Serpentinite enigma of the Rakhabdev lineament in western India: Origin, deformation characterization and tectonic implications

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Serpentine mineralogy controls fault rheology in the ocean and continental rift settings to subduction settings and hence can be used to discern the paleo deformational conditions. The Rakhabdev lineament from Rajasthan, India, provides a unique opportunity to understand its tectonic evolution inferred from the deformation microstructures. However, the complexity of surrounding calc-silicate rocks had resulted in a long-driven debate on the origins of these serpentinite rocks. The source rocks of the serpentinites also cannot be determined previously due to complete serpentinization and metasomatism rendering complete alteration of the source rocks. In this study, the serpentinite mineral was analyzed using Raman spectroscopy to accurately characterize its molecular structure. The presence of the antigorite-variety of serpentine mineral indicate towards the origin of Rakhabdev serpentinites in the upper mantle condition. The antigorite serpentinite of Rakhabdev is a hydration product of mantle materials showing high Mg# values obtained from EPMA data. The microstructural and EBSD analysis also indicates two stages of deformation, with deformation of antigorite at upper mantle conditions, followed by their shallow crustal carbonate metasomatism and subsequent deformation of the carbonates, with later stage calcite vein intrusion. This resulted in the appearance of antigorite in contact with calcite, dolomite, talc, tremolite, and chlorite. The exhumation of mantle wedge antigorite serpentinite is, therefore, indicating a paleo-subduction zone culminating in a crustal-scale collision boundary expressed as arcuate discontinuous bodies forming the Rakhabdev lineament.

Keywords: Rakhabdev serpentinite, Antigorite deformation, Calcite CPO, Raman spectroscopy

INTRODUCTION

Serpentinites typically form due to the hydration of peridotite at temperatures ranging from ~ 200 to ~ 600°C, resulting in the alteration of Mg-rich olivine and orthopyroxene to serpentine minerals (Evans et al., 2013). The common serpentine minerals chrysotile, lizardite, and antigorite have similar chemical composition, yet very different crystal structure (Wunder et al., 2001; Rinaudo et al., 2003). Chrysotile has a coiled structure responsible for its asbestiform properties, while lizardite has a planar structure, and antigorite has a wave-like structure (Wicks and Whittaker, 1975; Uehara, 1998). Structural type of

serpentine mineral present provides insights into the depth of serpentinization (Wunder et al., 2001; Bromiley and Pawley, 2003), and the microstructures developed within them help in characterization of the deformation processes activated in the region of their occurrence. The antigorite, formed as a result of serpentinization of mantle peridotites, is stable in the upper mantle condition. Their formation occurs in an MSH (MgO-SiO₂-H₂O) or MASH (MgO-Al₂O₃-SiO₂-H₂O) system (Wunder et al., 2001). The antigorite develops from water stored in mantle conditions in the high *P-T* stability field of the serpentine, primarily by slab dehydration below mantle wedge overlying a subducting slab (Arai and Ishimaru, 2008; Debret et al., 2019; Uno and Kirby, 2019). The unique alternating wave structure characteristic of antigorite and its crystal preferred orientation (CPO) are con-

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trolled by the original olivine fabric of the mantle wedge (Boudier et al., 2009) and acts as a controlling factor in fluid flow pathways and seismic anisotropy of the subduction zones (Hirauchi et al., 2010; Kawano et al., 2011; Campione and Capitani, 2013; Mizukami et al., 2014).

The studies on antigorite deformation, however, are limited to recent subducting slab (Andreani et al., 2007; Hirauchi et al., 2010; Plissart et al., 2019), and studies on paleo-subduction zones have been rarely investigated especially the completely metasomatized paleo-subduction zones. Since, different serpentinite minerals can deform at variable depths, identifying the chemical and structural variety of the serpentine minerals help in their characterization as metasomatized mantle wedge materials. The study of such serpentinite bodies can help in the understanding of the conditions of the metasomatism, deformation mechanisms, and tectonic evolution of the subduction zone. Such paleo-subduction zones are major components of the orogenic belt that stitch older cratonic blocks and occurs within present-day continental crust.

One such orogenic belt is the Aravalli-Delhi Mobile Belt (ADMB) in the state of Rajasthan, India, where few studies (Deb et al., 1989; Sarkar et al., 1989; Sugden et al., 1990; Verma and Greiling, 1995; Deb and Thrope, 2001; Singh et al., 2010; Purohit et al., 2015; Tiwari and Biswal, 2019) have reported the occurrence of serpentinites within Proterozoic meta-sediments. The fragmented bodies of serpentine rock occur along Rakhabdev lineament between the Jharol Group and the lower-middle Aravalli Group of rocks. However, due to a lack of studies in chemical characterization and complexity of the deformation and metamorphism in the area, the origin of these rocks is still an enigma. Two prevalent theories about their origin are the serpentinitization of structurally-controlled ultramafic intrusions and the serpentinitization in the paleo-subduction zone (Sarkar et al., 1989; Verma and Greiling, 1995). Lack of evidence and dearth in studies have rendered both the theories debatable (Sugden et al., 1990; Abu-Hamattah et al., 1994; Gathania et al., 1995; Shekhawat et al., 2010).

The present study aims at identifying the molecular structure of the serpentine mineral specimen present within the serpentinite bodies to give insights into its origin. The study of deformation microstructures is also attempted to determine deformation conditions and their relation to the Aravalli-Delhi orogeny (Prasad et al., 1998). Few studies have been undertaken based on microstructures and geochemistry of the antigorite (Bhu et al., 2006; Purohit et al., 2015). However, accurate characterization of serpentinite and usage of EBSD to determine deformation mechanisms have not yet been reported from this area. This could help in elucidating the

origin of the serpentinites which is present as arcuate discontinuous bodies within the metamorphosed Aravalli Supergroup of the ADMB.

GEOLOGICAL BACKGROUND AND SAMPLE LOCALITY

Rakhabdev (or Risabhadev) serpentinites constitute a series of continuous to detached (at places) bodies, occurring with a linear N-S trend for a distance of about 90 km between Gogunda in the north and Antri in the south, that defines the Rakhabdev lineament (Fig. 1), within the Aravalli Supergroup (Bhu et al., 2006). The ADMB comprises two major supracrustal units, viz., the Aravalli Supergroup and the Delhi Supergroup, and deposited on an Archaean basement known as the Banded Gneissic Complex (BGC) (Gupta et al., 1995; Heron, 1953; Roy and Jakhar, 2002; Sharma, 2010). The Aravalli Supergroup has been subdivided into the Lower, Middle and Upper Aravalli Groups (Roy and Jakhar, 2002). The lowest part of the succession is dominated by stromatolitic phosphorites and U-bearing black shales along with metabasalts. The middle part mostly comprises greywacke, carbonaceous phyllite, and conglomerate, and the upper part is dominantly quartzite, conglomerate, and phyllite which grades into mica schists with thin quartzite bands in the western side. Roy and Jakhar (2002) has designated the dominantly phyllite-mica schist-quartzite units as Jharol Group – a deep water facies, in contrast to the shelf facies associations dominating the eastern part of the Upper Aravalli Group (Gupta et al., 1997). The boundary between Jharol Group and Upper Aravalli Group is roughly marked by the occurrence of the serpentinites of Rakhabdev lineament (Bhu et al., 2006; Purohit et al., 2015), with type locality around Risabhadev area. The present study focusses on serpentinite samples collected from Gogunda and Risabhadev area within the Rakhabdev lineament (Fig. 1). A total of six serpentinite samples were collected (Table 1 and Fig. S1; Fig. S1 is available online from <https://doi.org/10.2465/jmps.191016>).

ANALYTICAL METHODS

The thinsections (~ 30 µm) were prepared along XZ section (perpendicular to foliation and parallel to lineation) and polished with 1 µm diamond paste, for petrographic observations. For EBSD analysis, two chip samples (cut along XZ section) of 1 cm by 1 cm were polished using IS Polisher 1000 at Kochi Core Center JAMSTEC facility to minimize the surface damage during polishing, followed by osmium coating at Hiroshima University.

Figure 1. Geological map of the Aravalli-Delhi Mobile Belt, showing the occurrence of Rakhabdev serpentinites (Modified after Sugden et al., 1990). Inset shows the location of the study area. The sampling points are shown in the two enlarged areas of the sample locality marked by black arrows (modified after Purohit et al., 2015). Google images and GPS values of sampling locations (WGS84 projection system) are included in Figure S1 and Table 1 respectively. The preliminary structural data (foliation and lineation of serpentinites and crenulation orientation) is presented in Figure 3.

Table 1. Sample locations and photograph locations (GPS values WGS84 projection system)

Sample locations			
Sample No.	Latitude (N)	Longitude(E)	Area
ACUD1403	24.7931	73.5712	Gogunda
ACUD1404	24.7931	73.5712	
RK 10	24.7897	73.5657	
RK 11	24.0784	73.7024	Rakhabdev
RK 12	24.0677	73.6913	
RK14	24.0463	73.6891	
Photograph location			
Figure No.	Latitude (N)	Longitude(E)	Area
Figure 2a	24.792627	73.571215	Gogunda
Figure 2b	24.078350	73.702373	Rakhabdev

Principle of Raman spectroscopic analysis of serpentinite minerals

Raman spectra of serpentines in low wavenumber spectral range (150–1100 cm^{-1}) corresponds to the inner vibrational modes of the lattice and to Si–O₄ vibrations,

while the high wavenumber spectra (3550–3850 cm^{-1}) corresponds with the stretching vibrations of the OH groups of the water molecules (Petriglieri et al., 2015). Low wavenumber Raman spectra obtained shows strong peak of 230 cm^{-1} , which is present in all serpentine polymorphs due to O–H–O vibrations, O being the non-bridging oxygen of a SiO₄ tetrahedron and H being the hydrogen of the outer OH group of the adjacent layer (Griffith and Wickins, 1967; Loh, 1973; Petriglieri et al., 2015). The peak at 680 cm^{-1} , due to deformation vibrations of the Si–O layer, the peak at 1044 cm^{-1} , due to antisymmetric stretching mode (ν_{as}) of S–O_b–Si groups, and the doublet at 3665 cm^{-1} and 3695 cm^{-1} , implies the presence of antigorite variety of the serpentine mineral in the collected serpentinites (Auzende et al., 2004; Enami, 2006).

Raman Spectroscopic analyses of two thin sections representative of the serpentine minerals were conducted by JASCO NRS-5100 Raman Spectrometer at the Geodynamics Research Center (GRC) facility of Ehime University. The analysis was done using 532 nm Nd-YAG laser and CCD detector. Collection times for Raman spectra were 6 or 12 accumulation of 10 s each.

Figure 2. Outcrops from Jharol belt and Aravalli belt. (a) Calcite bearing talc-tremolite schist inset showing the development of sub-vertical foliation in the rock. (b) Deformed serpentinites occurring sporadically between talc-tremolite schists and pelitic schist. The crenulated nature visible in outcrop is a product of shallow crustal deformation overprinting the pre-existing foliation of the serpentinites.

Electron Micro Probe Analysis

The major element chemical compositions of the serpentines and the associated minerals including opaque minerals were done by EPMA (JEOL JXA-8200) at Natural Science Center for Basic Research and development (N-BARD) facility of Hiroshima University with accelerating voltage of 15 kV and 2 μ m beam size. ZAF method was applied for matrix corrections.

Electron Backscatter Diffraction Analysis

The crystallographic preferred orientation (CPO) was obtained point by point, for the calcite and antigorite by electron backscattered diffraction (EBSD) using the Nordlys system attached to scanning electron microscope JEOL JSM6390A, with accelerating voltage of 15 kV and sample tilt of 70°. The obtained EBSD patterns were processed using the HKL Channel 5.0 software package.

RESULTS

The serpentinites occur in the field as detached bodies bounded by metapelites in the west and talc-tremolite schists (Fig. 2a) in the east. The actual contact of the metapelites and talc-tremolite schists with the serpentinites could not be identified in the studied area. The detached serpentinite bodies show a trend of NNE-SSW with subvertical foliation overprinted by crenulation cleavage trending NE-SW (Fig. 2b). The preliminary structural data of the foliation and lineation obtained from the serpentinite bodies and the measured crenulation cleavages are presented in Figure 3. The serpentinite samples, in thinsections show two types of assemblages. One as-

Figure 3. Stereonet plot of foliation and lineation of the serpentinites with the crenulation data. Crenulation cleavages ($n = 6$) are plotted as great circles in the stereonet. The serpentinite foliations ($n = 17$) represented by great circles and stripping lineations (solid circles, $n = 8$) are plotted on them. The data is overlapped hence plotted data appears less than actual data.

semblage shows the presence of dominantly serpentine minerals (the type of serpentine mineral is identified using Raman spectroscopy) with opaque minerals (mineral composition determined by EPMA). The opaque minerals of this assemblage are primarily magnetite, with few relict Cr-spinel. The other assemblage comprises of serpentine minerals with calcite, dolomite, talc, tremolite, and chlorite. The opaque minerals of the second assem-

Figure 4. Raman spectra with x -axis as Raman shift (cm^{-1}) and y -axis as Raman intensity measured from representative serpentinites, the peaks at high wave number implies the antigorite variety of the serpentine minerals.

blage include dominantly magnetite and also include minor alteration to ulvöspinel. Cr-spinel is not present in the serpentinite samples with calcite veins. Also, in the currently studied samples, no olivine grains could be identified. The serpentines exhibit massive (with interpenetrating blades) to foliated textures, with clasts of dolomite interspersed with massive serpentine minerals (discussed in later sections).

Raman spectral analysis of serpentine

The serpentinites primarily exhibit massive and randomly oriented blades or foliated nature observable in thinsections. Raman spectra of serpentines in low wavenumber spectral range ($150\text{--}1100\text{ cm}^{-1}$) shows strong peak at ~ 230 , ~ 373 , ~ 680 , and $\sim 1041\text{ cm}^{-1}$ (Fig. 4). The high wavenumber spectra ($3550\text{--}3850\text{ cm}^{-1}$) shows the doublet at ~ 3665 and $\sim 3695\text{ cm}^{-1}$ (Fig. 4).

Microstructural characterization and CPO

The serpentinites contain dominantly antigorite and occur as massive or foliated. The massive antigorite shows the interpenetrative texture of the blades of antigorite (Fig. 5a). The Cr-spinel grains preserved in the massive antigorite are fractured and show gradational zoning (Fig. 5b) of progressive Fe-enrichment from the unaltered core towards the Fe-rich rim. They are present with the assemblage of interpenetrative antigorite without any secondary alteration minerals like chlorite. The massive antigorite is deformed into foliated antigorite (Fig. 5c), showing a slight variation in BSE contrast, corresponding to their variation in mineral content (composition). Few relict grains preserve the interpenetrative texture of the original antigorite and shape of original parent grains (Fig. 5d). The magnetite present in the foliated antigorite samples (Fig. 5e) does not exhibit any zoning. Prominent calcite veins (Fig. 5f) crosscut the foliation of the antigorite and

Figure 5. (a) XPL image of original antigorite shows interpenetrating structures. (b) SEM-BSE image of representative Cr-Spinel within the original antigorite shows Fe-enrichment from core to outer rim. (c) SEM BSE images of foliated antigorite. (d) Pseudomorphic grains preserving interpenetrative texture and showing alteration to foliated antigorite. (e) SEM-BSE image of magnetite grains in the foliated antigorite sample. (f) Thin section of foliated antigorite with later stage calcite veins cutting them at an angle. (g) Calcite vein crosscutting the foliated antigorite with assemblage of talc-tremolite-chlorite. (h) The XPL image of calcite veins [marked by rectangle in (f)], with recrystallized grains (arrows) and larger grains showing twin development and undulose extinction.

are deformed by twinning and recrystallization (Figs. 5g and 5h).

The EBSD analysis of the foliated antigorite (Fig. 6) obtained from XZ sections of the samples, exhibit well developed CPO (Fig. 7a), with multiples of uniform dis-

Figure 6. Representative area showing foliated antigorite grains for EBSD analysis.

tribution (m.u.d) values 0.03–5.46. The $[010]$ axes of antigorite are concentrated parallel to the lineation and $[100]$ axes subnormal to lineation. This CPO pattern suggests the activation of $[010] (001)$ slip system in antigorite. The calcite within the veins crosscutting the foliated antigorite has also developed a weak but distinctive CPO (m.u.d. 0.19–2.15), with $[0001]$ axes parallel to vein direction and $\langle 11-20 \rangle$ axes spread normal to the vein (Fig. 7b). This CPO pattern is due to the dislocation creep with (0001) as the dominant slip plane for calcites deformed at mid- to high temperatures (de Bresser and Spiers, 1993; Austin et al., 2008; Vauchez et al., 2015).

Mineral Chemistry

The detailed compositions of the antigorite (inferred by the textural similarity with the representative antigorite grains determined by Raman spectroscopy) are listed in Supplementary Table S1 (available online from <https://doi.org/10.2465/jmps.191016>).

The major total oxide of the antigorite ranges 84.58–88.25%. Recalculations of atomic proportions were done

Figure 7. (a) Lower hemisphere pole figures of CPO of antigorite, shows development of $[010] (001)$ slip system in the antigorite. (b) Lower hemisphere pole figures of CPO of deformed calcite veins, showing activation of $[0001] \langle 11-20 \rangle$ slip system.

Figure 8. (a) Mg versus (Fe + Mg) plot for the antigorite samples, showing two distinct clusters with higher Mg#(95–98) for original antigorite and a Mg#(88.1–91) for antigorite in contact with calcite/dolomite veins. (b) Mg#[Mg/(Mg + Fe)] versus Cr# [Cr/(Cr + Al)] for the chromian spinels shows compositional affinity of the spinel in the common region of forearc and abyssal peridotites (Gamal El Dien et al., 2019).

based on the seven oxygen and yields Mg number (88.80–91.13) for samples containing calcite and dolomite. The antigorite from zones without calcite and dolomite assemblage shows a higher Mg number (93.78–98.70). No zoning within the individual antigorite grains was observed in SEM BSE, however, there is a variation in the Fe content of all the antigorite samples. The Mg versus (Fe + Mg) plot (Fig. 8a) shows clustering in two distinct zones, representing alteration in the antigorite chemistry in response to the presence of carbonate phases. The representative composition of the talc, chlorite, and spinel are also listed in Table S1. The mineralogical variation in Cr-spinels is inferred by plotting its Mg number against its Cr number (Fig. 8b) and shows the compositional affinity of the spinel in the common region of forearc and abyssal peridotites (Arai and Ishimaru, 2008; Gamal El Dien et al., 2019).

DISCUSSIONS

Generation and deformation of antigorite

The absence of mantle minerals like olivine and pyroxenes within the observed samples renders the determination of source-rock mineralogy difficult. However, the predominance of only antigorite within the serpentinite (inferred from Raman spectroscopic analysis), and the mineralogy of the Cr-spinels (Fig. 8b) suggest the possibility of forearc peridotites as the source rock (Gamal El

Dien et al., 2019).

The interpenetrating microstructures of the original antigorite (Fig. 5a) that is also preserved in pseudomorph (Fig. 5d) shows deformation into oriented blades of antigorite forming the foliated antigorite (Figs. 5c and 5d). Since, all other phases are post to this massive antigorite and the foliated antigorite, therefore the serpentinization is a product of olivine + orthopyroxene system at pressure >1.5 GPa (Nakatani and Nakamura, 2016). The hydration of olivine and orthopyroxene resulting in the formation of antigorite is given by the following reaction (Pawley, 1998; Murata et al., 2009; Watanabe et al., 2011; Auzende et al., 2015).

The presence of the Cr-Spinel as opaque mineral (without any hydrous phases like chlorite) and the Mg-rich antigorite (Fig. 5b) also suggest a high temperature (400–600 °C) for the serpentinization, accommodated by compositional adjustment of olivine at higher rates of Mg-Fe diffusion (Evans, 2010; Evans et al., 2012). The subsequent Fe rich alteration of the Cr-spinel and presence of Fe-oxides and Fe-Ti oxides suggests their mobilization in later metasomatic process, post to the serpentinization of the peridotites (Gamal El Dien et al., 2019; Nozaka, 2018).

The CPO pattern in the foliated antigorite (Fig. 7a)

Figure 9. Schematic diagram showing the stages of formation and ensuing deformation of the Rakhadev serpentinites. The generation of random blades and interpenetrative antigorite as a result of hydration of mantle peridotites at high pressures, followed by deformation into foliated antigorite. The foliated antigorite underwent metasomatism and alteration resulting in formation of talc-tremolite and calcite, with dolomite and chlorite. Calcite vein injection postdate all other deformation and fills into the fractures during exhumation.

represents the antigorite CPO pattern common with other antigorite samples reported from forearc mantle body (Nishii et al., 2011), with the *c*-axis perpendicular to the foliation and the *b*-axis parallel to the stretching lineation. The σ_1 direction being parallel to foliation suggests horizontal orientation. The antigorite deformation behavior is controlled by the structural relationship between olivine in peridotite and antigorite, which again depends on the fluid flow pathways controlled by the geometry of the initial olivine fabric and the topotactic relationship between olivine and antigorite (Watanabe et al., 2011). Few pseudomorphs of antigorite are present (Fig. 5d). However, in the absence of parent mantle mineral grains such inferences cannot be drawn from these samples.

Metasomatism and deformation at shallow crust

The presence of calcite + talc + tremolite + chlorite + dolomite assemblage suggests secondary metasomatism of already-hydrated mantle peridotite at a shallower depth, with high CO_2 partial pressure (Schandl and Naldrett, 1992; Soda and Takagi, 2010) leading to alterations of the relict forsterite grains associated with bladed antigorite. This secondary metasomatism led to the generation of calcite, talc-tremolite, and chlorite within the weak planes of antigorite. Deformation at a shallower level has resulted in the development of weak crenulation of foliated antigorite and alteration to talc-tremolite and chlorite in the microlithon domains. The variation in Al content (Table S1) of the massive and deformed antigorite in contact with late calcite/dolomite, implies a possible variation in mineralogy of the parent mineral. The calcite veins cutting the foliated antigorite and the talc-tremolite grains suggest their injection at a later stage of CO_2 metasomatism postdating the antigorite-forming hydration event, during shallow crustal deformation. The CPO of calcite within the veins, suggests the develop-

ment of $c\langle a \rangle$ as dominant slip system (Fig. 7b), as a result of plastic deformation of calcite at $>250^\circ\text{C}$ (Burkhard, 1993). The orientation of maximum principal stress (σ_1) axes at shallow crustal deformation is represented by the orientation of the *c*-axes of calcite crystals (Fig. 7b) (Kamb, 1959; Romeo et al., 2007).

TECTONIC SCENARIO

The antigorite serpentinites of the Rakhadev lineament record the deformation in three stages (Fig. 9). The first being the deformation of antigorite in upper mantle conditions (Fig. 5c). The deformed antigorite underwent exhumation and further shallow crustal CO_2 rich metasomatism resulting in the alteration of serpentinites into talc-tremolite-calcite-dolomite-chlorite assemblage. This was followed by intrusion of calcite veins (at a later stage of CO_2 metasomatism), cutting across all other features (Fig. 5f). No relict upper mantle minerals except Cr-spinels (with Fe-enrichment towards the rim) are observed in the analyzed samples since the entire rock is metasomatized. The exhumation of the hydrated upper mantle material and its subsequent shallow crustal metasomatism is possible during ongoing subduction, along the exhuming channels at the interface of the subducting slab and mantle wedge. Also, since metamorphosed deep-sea sediments are reported from the western margin of the presently studied antigorite exposures (Roy and Jakhar, 2002), it further supports the idea of Rakhadev being a tectonic lineament preserving hydrated mantle rocks. Further studies are required on the regional deformation pattern of the Rakhadev lineament to elucidate the related tectonic scenario. The timing of this tectonism is also important to determine the integrated tectonics of this paleo-subduction zone and the eventual closure of the paleo-ocean in the overall evolution of ADMB.

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SUPPLEMENTARY MATERIALS

Supplementary Figure S1 and Table S1 are available online from <https://doi.org/10.2465/jmps.191016>.

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