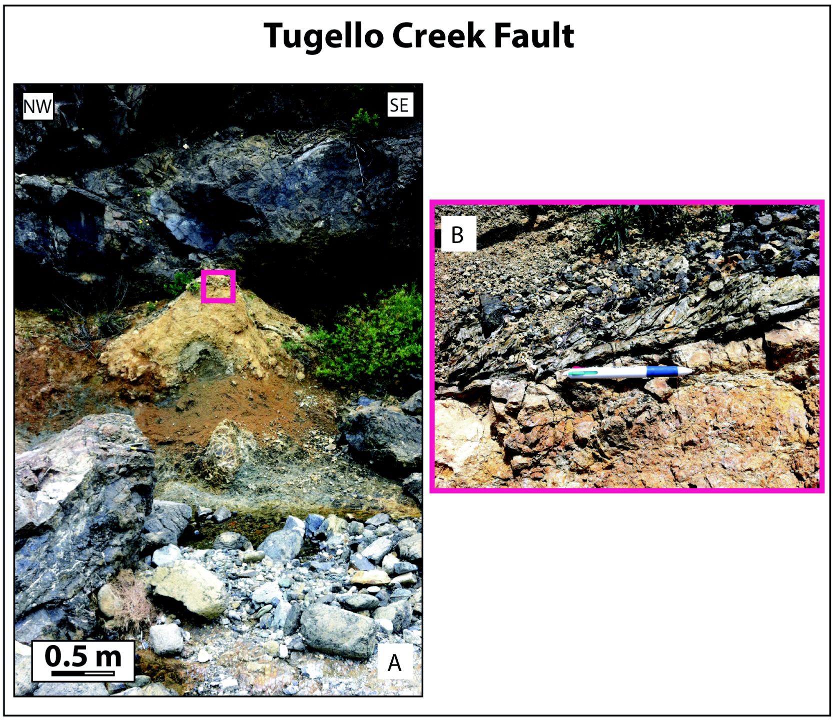

Fig. 5 - Tugello Creek Fault, the pink square highlights the magnification of the sepiolite level, with duplex structures.

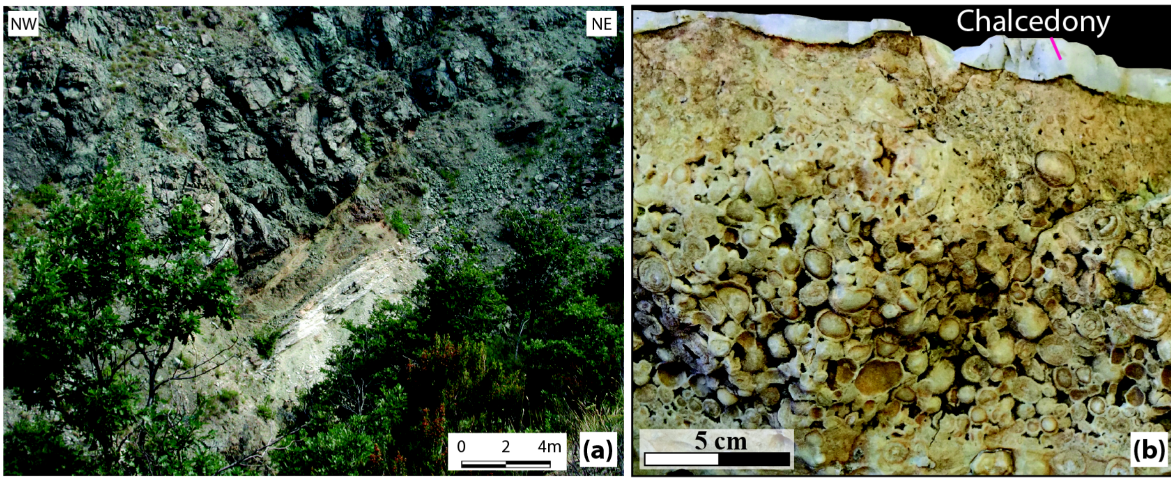

Fig. 6 - Bisciarelle Creek Fault, (a) Detail of the Bisciarelle Creek Fault; (b) Magnification of the sample of the spherulites, sampled from the central part of the fault core.

On the basis of the geological survey and the detailed study of these structures, their orientations, their tectonic directions of movement and for their deformational events, the Moncalero Creek Fault has been related to the older system of regional reverse shear zones (RSZ1), the Tugello and the Bisciarelle Creek Fault to the younger system of regional shear zones (RSZ2), and the Paganella Creek Fault to the minor system of strike-slip/oblique slip systems and in particular a P-shear fault, related to the younger fault structures recognized in the investigated area. In particular the Bisciarelle Creek fault has been studied for the occurrence of dolomitic spherulites within the fault core (Fig. 6b).

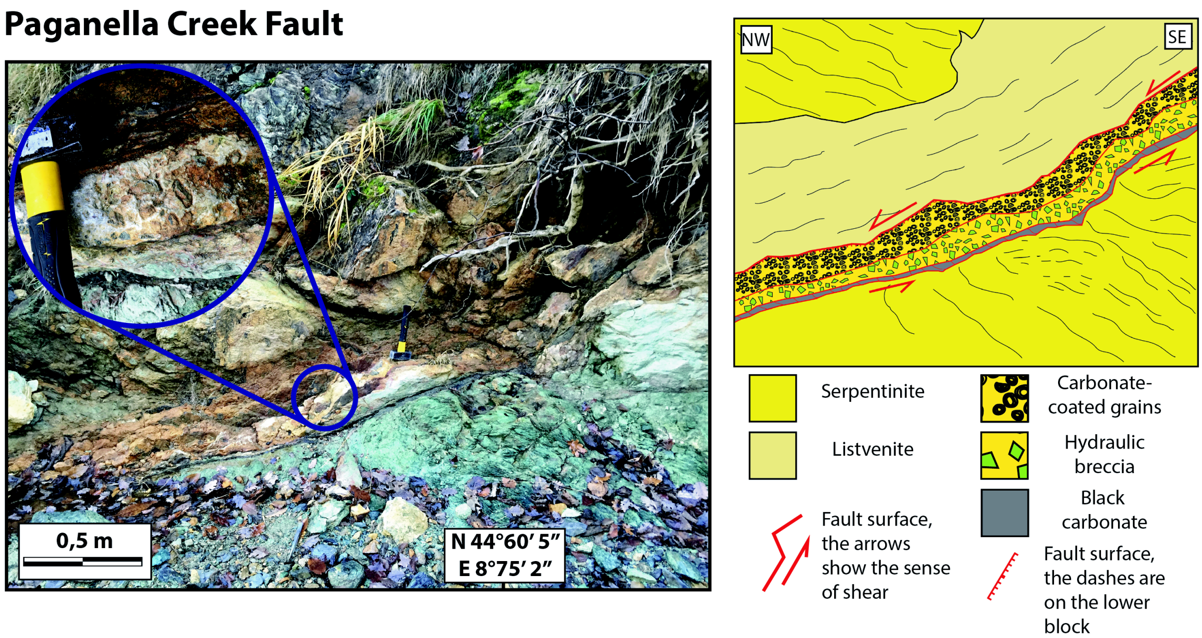

Fig. 7 - Paganella Creek Fault, with structural sketch, and in the right part of the figure are reported the structural elements of the fault.

RESULTS AND DISCUSSION

The oldest UCD event is characterised by the coexistence of dragged folds with chevron style, and the oldest system of reverse shear zone (RSZ1, Moncalero Creek Fault), that are developed under greenschist to low greenschist metamorphic conditions. Commonly, RSZ1 structures develop in the reverse limbs of the D3 folds or cut them; locally the RSZ1 may be folded. This testifies to an early development of the RSZ1, under ductile regime and their subsequent evolution from the ductile to brittle-ductile regime. The RSZ1 structures (Fig. 2a, b) are developed under greenschist to low greenschist metamorphic conditions, they show a main sense of shear with top to S-SW, with a minor top to N-NW (*e.g.* Moncalero Creek Fault), and are mainly developed in serpentinites and lherzolites. They show a symmetric damage zone with development of damage zones up to 1 m thick. The symmetric development of the damage zones is the product of ruptures along faults separating bodies with the same elastic characteristics. This limited damage zones implies small variations in terms of porosity and permeability conditions with respect to the characteristics of the host rocks. The damage zones linked to the RSZ1 structures show veins (V1, Fig. 2c) and the occurrence of a former intense carbonation (ALT-1) with the development of listvenites and are related to the oldest gold-bearing mineralisations of the area. This ALT-1 event is volumetrically limited since the alteration occurs only near the main faults or near P and R fractures and joints. This evidence points out the importance of such system in influencing the fluid circulation, and the limited development of the carbonation implies that this process leads to a reduction of the porosity and permeability turning the structures from a preferential pathway to a potential seal for the fluid circulation. The temperatures constrained by Giorza (2010), for the ALT-1 using fluid inclusion in magnesite and Fe-dolomite, are of ca. 270-300°C. Considering a possible range of geothermal gradients the ALT-1 has been developed at about 4-11 km.

The RSZ2 structures (Fig. 3a) are developed under zeolite facies to non-metamorphic conditions, are widespread in the study area (*e.g.* Tugello Creek Fault and Bisciarelle Creek Fault). This system in place locally reactivates pre-existent RSZ1 structures. The RSZ2 structures show scattered slickenlines (Fig. 3b), and a variable sense of shear, with at least two different stages (A-B) of tectonic movements. The older stage (RSZ2-A) is characterised by a main top to NW, instead the younger stage (RSZ2-B) is characterised by top to - SW and - NE. The temperatures constrained by Giorza (2010), for the ALT-2 using fluid inclusion in magnesite and Fe-dolomite, are of ca. 250°C. Considering a possible range of geothermal gradients the ALT-2 has been developed at about 3-9 km. With respect to the oldest RSZ1 system, the younger RSZ2 system shows a volumetrically more developed carbonation (ALT-2) and completely different structural and textural characteristics. The RSZ2 reverse faults, developed quartz-carbonate faults hosting orogenic Au deposits. They are developed near the lithological transition between serpentinized lherzolites, at the hanging wall, and the serpentinites that make up the footwall. These faults show an asymmetric damage zone, more developed in the footwall with respect to the hanging wall, with several meters of different fault rock types. The asymmetric development of the damage zones is the cumulative product of earthquakes ruptures on bimaterial faults separating different elastic bodies (Ben-Zion, 2001; McGuire *et al.*, 2002; Ben-Zion & Shi, 2005; Shi & Ben-Zion, 2006; Mitchell *et al.*, 2011). Hence, the asymmetry that characterises the damage zone of the RSZ2 fault system should be the result of cumulative seismic ruptures along the bimaterial interface represented by the serpentinized lherzolites and the serpentinites.

The core of the analysed structures, that was here ascribed to the RSZ2 system (the Tugello Creek fault and the Bisciarelle Creek fault) host levels of carbonates (up to two levels) characterised by carbonate-coated grains, cockade or hydrothermal breccias and by spherulites (only the Bisciarelle Creek fault), coeval with sub-micron carbonates coating of slip surfaces, chalcedony shear veins and shear surfaces, injection veins, syn-kinematic filamentous phase, and sepiolite duplex structures. The chalcedony shear veins show textures similar to the moss, jigsaw and flamboyant textures, described as related by crystallization after silica gel deposition (Moncada *et al.,* 2012). The presence of sepiolite, as the presence of chalcedony crystallized over a previous deposition of amorphous silica gel, along fault slip surfaces have been found to play a significant role in controlling fault strength leading to an almost complete loss of strength facilitating shear localization (Sánchez-Roa *et al*., 2017, 2018). At local scale, the presence of these "soft" layers along the fault surfaces may have localised the shear deformation along the faults of the RSZ2, reactivating them, as highlighted by the development of different fault rock types along the structures ascribed to this system in the study area.

The occurrence of syn-tectonic fluid flow and consequent fluid-rock interaction (carbonation or listvenitization) along the two systems of Reverse Shear Zones (RSZ1-RSZ2), is testified by metasomatic alteration (ALT-1, ALT-2, and ALT-3), by at least three systems of veins (V1, V2, and V3), and by the development of hydraulic and cockade breccias. The evidence that the carbonation took place along these structures, in particular along the RSZs systems, and within their damage zones, highlights how these structures acted as important fluid pathways and played a major control in the distribution of the ore deposits.

The detailed study of RSZs structures in the Lavagnina Lakes area showed that, though they are similar in attitude to other RSZs widespread throughout the Voltri Massif, they contain particular mineralisations (*e.g*. gold and chalcedony) and structures that have been described for the first time in this work. Analysing RSZ2 structures and in particular the Bisciarelle Creek Fault, it appears clear that the fluid involved in the development of gold mineralization of the Lavagnina Lakes area can be related to a tectonically driven hydrothermal system and/or orogenic system gold deposits.

The mapping of gold mineralization and of chalcedony occurrences in the Voltri Massif allowed to highlight that, at a km-scale, they occur along a lineation of mineralization associated with chalcedony and quartz veins. At regional scale this lineation is oriented mainly NNW-SSE, similarly to the axes trend of D4 folds. Hence the RSZ2 in the Voltri Massif could effectively represent a regional tectonically driven hydrothermal systems.

A detailed outcrop-scale investigation highlighted that the Bisciarelle Creek fault, that was related to the RSZ2 system, shows peculiar fault textures, organized in bands, called spherulites. This fault (exposed for ca. 100 m) shows a complex core characterised by parallel to sub-parallel multiphase slip surfaces that are mainly sub-horizontal or gently dipping to W-SW with top to NE; it is coeval with syntectonic Au-bearing quartz and chalcedony shear veins.

Fault activity was accompanied by fluid infiltration that caused an almost complete replacement of the original host rock mineralogy, now characterised mainly by dolomite and ferroan-dolomite and smectite alteration. Fluid infiltration and related mineral by mineral replacement were enhanced by schistosity and veins. The majority of the spherulites have a circular shape irrespective of the direction of observation of the spherulite levels, and have an average dimension of about 0.5 cm. The largest ones show a complex internal texture whereas the smallest ones show a simpler and more massive internal texture. Large spherulites are made up of relatively thick (up to 8 mm) and fibrous dolomite bands that alternate regularly with thinner (up to 2 mm), massive, and partly laminated dark carbonate bands, whereas, in general, small spherulites (size < 3 mm) do not show concentric texture. Several alternating levels can be identified in the larger spherulite (up to 9 levels). The central part of the spherulites is made of, in decreasing order of abundance by: *i*) carbonates crystals (80%), *ii*) relic of carbonate clasts (15%), and *iii*) relic of carbonated serpentinite clasts (5%).

Spherulites are different from clast-cortex grains described in literature in other fault contexts (Smith *et al*., 2011; Rempe *et al*., 2014; Rowe & Griffith, 2015), because these are composite grains that form at the slipping zones of faults and are made of a central clast and an outer fine-grained cortex of smaller existing clasts, sticked together. In contrast with the spherulites, the shape of these aggregates is poorly rounded and their textures is cataclastic and lacks banding; thus, their genetic mechanism must be distinct from that of the spherulites. By contrast, spherulites are really similar to pisolite or cave pearls (Nader, 2007), related to vadose groundwater circulation or to oolite/pisolite deposits related to hydrothermal circulation (Sant'Anna *et al*., 2004; Wu *et al*., 2014).

The possibility of a pre-existent pisolite-rich limestone cut by the fault was rule out in this work because its movement would have resulted in a pervasive cataclasis of the carbonatic horizon, and because some spherulites have a carbonated serpentinite central clast. The growth of pisolites did not occur after the end of the fault activity because evidence of truncation of the grains by both shear surfaces and chalcedony shear veins locally exist; if these grains formed during a period of inactivity of the fault they would again been damaged by the following slip. Conversely, it is here suggested that the spherulites are coeval with the fault activity, for the presence of spherulites with carbonated serpentinite core clast, and the similarities with cave spherulites derive from a convergence in the way of growth, irrespectively of the different genetic environment: actually, both record the alternation of different types of fluid circulation. Moreover, the spherulites are associated with injection veins, carbonate shear surface with characteristic similar to mirror-like surface, vescicular porosity, chalcedony shear veins with textures testifying crystallization over silica gel, chalcedony shear surface characterised by quartz surrounded by filamentous silica matrix; in literature, all these textures are related to fast slip and possible paleo-seismic events.

Single-spot SEM-EDS analyses revealed that ferroan dolomite makes the spherulite bands (CaO: 29.13±2.33 wt.%; MgO: 19.03±4.86 wt.%; FeO: 1.77±0.87 wt.%) and that minor compositional variations across large and thin bands exist. Elemental imaging by Laser Ablation Inductively Coupled Plasma Time-of-Flight Mass Spectrometry provides a detailed account of the distribution of chemical elements within spherulites, matrix, and chalcedony shear veins, confirming the hypothesis of hydrothermal derivation of the fault rock.

Hence, considering all the constraints, two models for the genesis of spherulites along fault core are here proposed:

i) the first model envisages the development of the spherulites during several cycle of hydrothermal fluid flows (T around 250°C), coeval with a stick-slip movement, with cycles of fluid pressure build-up, fault opening, fluid flushing, and mineral precipitation during the seismic failure of the fault, according to what proposed by Sibson *et al.* (1988);

ii) the second proposed mechanism is the process known as transient boiling in microfluidic industry (Zhao *et al*., 2000), which is also described as "nucleate" boiling. Thus, the closely packed spherulites would represent the mineralized products of vapor generated by the transient boiling of the hydrothermal fluid at ~250°C and the porous spherulite matrix would represent the co-existing liquid phase.

In conclusion, the analysed reverse shear zones, that were here related to the RSZ2 system, are characterised by several textures (*e.g.*, carbonate shear surface with characteristic similar to mirror-like surface, vescicular porosity, chalcedony shear veins with textures testifying crystallization over silica gel), and by gold mineralisations that in literature are both related to fast fault slip and possible paleo-seismic events (Weatherley & Henley, 2013). This evidence could support the hypothesis that at local and probably at regional scale the RSZ2 system can be considered a system of paleo-seismic structures. Further investigation may provide important information about this possibility.

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