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Serpentinites of the Pollino ophiolite massif (Calabria, southern Italy), mineralogy, petrography and future perspectives

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ABSTRACT

Serpentinites of the Pollino ophiolite Massif have been widely studied for their mineralogical, petrographic, and chemical characteristics, providing major findings on their impact on environmental and human health issues. The mineralogy and the petrography of the studied serpentinites revealed the presence of lizardite, antigorite, crysotile, Cr-chlorite, magnetite, tremolite, actinolite, pyroxene, calcite, dolomite, aragonite, and talc, and the most commonly occurring minerals are different polymorphs of serpentine. Capturing atmospheric CO_2 and storing it in natural rock systems through carbonation reactions is a promising greenhouse gas mitigation research, and largely depends on the use of hydrated ultramafic rocks, which are widespread in orogenic belts around the globe. The integration between petrographic and mineralogical studies of serpentinites in Southern Italy ophiolites can represent a starting point for the study of natural materials, which can be used for CO_2 storage and sequestration. A particular attention must be given to those carbonate phases that are produced by carbonation processes to check whether the serpentinites of the Pollino Massif can be used for the induced mineral Carbon Capture Storage (CCS). If successful, this technique and the Pollino Massif serpentinites can become highly significant in safeguarding the health of our planet's climate.

Keywords: serpentinites; health risk; carbon storage and sequestration; mineralogy and petrography; Pollino Massif; Calabrian ophiolites.

INTRODUCTION

Carbon dioxide (CO_2) is a greenhouse gas and plays a major role in the regulation of Earth's temperature and affecting global warming patterns. Its concentration in the atmosphere has increased significantly over the past several decades (Hegerl et al., 2006; Foster et al., 2017; Global Carbon Budget, 2023; Figure 1).

The interest in carbon management involves the use of a range of technologies to capture, store, transport and use CO_2 emissions, as well as to remove it from the atmosphere (EU Regulation 2021/1119 of the European Parliament and of the Council, European Climate Law). The EU Industrial Carbon Management Strategy seeks to develop these technologies and the regulatory and investment framework to support them. It focuses on i) the capture of CO_2 for storage (CCS); ii) the capture of

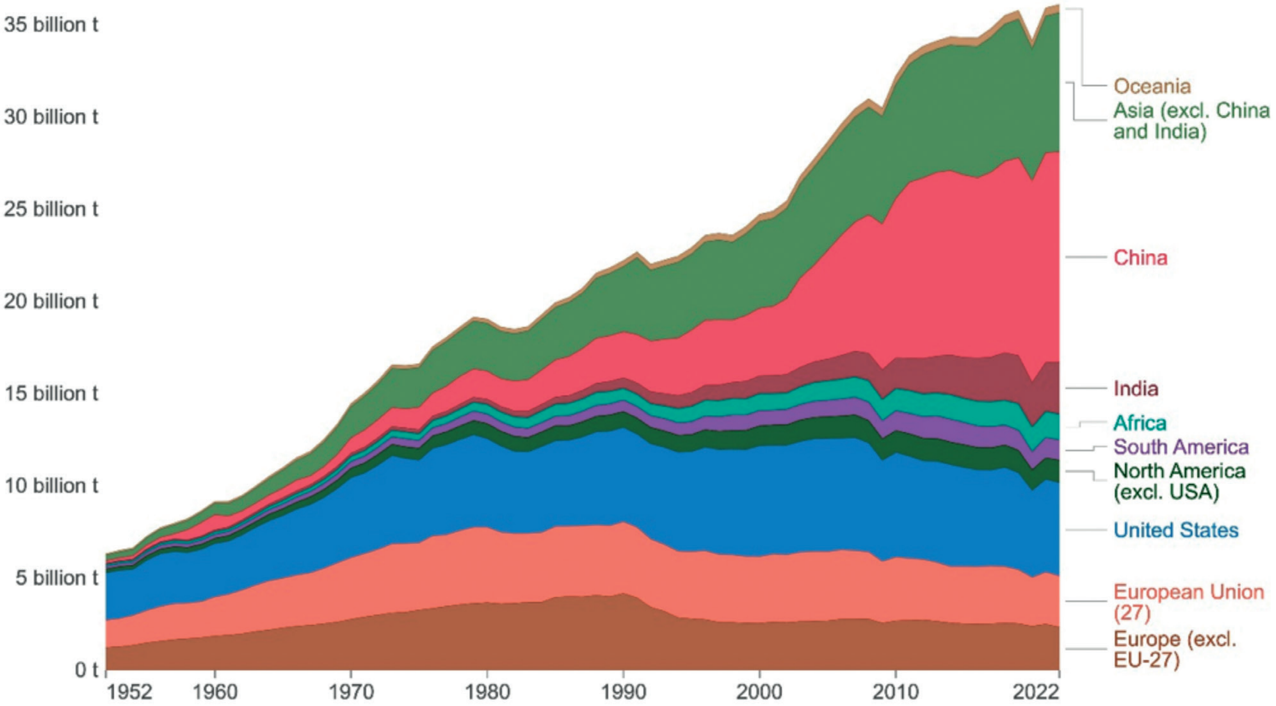

Figure 1. Annual CO_2 emissions by world region (modified after Global Carbon Budget, 2023).

CO_2 for utilisation (CCU) and iii) the removal of CO_2 from the atmosphere.

Most carbon on Earth is bound in minerals and sequestered in deep-sea sediments, reminding us that increasing the uptake of CO_2 in minerals could reduce the greenhouse gas content in the atmosphere. Mineral carbonation is the process by which carbon dioxide (CO_2) reacts with minerals in rocks to form solid carbonates and represents the most environmentally and geologically stable method for carbon disposal (Oelkers et al., 2008). This reaction can occur naturally over geological timescales or can be artificially accelerated for carbon capture and sequestration (Herzog, 2002). Moreover, anthropogenic gas emissions are projected to alter future climate with potentially non-trivial impacts (Keller et al., 2008), and the effects of increased CO_2 concentrations can be clearly documented through greenhouse effects, ocean surface acidification, and ecosystem fertilization (Lackner et al., 1995; Huijgen and Comans, 2003). The observed increase in atmospheric carbon dioxide (CO_2) concentrations motivates the study of carbon capture and storage (CCS) as an important component of multilateral strategies to mitigate the risks of future climate change. Proposed reservoirs for CO_2 storage include terrestrial biomass, deep sea, saltwater aquifers, and minerals, and these reservoirs differ significantly in terms of the rate at which stored CO_2 leaks into the atmosphere. Characteristic storage periods increase from terrestrial pools (tens to hundreds of years), deep sea (hundreds of years), geological reservoirs (thousands of years), to thermodynamically stable minerals (> thousands of years) (Lackner, 2003; Freund et al., 2005). Carbonation is a process that can be performed on metal-rich, non-carbonate minerals (e.g., Mg_2SiO_4 i.e., Mg_2SiO_4 olivine, $Mg_3Si_2O_5(OH)_4$ serpentinite, $CaSiO_3$ wollastonite) to geologically and thermodynamically stable mineral carbonates [i.e., $MgCO_3$ magnesite, $MgCa(CO_3)_2$ dolomite, $CaCO_3$ calcite, $FeCO_3$ siderite, $NaAl(CO_3)(OH)_2$ dawsonite] by induced exothermic alteration (Lacinska et al., 2017). This technique attempts to mimic the natural, low-temperature alteration (carbonation) of a wide range of silicate rocks that safely trap CO_2 over geological time (Lackner et al., 1995; Seifritz, 1990; Goff and Lackner 1998). Among various mineral species that can undergo carbonation reactions, Mg-rich minerals, as well as Ca-carbonates, are significant acting as important reservoirs of CO_2 since they are highly abundant at the Earth's surface.

The mineral carbonation process is typically a reaction of olivine or serpentinite with carbon dioxide to form magnesite + quartz + water: $Mg_3Si_2O_5(OH)_4 + 3CO_2 = 3MgCO_3 + 2SiO_2 + 2H_2O$ (Power et al., 2013). The rate of CO_2 sequestration in nature is controlled primarily by reactive surface area, temperature, pH, and CO_2 partial pressure. There is an obvious advantage of in-situ carbonation since CO_2 is injected in-situ, with no need to mine, pulverize, or activate the serpentine rock. The ophiolites of the South Apennines (Italy) represent a natural analogue for in-situ mineralogical sequestration of CO_2. By binding the CO_2 in silicate rocks through mineral carbonation, it is possible to produce stable, also naturally occurring, carbonate rocks (Zevenhoven and Fagerlund, 2010).

The typical procedure starts with a complete characterization of the mineralogy and composition of the rock, followed by laboratory and pilot-scale reactivity tests to determine the rate and extent of the carbonation reaction of a given rock. In the case of field studies, once carbonation has started, the process is monitored to determine the extent of carbonation over time and to ensure long-term carbonate stability (Spadea, 1976; Beccaluva et al., 1982; Lackner et al., 1995). In the quarry site, the serpentinite rocks reacting with calcite in the presence of CO_2 could be a useful sink for carbonation processes and CO_2 storage.

Serpentinites cropping out in the Pollino mafic-ultramafic massif in the Calabria region of southern Italy have been studied extensively and analyzed for their mineralogical, petrographic, and chemical characteristics, which also provided relevant findings on their impact on environmental and human health (Bloise et al., 2012, 2017, 2019; Caputo et al., 2018; Dichicco et al., 2018, 2019; Punturo et al., 2023 a,b; Rizzo et al., 2016, 2018).

Cataclastic serpentinites are more apt to be carbonated due to their texture with extensive fracturing of individual grains that increases the reactive surface area of minerals. Regarding future perspectives, it can be interesting to undertake a petrological study of the serpentinites of the Pietra Pica quarry with particular attention to the carbonate phases present and to the existence of talc to check whether the serpentinites of the Pollino Massif can be used for the induced mineral Carbon Capture Storage. In order to mitigate climate change, as part of international efforts related to European climate protection directives, this paper proposes the use of serpentinite from the Pollino massif as a high-potential carbon capture and storage site.

GEOLOGY OF THE POLLINO OPHIOLITE MASSIF

The Pollino Massif in the Southern Apennines (Italy) is located within the continental convergence zone between Eurasia and Africa and is composed mainly of Mesozoic and Tertiary magmatic and sedimentary rocks derived from the Ligurian ocean basin and the African passive margin. These oceanic and continental crustal rocks are overlain by Pliocene-Pleistocene terrestrial deposits (Figure 2a; Cello and Mazzoli, 1999). Collectively, the Pollino Massif and the continental crustal rocks make up the Liguride Complex of the Apennines and the Calabria region (Spadea, 1982; Bonardi et al., 1988; Knott, 1994; Monaco and Tortorici, 1995; Stampfli et al., 2002). The Pollino Massif (Jurassic palaeo-oceanic lithosphere) is part of the Frido tectonic unit (Knott, 1987), which includes a metasedimentary sequence (phyllite, meta-arenite, quartzite, and blocks of calcschist and metapelite; (Monaco et al., 1991; Monaco and Tortorici, 1994) and a Hess-type, incomplete ophiolite, composed of serpentinite, metabasalt, metagabbro, metapillow lavas, and dismembered metadolerite dikes (Spadea, 1982). The Pollino massif likely represents a fossil ocean-continent transition zone or a continental margin ophiolite (Dilek and Furnes, 2011, 2014), reminiscent of the modern Western Iberia rifted margin (Pinheiro et al., 1996). The second major tectonic entity in the Frido Unit is a non-metamorphic Calabro-Lucano Flysch or the North Calabrian Unit. The Pollino Massif tectonically overlies the North Calabrian Unit and forms the structurally highest tectonic unit in the Liguride Complex.

Different interpretations have been proposed for the geological origin of the Liguride complex (Di Leo et al., 2005; Cavalcante et al., 2009; Tortorici et al., 2009). Bonardi et al. (1988) have suggested that metapelitic rock assemblages, including both continental crustal and ophiolitic rocks, constitute a mélange zone (Episcopia–San Severino mélange). Other interpretations consider the Liguride complex as a stack of thrust sheets in an accretionary prism complex, characterized by variable lithologies and metamorphic overprints (Monaco et al., 1991; Monaco and Tortorici, 1995). Different rock assemblages in the Frido Unit underwent HP-LT, blueschist facies metamorphism (Spadea, 1994). In the study area the outcropping complex (Liguride complex) is covered by thrust top basin deposits consisting of siliciclastic, calciclastic and Pleistocene sediments.

MATERIALS AND METHODS

A total of fifteen serpentinite samples of the Frido Unit selected for this study was collected at Pietrapica quarry, near the settlements of San Severino Lucano and Episcopia (Calabria-Lucanian boundary, Basilicata region, southern Italy), where outcrops of the Pollino massif serpentinites are widespread (Figure 2b).

At the scale of the outcrop the studied serpentinite looks dark green to greysh colored; they appear to be quite massive; however, at the mesoscale several cross-cutting fractures were observed as well as fibrous minerals within (Figure 3). Indeed, the massive serpentinite samples

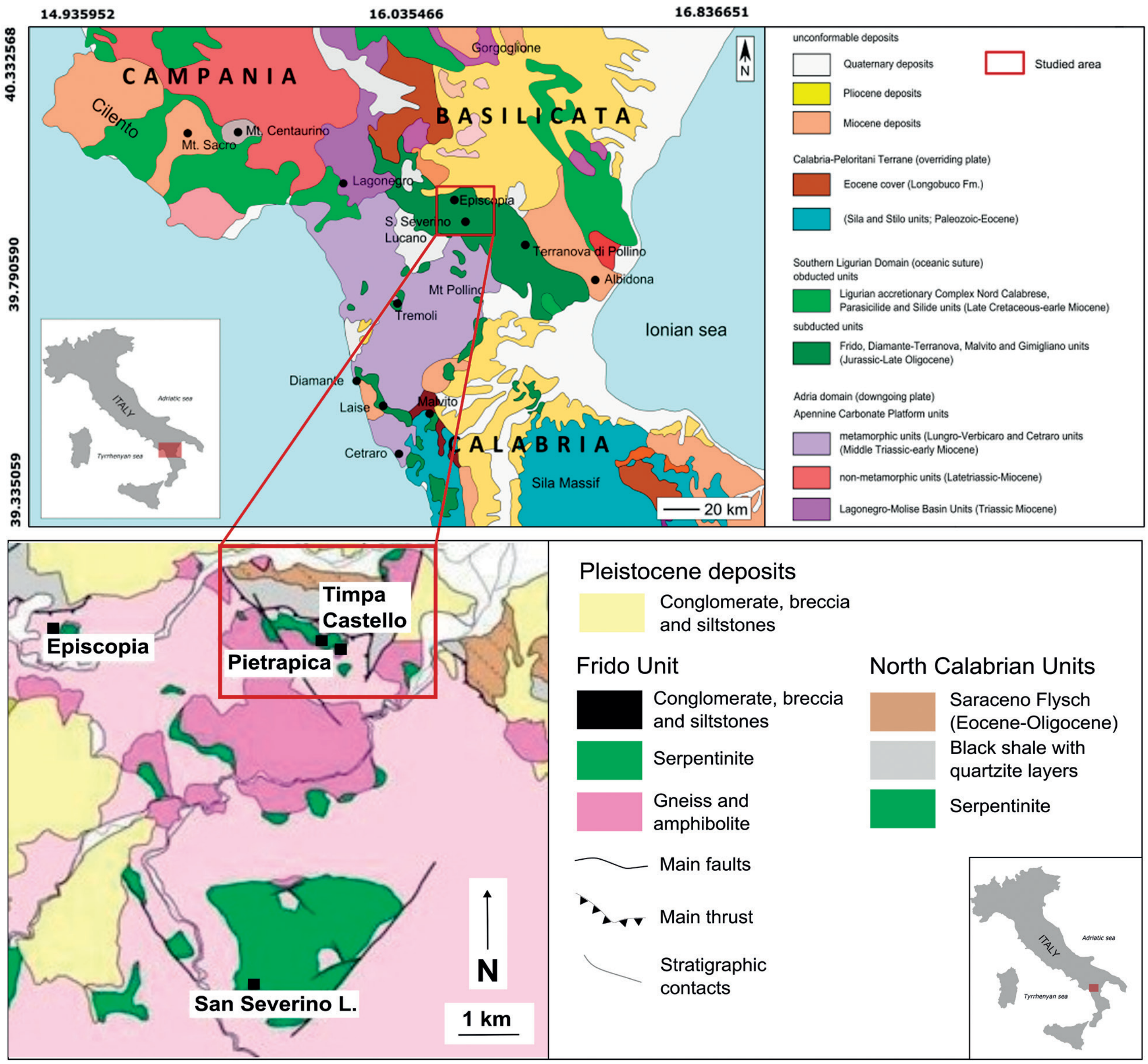

Figure 2. a) Geological map of Southern Apennines (modified after Sansone et al., 2012; b) Simplified geological map of the Liguride Complex in the Pollino Massif with the location of the Pietrapica quarry site and the outcrop features of the Pietrapica serpentinites (modified after Rizzo et al., 2023).

are weakly fractured and deformed and sometimes it is quite difficult to cut them for obtaining rock slab. Macroscopically, the analysed serpentinites show quite homogeneous features which permit them to be divided into two main types: the first type of serpentinite samples is cataclastic type, showing fracturing and rigid-body rotation of mineral grains or aggregates while the second type of serpentinite samples is a massive type, consisting of fractured serpentinites embedded in cataclastic serpentinites (Figure 4). Recent petrographic and mineralogical studies have documented the occurrence of asbestos minerals in the area (e.g. Dichicco et al., 2018, 2019; Bloise et al., 2019; Ricchiuti et al., 2021; Punturo et al., 2023; Rizzo et al., 2023).

Petrographic investigation of all the specimens was carried out by using a Nikon Alphaphot-2 YS2 optical microscope on thin sections of rock samples. Semi-quantitative mineralogical analyses were performed on randomly oriented powder by using a Philips X'Pert 3040PW with CuKα radiation, 40 kV and 40 mA, 1 s per step, and step scan of 0.01 °2θ, in the Department of Sciences at University of Basilicata. The analyses by

Figure 3. Photograph of the sampled Pietrapica serpentinite quarry.

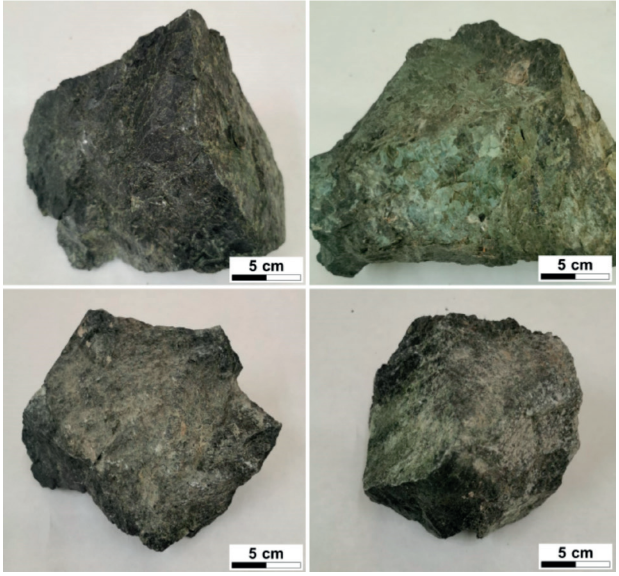

Figure 4. Macroscopic features of the studied serpentinite samples.

Raman were carried out at the Department of Sciences, University of Basilicata. The used spectrometer, a Horiba Jobin-Yvon LabRam HR800, is equipped with a HeNe laser source with a wavelength of 633 nm. The detector operated at -70 °C with an edge filter excluding the detection shift below 150 cm^{-1}. The evaluation of the calibration of the instrument was made by checking the position of the Si band at ±520.7 cm^{-1}. The measurements were performed using optical microscope Olympus with objective of 10×, 50× and 100× and the Raman spectra were collected in the spectral ranges of 200-1200 cm^{-1} and 3200-3800 cm^{-1} with an average of 5 acquisitions of 10 s. Micromorphological analysis was undertaken by using Scanning Electron Microscopy (ESEM) at the Microscopy Laboratory of the Department of Basic and Applied Sciences, University of Basilicata, Italy. SEM-EDS investigation was carried out on serpentinite representative fragments with a Tescan- Vega\\LMU scanning electron microscope (SEM), equipped with an energy-dispersive X-ray spectrometer (EDS) Edax Neptune XM4 60 (SEM laboratory, Department of Biological, Geological, Environmental Sciences, University of Catania.

RESULTS

The studied serpentinite rocks are characterised by pseudomorphic and vein textures. The pseudomorphic texture (Figure 5a) is represented by a mesh texture (Wicks and Whittaker, 1977; Wicks et al., 1977; Prichard, 1979) in which serpentine and magnetite phases statically replaced olivine grains whereas yellow brown bastite replaced orthopyroxene. The vein texture is represented by different sub-millimetric veins intersecting each other and criss-crossing the pseudomorphic texture.

In the Pietrapica serpentinites, the protolith minerals are olivine, pyroxenes (both orthopyroxene and clinopyroxene), and spinel, whereas the secondary (i.e. metamorphic) minerals are serpentine, chlorite, magnetite, prehnite, and amphibole. Olivine is locally recognisable in the core of serpentine + magnetite pseudomorphs in the mesh texture (Figure 5b). Orthopyroxene is observed as fresh porphyroclasts as well as bastite pseudomorphs, which retain the prismatic habit of the original orthopyroxene (Figure 5c); orthopyroxene also occurs as lamellae within clinopyroxene porphyroclasts. Clinopyroxene porphyroclasts are commonly rimmed by amphibole, which is identified as belonging to the tremolite-actinolite series based on optical characteristics (Figure 5d). Spinel forms red-brown coloured, xenomorphic, holly leaf-shaped porphyroclasts defined as typical of porphyroclastic upper mantle peridotites (Mercier and Nicolas, 1975).

Serpentinites are crosscut by several types of veins, filled with various serpentine minerals (Figure 5e). In some samples, kinked orthopyroxene is replaced by pseudomorphic amphiboles (Figure 5f), which are interpreted as belonging to the tremolite-actinolite series based on optical characteristics. Clinopyroxene porphyroclasts are sub-euhedral and commonly surrounded by amphibole, which belongs to the tremolite-

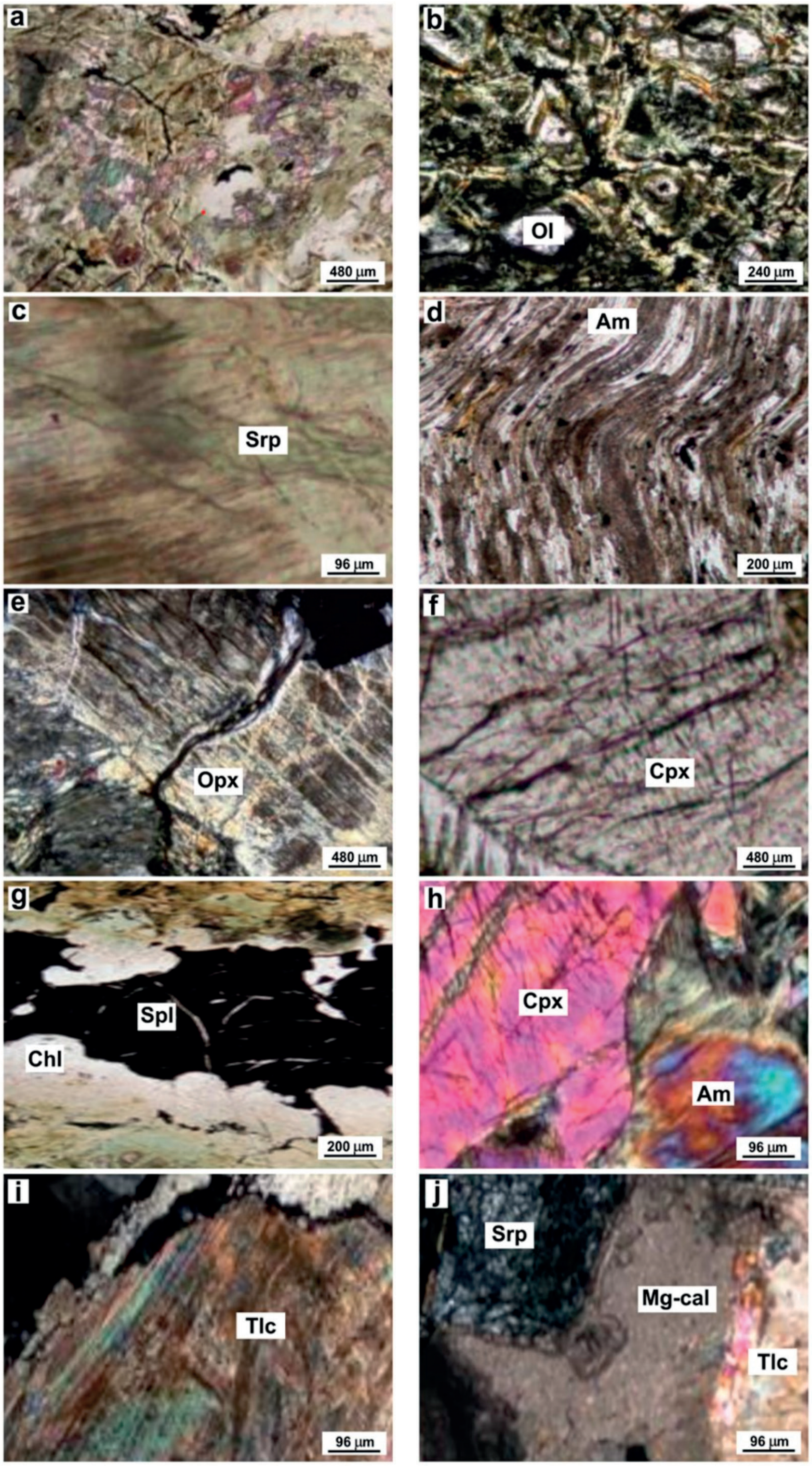

Figure 5. Representative photomicrographs of the Pietrapica serpentinite. a) pseudomorphic textures, lower polarizer; b) mesh texture with relicts of olivine crystals, crossed polarizers; c) cross-cutting vein with serpentine fibres lower polarizer; d) orthopyroxene deformed by kink fold band, lower polarizer; e) orthopyroxene crystal, crossed polarizers; f) relicts of clinopyroxene porphyroclasts 1N, 10X; g) spinel porphyroclasts, lower polarizer; h) clinopyroxene with tremolite rim (acicular amphibole) and chlorite; crossed polarizers; i) Mg-carbonate with talc and serpentine 2X, 1X; j) talc in carbonate vein with serpentine 2X, 1X. Legend: Ol: olivine; Srp: serpentine; Am: amphibole; Spl: spinel; Opx: orthopyroxene; Cpx: clinopyroxene; Tlc: talc. Mineral symbols are after Siivola and Schmid (2007).

actinolite series based on its optical characteristics (Figure 5g). Spinel forms red-brown coloured, xenomorphic, holly-leaf shaped porphyroclasts (Figure 5h). Talc is detected in some of the studied samples (Figure 3i) and is spatially associated with carbonate and serpentine veins (Figure 3j).

The results of the semi-quantitative XRPD X-ray diffraction analysis have shown that the mineralogical phases of the serpentinite samples mainly include serpentine (i.e. lizardite, antigorite and crysotile), Cr-chlorite, magnetite, tremolite, actinolite, pyroxene, and calcite. The most commonly occurring minerals are polymorphs of serpentine including lizardite (d=7.27 Å), crysotile (d=7.32 Å), and antigorite (d=7.23 Å). Amphibole minerals such as actinolite (d=8.31 Å) and tremolite (d=2.94 Å) were also detected in several samples (Figure 4). The presence of uvarovite in some samples is likely the result of a Ca-metasomatic process, in which the addition of Ca destabilises the iron stored in the iron-bearing minerals of the serpentinites (such as magnetite). favouring its formation (Plümper et al., 2014). Uvarovite formation may also be related to degradation of ortho- and clinopyroxene phases in the rocks.

In addition to X-Ray powder diffraction analysis (Figure 6), Raman spectroscopy was used to identify the main mineralogical phases present in the samples analyzed (Figure 7). The main sbestos minerals such as serpentine, chrysotile, lizardite and antigorite were identified based on their major peaks (Kloprogge et al., 1999; Rinaudo et al., 2003; Petriglieri et al., 2015; Militello et al., 2019). Serpentine showed the main peaks at 696 cm^{-1}, 394 cm^{-1}, 236 cm^{-1} at low wavenumbers, regarding the high

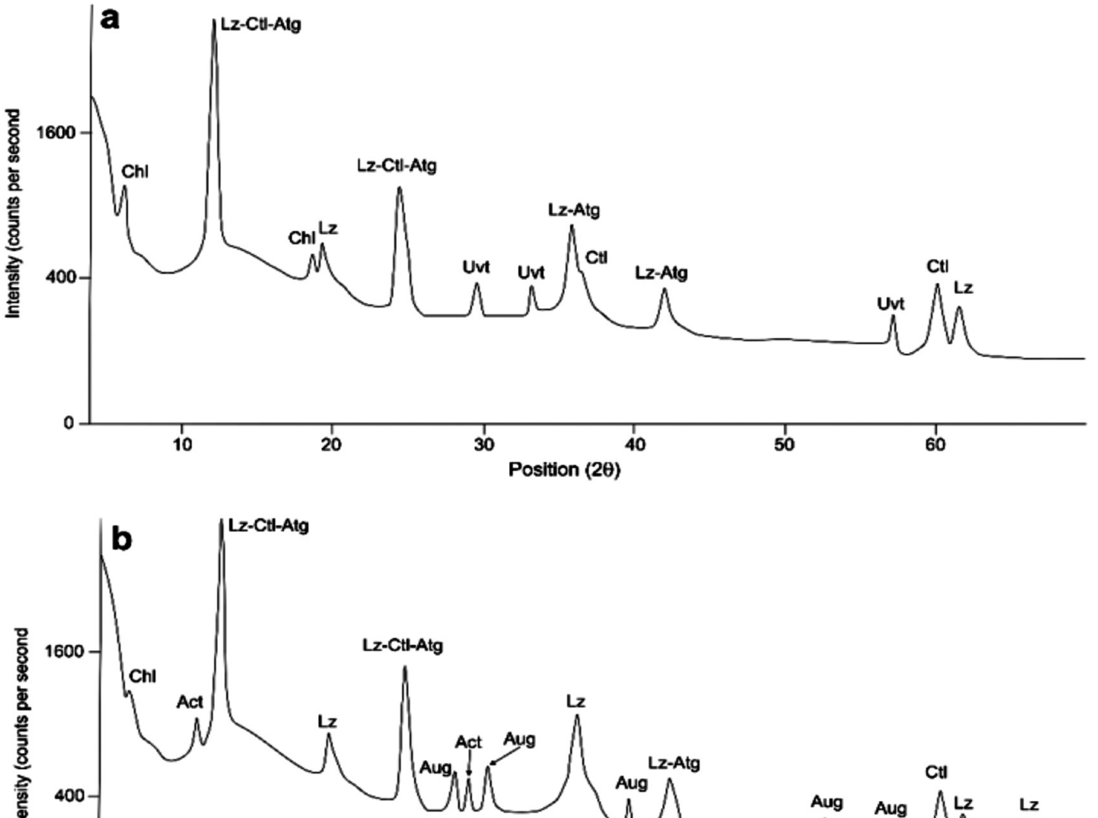

Figure 6. XRPD patterns of a massive a) and cataclastic b) serpentinite sample. Legend: Lz-Lizardite; Ctl-Chrysotile; Atg-Antigorite; Chl-Chlorite; Act: Actinolite; Aug-Augite; Uvt-Uvarovite.

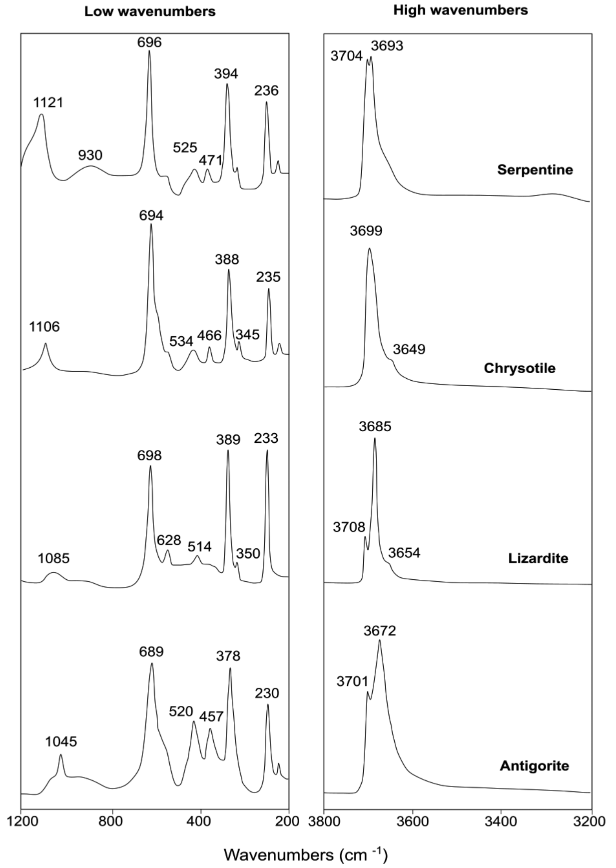

Figure 7. Raman spectra at low and high wavenumbers of Pietrapica quarry serpentinite samples.

wavenumbers we identified peaks at 3704 cm^{-1} and 3693 cm^{-1}. The chrysotile has the main low wavenumber peaks at 694 cm^{-1}, 388 cm^{-1}, 235 cm^{-1} while the high wavenumber peak was identified at 3699 cm^{-1}. The main low wavenumber peaks of lizardite were observed at 698 cm^{-1}, 389 cm^{-1}, 233 cm^{-1} while the main high wavenumber peak was identified at 3685 cm^{-1}. Finally, regarding the low wavenumbers, antigorite showed the main peaks at 689 cm^{-1}, 378 cm^{-1}, 230 cm^{-1} whereas the main high wavenumber peaks were identified at 3701 cm^{-1} and 3672 cm^{-1}.

Scanning Electron Microscope micromorphological analysis revealed either the typical lamellar habit of serpentine polimorphs such as antigorite and lizardite (Figure 8 a,b) with parts were separable serpentine fibres do occur (Figure 8 c,d); furthermore, the Fe-oxides mineral phases have been also observed (Figure 8 a-d).

DISCUSSION

This work aims to the mineralogical and petrographic characterization of the Pietrapica quarry serpentinites in the Pollino Massif. The petrographic and mineralogical study of serpentinites from the Frido Unit shows that Mg silicates, mainly serpentine and secondarily olivine, can be used for carbonation purposes. The use of serpentine rocks for CO_2 storage is an emerging technique in the field of carbon capture and storage (CCS). This technology aims to remove carbon dioxide (CO_2) produced by industries and power plants and deposit it underground, thereby reducing greenhouse gas emissions.

This paper also proposes to launch the idea for the suitability of natural analogue systems using different approaches in various sites in the Pollino area (Southern Apennines). Recent work has obtained precise estimates of the content of carbon-bearing minerals that could sequester atmospheric CO_2. Wilson et al. (2006) have shown that the amount of atmospheric CO_2 that has been bound in mineral form can be estimated from the Rietveld results for weight-percent abundance of hydrated magnesium carbonates. Wilson et al. (2006) carried out a study on serpentinite-rich sterile samples from Clinton Creek and Cassiar (Australia) and it has been estimated that a total of 164,000±16,400 tons of CO_2 are bound within the Clinton Creek sterile pile (applying a relative error of 10% corresponding to 4.36 wt% of hydro magnesite). However, this high degree of accuracy is difficult to obtain with other quantitative phase analysis methods (Wilson et al., 2006).

Large-scale application of mineral sequestration can represent high potential to fund remediation of mine sites and to provide reduction in greenhouse gas emissions. This refining technique could be applied to serpentine samples allowing for improved characterization of many tailings' materials and quantification of CO_2 sequestration. Recent work by Lavikko et al. (2016) have shown the importance of mineralogical and petrographic studies to improve the mineral carbonation process. The hypothesis that lizardite is more prone for carbonation than antigorite was initially thought to be incorrect, but it was on the right track because the characteristics that positively influence the suitability of a serpentinite for mineral carbonation are more common in lizardite than in antigorite. In contrast, amphiboles and pyroxenes have already been reported to be unsuitable for mineral carbonation (Sjöblom and Eklund, 2014; Byggmästar, 2011).

Characteristics that influence the suitability of a serpentine such as mineralogical structure, parent rock, and subsequent rock transformation through metamorphosis must be considered when working with carbonation processes. These integrated studies can be useful for selecting injection sites on a field scale and potential additives or pre-treatment strategies to optimize carbonation reactions of Mg-silicate metals.

Therefore, the petrographic and mineralogical study of natural occurring serpentinite rocks hosted in ophiolite

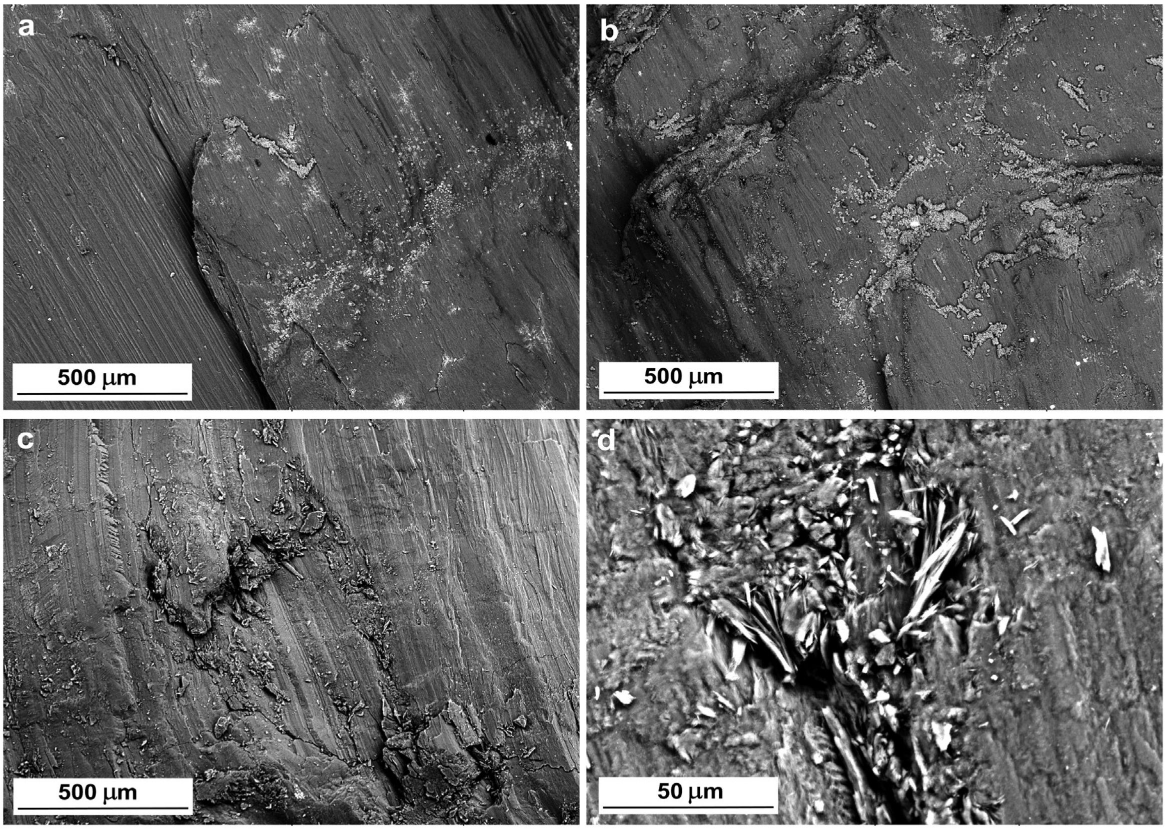

Figure 8. SEM images of selected serpentinite samples highlighting the microstructural features and the different habits of serpentine minerals, together with presence of Fe-oxides. a) and b) massive portion of the specimens, from which cleavage fragments are prone to split; c) tiny serpentine fibres occurring in the micro-fractured parts of the main matrix and d) chrysotile separable fibres.

sequences cropping out in the southern Apennines of Italy can serve as a starting point for the study and analysis of natural materials that can be used for CO_2 storage. Therefore, it is crucial to conduct extensive research in this area to develop more efficient and sustainable methods for reducing carbon emissions and mitigating climate change.

CONCLUSIONS

Petrographic analysis, XRPD, SEM and Raman spectroscopy revealed that the serpentinites of Pietrapica quarry (Pollino Massif, southern Italy) show a mineralogical composition consisting of olivine, pyroxenes and spinel with the presence of secondary minerals such as serpentine, chlorite, magnetite, prehnite, and amphibole. The mineralogical phases of the serpentinite samples mainly include serpentine (i.e. lizardite, antigorite and chrysotile), chlorite, magnetite, tremolite, actinolite, pyroxene, and calcite.

In the last years, the prospects of using serpentine rocks for CO_2 storage are promising. The market for CO_2 storage and sequestration that is developing in parallel with the European regulated market is an economically promising sector. The growth of this market is based on the prospects for the development of capture and storage technologies over the next decade.

CO_2 storage in rocks, particularly in serpentine rocks, is important for several reasons:

1. First, this technique allows some of the carbon dioxide in the atmosphere to be isolated and injected into deep reservoirs reducing the amount of CO_2 in the atmosphere, and hence helping to mitigate the greenhouse effect and global warming. CO_2 storage can facilitate the energy transition process.
2. As we move toward cleaner energy sources, CO_2 storage can help manage residual CO_2 emissions.
3. Deep reservoirs that were previously occupied by gas and/or oil can be used for CO_2 storage since these reservoirs are able to naturally contain gas and fluids, making them suitable for CO_2 storage.

Some projects are currently exploring the possibility of turning CO_2 into rock, a process that could offer a long-term and safe method of storage. However, it is important to note that these techniques are still under development and require further research to be implemented on a large scale. Each contribution aimed at providing detailed geological and petrographic features of natural outcrops may add a new piece of information towards sustainable solutions for greenhouse gas reduction.

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