

Raman spectroscopic study of natural cordierites and indialites in the metamorphic aureole of Mishow, located in the southwest of Marand, East Azerbaijan Province, Iran

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Abstract

The contact aureole of Misho is about 1 km wide around Cadomian igneous intrusions and is located in the northwest of Iran near the city of Tabriz. The granite intrusion has injected into the Kahar formation, causing thermal contact in country rock ranging from the albite-epidote hornfels facies to the pyroxene hornfels facies. The Misho's aureole country rock, located south of Eyshabad, exhibits the following metamorphic stages in ascending order of metamorphic grade: 1) Cordierite Hornfels, Muscovite, and spotted slate chlorite 2) Biotite-Cordierite-Hornfels differentiates the two minerals from their initial occurrence in country rocks. 3) Garnet, Andalusite, Cordierite, Biotite, Hornfels, which are composed of porphyroblasts cordierite and andalusite with anatectic Migmatite, have partially melted during high-grade metamorphism. Due to contact metamorphism, the mineral cordierite, which composes the majority of pelites, has developed in large quantities throughout the aureole. The mineral cordierite is polymorphic and has a high temperature Cordierite is called Indialite, crystallizes as a hexagon. The Raman spectra of cordierite at specific distance from the intrusive granites have been analyzed in this paper. The substitution of Fe and Mg in the structure of orthorhombic cordierite is responsible for the shift in the Raman spectra of these cordierite toward lower wavelength bands at distances between 500 and 350 M from the igneous contact. This shift is characteristic of pure Mg-cordierite and is proportional to the concentration of Fe. These cordierites' Raman spectra exhibit a distinct band that is identifiable as indialite at a distance of 100 to 300 meters adjacent to the intrusive contact. Petrography investigations indicate that hexagonal cordierite crystallizes most likely as a result of melting cordierite around 700 C°. As a result of this melting, the cordierite structure becomes distorted.

Key words: contact metamorphism, cordierite, indialite, Raman spectra, Misho, Iran

Introduction:

The Ediacaren Misho granites and its metamorphic aureole are situated southwest of Marand City in northwest Iran, between the latitudes of $45^{\circ} 31' 5''$ to $45^{\circ} 38' 13''$ and the eastern longitudes of $38^{\circ} 22' 27''$ to $38^{\circ} 19' 48''$ (Fig1). According to the geological divisions of Iran, the study area is located in the Alboz-Azarbaijan zone. The research region is bordered to the north and south by two sub-parallel, dextral oblique-slip fault systems. The Cadominan Misho granite is exposed over an area of more than 50 km^2 , with a horseshoe-like body form. Leucogranitic and diorite constitute the majority of the Misho granites. According to previous studies, the S-type granites could have been associated with anatexis of the upper crustal materials, while the I-type granites are based on zircon U-Pb ages that imply early Cambrian (600–520 Ma) (Shahzeydi, 2011). The diabasic rocks (most likely Percambrian in age) that comprise up the Kahar Formation include the rocks that surround the Misho granites. Only a few of the Soltanieh and Barut formations can be found along the southern margin of the Misho granites. Contact metamorphism and the creation of different hornfels rocks, ranging from low-grade to high-grade metamorphism and anatectic migmatites, have been generated by the intrusion of granite into these rocks. Raman spectroscopy has been used in this study to evaluate the cordierite in the Hornfelses, with an emphasis on the distance from the granites that is intrusive. cordierites are abundant in the Percambrian rocks and rocks that are part of the Misho metamorphic aureole are known to contain these cordierites. Cordierite $[(\text{Mg,Fe})_2\text{Al}_4\text{Si}_5\text{O}_{18} \cdot m\text{H}_2\text{O} \cdot \text{CO}_2]$ is a significant Mg-Fe-Al containing Cyclic silicates that is stable in a variety of geological settings (e.g., Vry et al. 1990; Carrington and Harley 1995, 1996; Smith 1996; Kalt 2000; Harley et al. 2002; Bertoldi et al. 2004). According to Pereira and Bea (1994), it was frequently described as the result of subsolidous metamorphic events. Both orthorhombic and hexagonal crystals may form from cordierite. Indialite (P6/mcc), a high-temperature cordierite, took on a hexagonal crystal structure and remained stable at 1450 C° (Schreyer and Schairer 1961; Putnis 1980; Meagher and Gibbs 1977). Two distinct tetrahedral (T_1 and T_2) sites, one octahedral M site, and two types of channel sites (Ch_0 , $\text{Ch}_{1/4}$) make up the unit cell of low-grade cordierite, which has an orthorhombic structure (see Fig.2). $(\text{M})_2 (\text{T}_1)_2 (\text{T}_2)_6 2(\text{T}_2)_2 (\text{T}_2)_1 2 (\text{T}_1)_6 \text{O}_{18} (\text{Ch}_0, \text{Ch}_{1/4})$ is the formula for this expression. While another set of tetrahedra (T_1) with 1Si + 2Al atoms connects various hexagonal rings (Fig.2), four Si and two Al atoms form six-membered rings of (Si,Al) O_4 tetrahedra (T_2) along the crystallographic c-axis that define a channel-like structure (Gibbs 1966). Vibrational spectroscopy has been widely utilized to

investigate various cordierite features, including polymorphism (e.g., Langer and Schreyer, 1969; Farrell and Newnham, 1967).

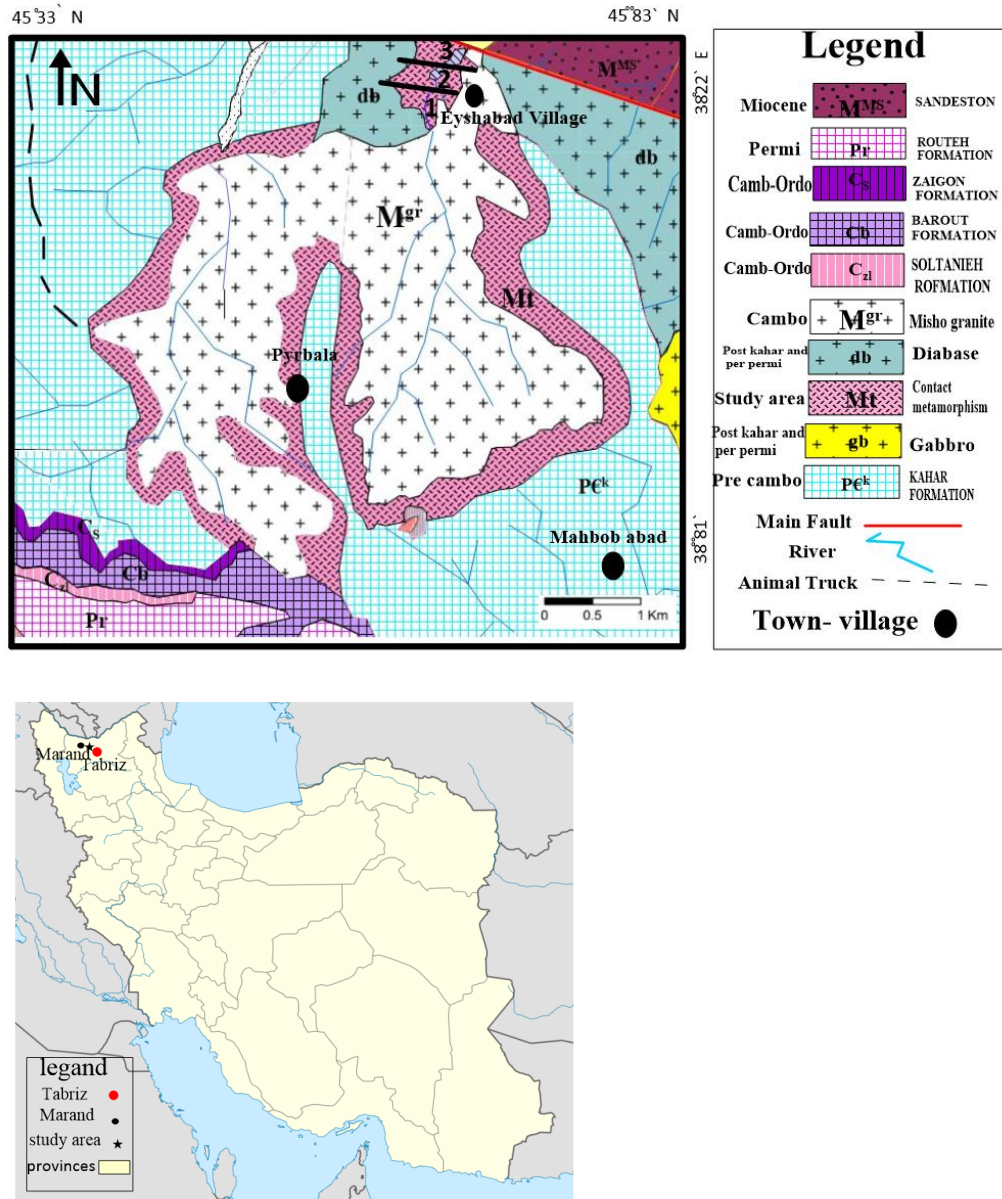


Figure 1: The study area's geological and geographical maps were created using the Geological Survey Organization's map.

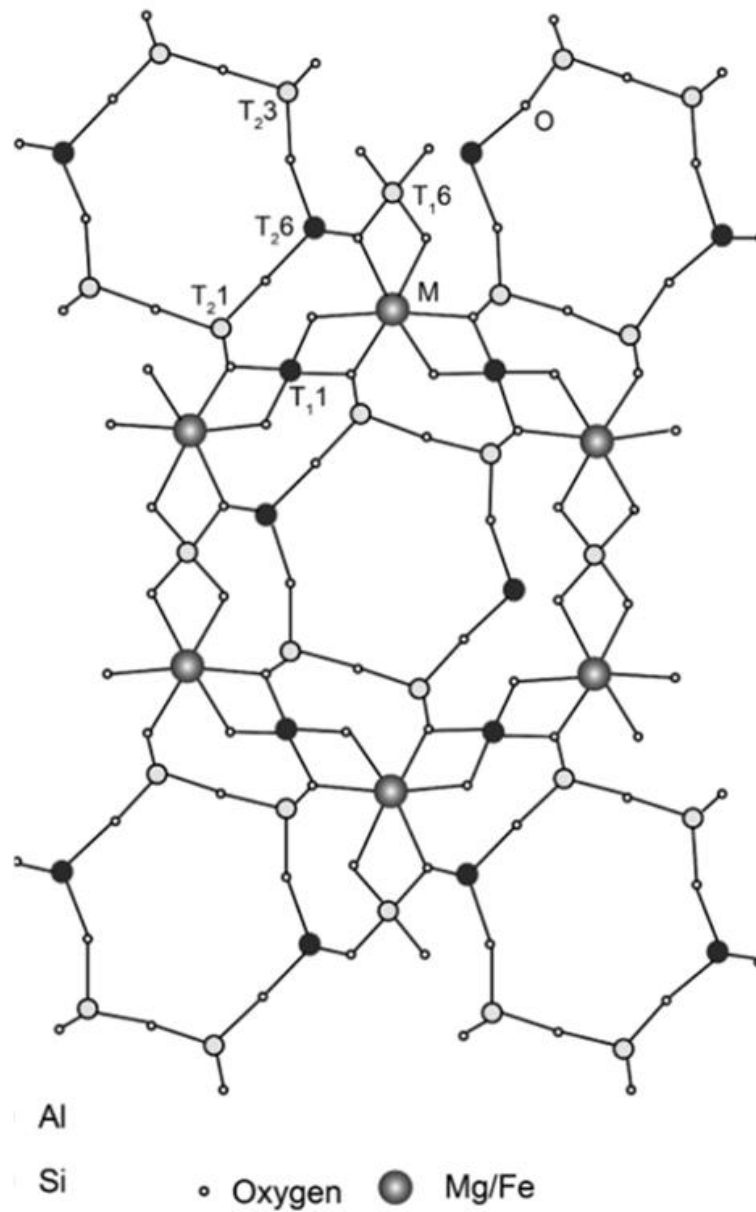


figure 2: The crystal structure of octahedral, tetrahedral and octahedral sites are depicted in the figure. Cohen et al. (1977).

Field Description and Geological study:

Fig. 3 depicts the contact that aureoles surround the Misho granites. Throughout the massif, the inner zone has a uniform overall aspect. There are fine to medium-grain hornfels and equigranular hornfels in this area. Its varied width spans between 100 and 200 meters. The hornfelses in this zone exhibit a gray to black coloration, which is remarkably comparable to other metamorphosed sediments in other areas. In the western and eastern regions of the granitic intrusive complex, where the metasediment outcrop is located close to the diorite massif, migmatitic rocks are most widely distributed. Oval-shaped cordierite crystals are aligned parallel to the existing regional metamorphic fabric (D1 schistosity). In the hand sample The splitting of cordierite-rich (with or without andalusite) layers is typically one of the boudinage structures linked to leucosome formation. Certain areas within the leucosome's boundary clearly contain different leucocratic minerals (K-feldspar, quartz) (fig. 3,4,5). Incoming index minerals have been used to identify metamorphic zones.

I chlorite zone (present at lowest exposed grades), II chloritoid-cordierite zone, III biotite zone, IV staurolite zone, V andalusite zone, VI K-feldspar and melting zone, VII garnet and melting zone, VIII sillimanite and melting zone. The mineral cordierite is abundantly observed in these contact metamorphism rocks, which in petrographic studies include various rocks such as slate, phyllite, and hornfels. Cordierite is a major constituent of the assemblages in zones II– VIII.

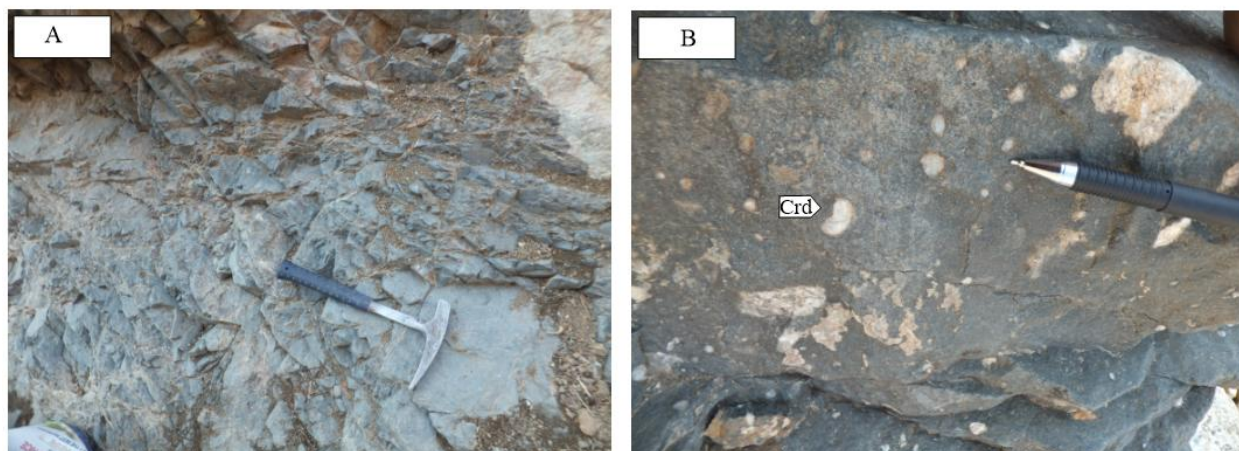


Figure 3: A) Outcrop of spotted slate near the village of Eyshabad. B) Distribution of oval-shaped cordierite spots located 400 meters away from the mass. This represents the first appearance of cordierite, with the size of the cordierite crystals reaching up to 1 centimeter.

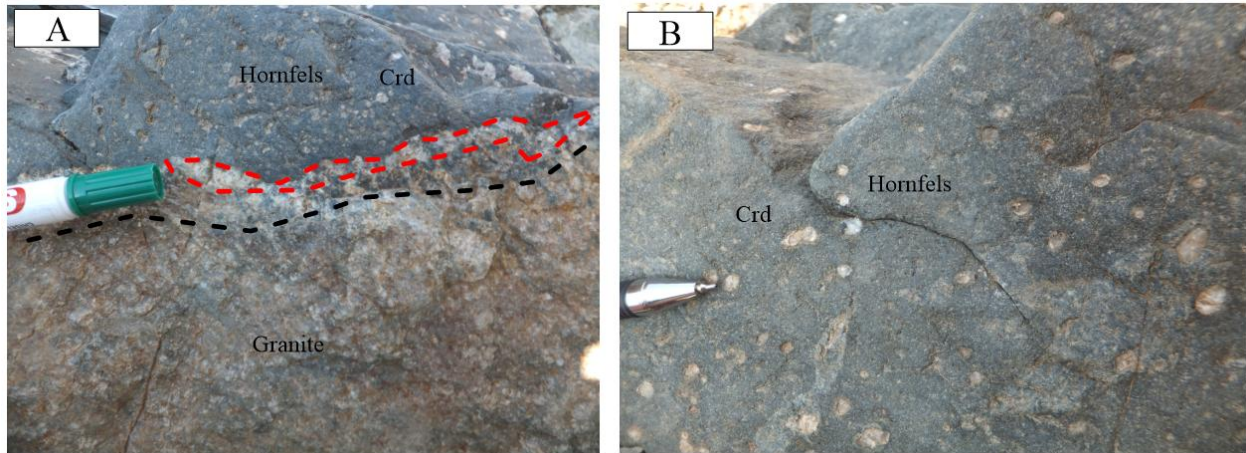


Figure 4: A, B) High-temperature rocks designated Hornfels are found in the area before high-grade intermediate and high-grade migmatites (Figure 4: A, B). Parallel to the current regional metamorphic fabric (D1 schistosity) are oval-shaped cordierite crystals. A random orientation is seen on the cordierite oval surfaces. Veins of quartz show when melting has begun.

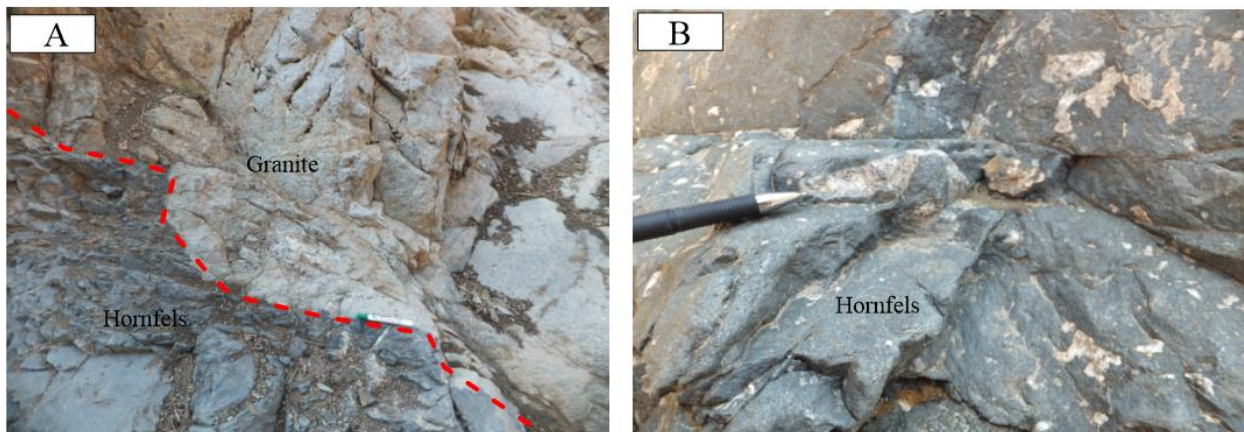


Figure 5: A, B) (A) Andalusite, biotite, cordierite hornfels are found close to the intrusive massive (B) shows leucosome veins, which are thought to be the initial indication of the rocks' migmatitic activity



Figure 6: The metapelite's primary layering is obvious on the image's right side. Cordierite-rich layers on the left are separated from leucosomes with a boudinage structure in the central region.

Contact metamorphism rocks from low grade to high grade are as follows:

1) **Spotted slate or chlorite-muscovite-cordierite ± And Hornfels:** The granite and granodiorite plutons of the Ediacharan have intruded low-grade metamorphic rocks, including the Kahar Formation slates. White spots in the slates at a distance of 500 meters from the intrusion in the outer zone of the Misho metamorphic aureole are thought to be the earliest indication of contact metamorphism. Muscovite, cordierite, and chlorite constitute the majority of the mineral assemblage. The mineralogy of the matrix inside the spots is identical to that outside the spots in the thin section. The oval-shaped cordierite, which forms the spots, ranges in size from 0.5 to 1 m. Nevertheless, it seems that the cordierite has rehydrated and changed into muscovite aggregates with fine grains. Cordierites show random orientation. Cordierite dots are still visible at longer distances, but they are found in the interior. However, even andalusite is seen in the interior of the speckled slate zone. These patches seem to be pseudomorphs, which show that one mineral has been replaced by another while maintaining its original form (Winter, 2001). This rock has a

porphyroblastic texture, and the cordierites have changed into quartz and muscovite at the edges due to the infiltration of pneumatolytic fluids in the region (a process known as pinitization). (Fig.7)

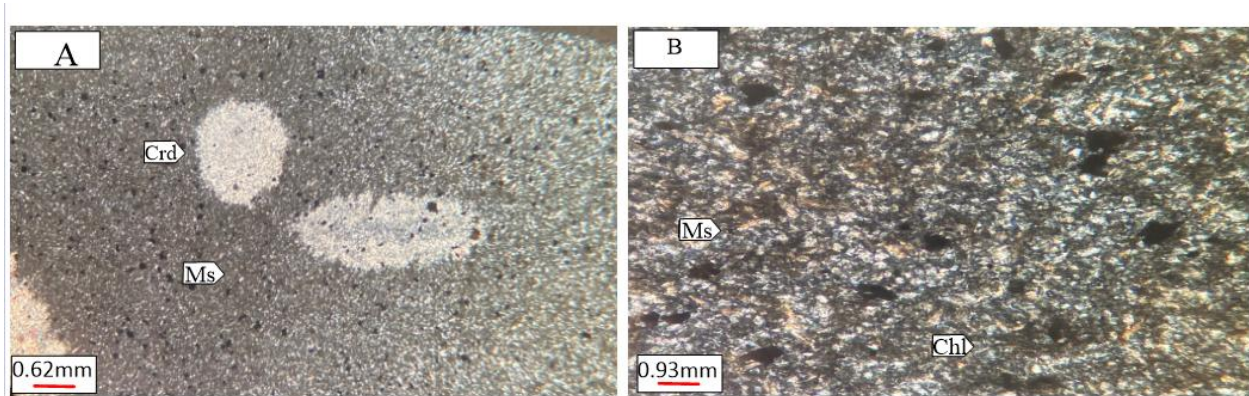


Figure 7 A: Spotted slate or chlorite-muscovite-cordierite Hornfels : cordierite spot shows on a thin section of a host slate. B: The slate's matrix contains the minerals muscovite and chlorite.

Cordierite biotite hornfels: As the degree of metamorphism increases, the mineral assemblage of cordierite, biotite, and quartz appears while, the proportion of muscovite and chlorites significantly reduces. The modal quantity of chlorites falls and the modal amount of biotite and cordierite increases as we get closer to the intrusive border, which is between 450 and 400 meters away. The first contact metamorphic mineral found in the Kahar sedimentary host rock is biotite. Low-grade regional metamorphism caused the biotite to crystallize in a platy form along the Kahar protolith's major layering. Cordierites appear as subhedral crystals that are comparatively large. Usually, there is a lot of biotite present with these cordierites. Numerous biotite, quartz, and black mineral inclusions can be seen in the intact cordierite porphyroblasts. The growth of the porphyroblast coincided with the trapping of these inclusions. The cordierite porphyroblast, with a size of 2 m, are extended along the cleavages and have a poikiloblastic texture . (Fig.8)

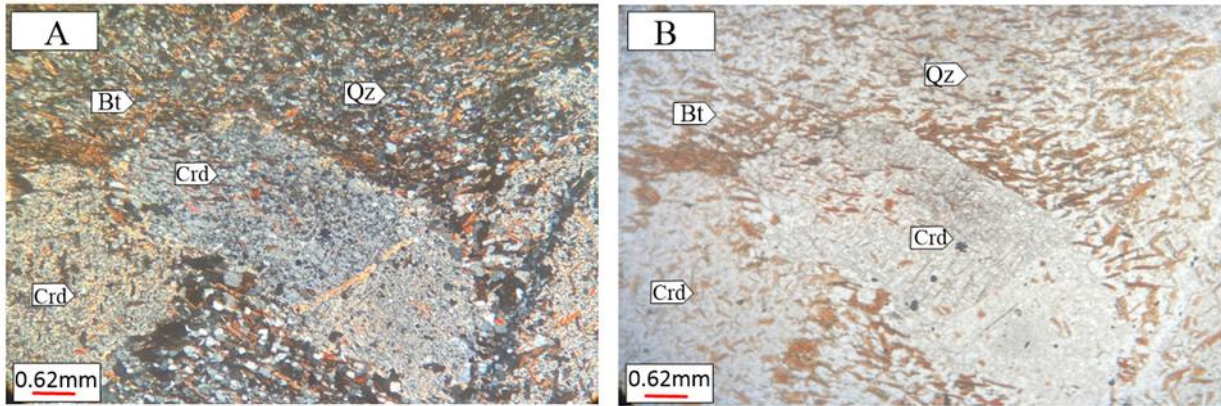


Figure 8: Cordierite biotite hornfels: Numerous biotite, quartz, and black mineral inclusions are found in well-preserved cordierite porphyroblasts that are trapped during the porphyroblasts' growth.

B: The identical picture in plane-polarized light (PPL).

Andalusite -Cordierite- Biotite -Hornfels

As one approaches the contact at distances between 300 and 350 meters, assemblages of And+Qtz+Bt+Ms+Crd are seen at higher grades in the hornfels. These rocks have a much lower concentration of muscovite and a matrix of quartz and biotite, with andalusite crystals resembling giant poikiloblasts. On the slates' cleavage planes, this andalusite is clearly visible. The andalusites, quartz, and biotite, which exhibit partial growth zoning (Fig.9), are located adjacent to the cordierites in the thin section. There is butterfly twinning in the cordierite. Compared to the cordierites, the andalusites crystals contain modest overgrowths. The initial foliation has been nearly retained by the biotite, which has crystallized in that orientation. The modal amount of muscovite is 5-8%, but biotite is more abundant than muscovite. These rocks have seen a higher grade of metamorphism, as evidenced by the presence of andalusite and recently formed chlorite. Andalusite sometimes exhibits strong undulatory extinction. Although undulatory extinction in andalusites is an indication of the presence of subgrains. However subgrains are not due to deformation because deformation related strain is not observed in the mineral sizes. Andalusite is more abundant as one gets closer to the intrusive. The mechanism of andalusite mineral development is most likely the cause of the undulatory extinction in andalusites. Biotite and cordierite assemblages are also seen in all samples that include andalusite. The area's breadth

ranges from 200 to 300 meters, and the slate outcrop with these assemblage characteristics is located in the northwest portion of the intrusive (Fig.10).

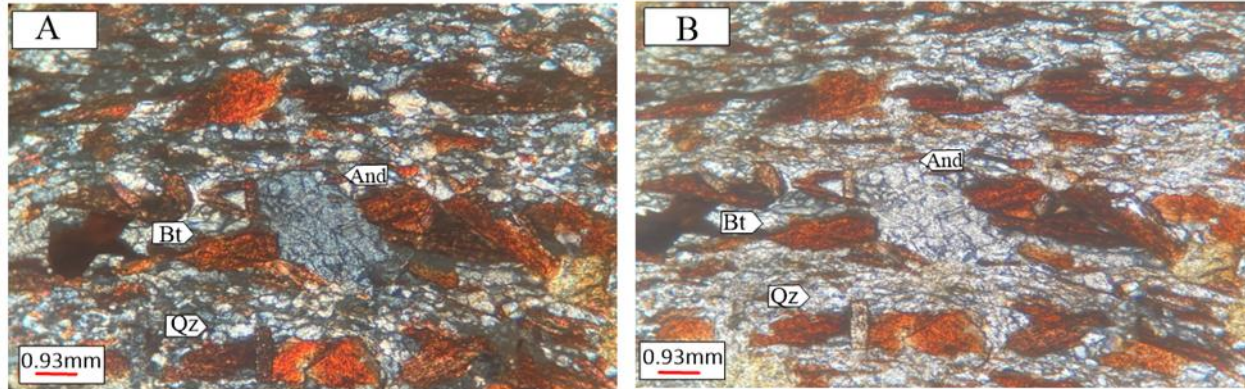


Figure 9: Andalusite -Cordierite- Biotite -Hornfels: Cordierites and Andalusites get into contact. Growth zoning is seen in biotite and quartz.

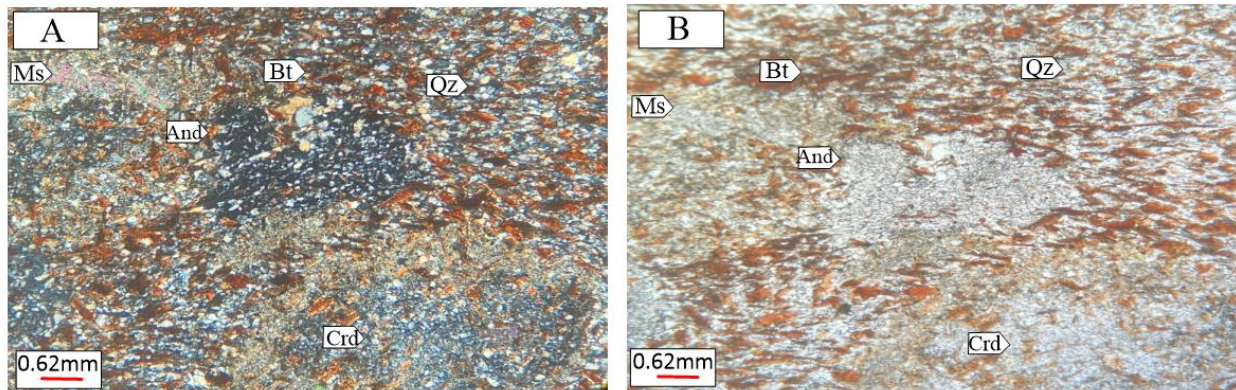


Figure 10: Andalusite -Cordierite- Biotite -Hornfels: Along with quartz and biotite, which show some degree of growth zoning, andalusites are found in thin portions close to cordierites. In contrast to cordierites, andalusite crystals have fewer inclusions, and cordierites exhibit butterfly twinning.

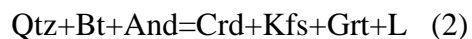
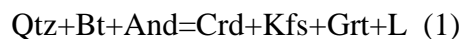
Cordierite- Garnet- Biotite – muscovite -Hornfels

The primary cordierites are well-formed, 2.5 m in size, and have a poikiloblastic texture in thin sections. Along the cleavages, they are extended. Numerous muscovite and opaque mineral inclusions can be seen in cordierite. Occasionally, inclusions of pre-existing regional biotites are also seen in these cordierites. Inclusion of muscovite and biotite usually retain the same

orientation of the slaty cleavage outside the cordierite. This shows that the expanding cordierite crystals have been contained by the oriented micas. Outside of the cordierite, the micas are typically coarser and more randomly oriented, suggesting that they formed or crystallized during contact (thermal) metamorphism. More cordierite patches in the thin section had entirely changed into fine-grained aggregates of pinitic or gray-brown sericite that show reverse pleochroism. The first sign of the rocks' migmatization has been thought to be Cordierite-Garnet-Biotite – muscovite -Hornfels with leucosome veins and cracks. In regions with high quartz abundance, it appears rounded and subhedral, whereas Quartz is found in a xenomorphic and interlayered form in regions with low quartz concentrations. (fig.10).

They are often covered by cordierites in the area between the leucocratic and melanocratic parts. Apart from cordierites, biotite and K-feldspar are also found, either as inclusions in the cordierite or as independent minerals. Aggregates of sericitized cordierite are seen in a thin section close to the leucosomes (or possibly the leucosomes themselves). The quartz that forms between these aggregates of cordierite has a xenomorphic and intergranular appearance. At the edges of the cordierite aggregates, biotite is frequently seen (fig.11).

as a result, regions of the molten zone's mineralogical composition at the beginning of the melting process contain K-feldspar, quartz, and cordierite. K-feldspar appears as a crystal, but cordierite crystals are comparatively coarse and have inclusions. The quartz is intergranularly arranged. The mineralogy of the area includes garnet, K-feldspar, and cordierite, as shown by the metamorphic mineral assemblage discovered in the pelitic rocks of the Khar Formation. Considering the mineral assemblage of the leucosomes, the potential melting reaction might be expressed as follows:



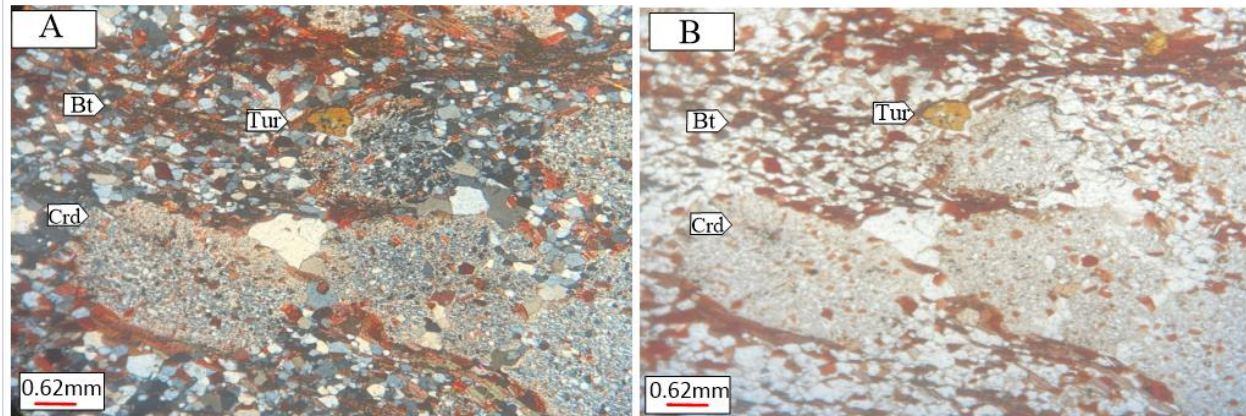


Figure 10: Cordierite- Garnet- Biotite – muscovite -Hornfels :the earliest indications of migmatization with visible veins and leucosome cracks.

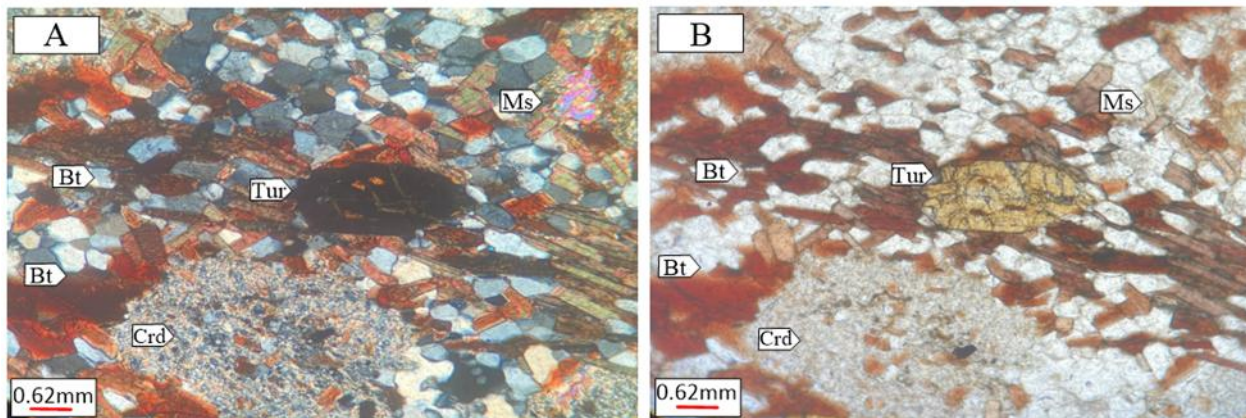


Figure 11: Cordierite- Garnet- Biotite – muscovite -Hornfels:The sericitized cordierite aggregates are seen. These aggregates of cordierite contain intergranular, isomorphic quartz. At the margins of the cordierite aggregates, biotites are frequently observed.

METHODS

Focused sampling of Neoproterozoic (Percambrian) slate and hornfels rock at specific distances from I-type granites intrusion and S-type granite intrusions in the Misho region was part of the research methodology. After a thorough petrographic analysis, thin sections were extracted from components that had visible prophyroblasts in hand samples. Lastly, the University of Tabriz's laboratory and research center used the Raman spectroscopy technique with 25 devices. With a wavelength of 523 nm, laser power between 0.5 and 70, a focus of 1 micro, and pixel sizes smaller than 1.5, the analysis was performed.

Raman studies of indialite

There are two main kinds of cordierite crystallization, cordierite and indialite. natural cordierite is typically a low-temperature mineral and an orthorhombic structure, With a three-dimensional space group (Cccm) (Gibbs 1966, Meager and Gibbs 1997). Indialite, a high-temperature cordierite with a disordered and hexagonal structure, is stable up to 1450 C° (Meager and Gibbs 1997; Takane 1942; Miyashiro and Iiyama 1954; Iiyama 1955; Harwood and Larson 1969; Deer et al. 2013; Finkelstein et al. 2015). The cordierite minerals have different peak points in their Raman spectra and are located at different distances from the intrusion. Cordierite that is 500 meters from the intrusion can have its Raman spectrometry separated into many portions, each with distinct spectrometric properties. The main peaks have been found at 260 cm⁻¹, 670 cm⁻¹, 971 cm⁻¹, and 1010 cm⁻¹. Additionally, bands have been observed between 522 and 645 cm⁻¹. According to Haefeker et al. (2013), the Mishow cordierite is Fe-Mg cordierite, as confirmed by a comparison of the Raman spectra with phases rich in magnesium and the data from RRUF. According to Haefeker et al. (2012), the tetrahedral stretching vibrations of the SiO₂ site connected to the M site are responsible for the band at 990 cm⁻¹, the strong band at 1031 cm⁻¹ is ascribed to stretching, and the band at 990 cm⁻¹ is attributed to the tetrahedral stretching vibrations of T₂₁, T₂₃ from the SiO₂ site connected to the M site. The stretching of T₁₁ Al-tetrahedra associated with the M site is responsible for the vibrational band at the lower wavenumber of 645 cm⁻¹, whereas the stretching vibration related to the orientation of the M site produces the peak at wavenumber 260 cm⁻¹. The main difference between the measured peaks and those of mg cordierite is that all of the peaks in iron-bearing cordierites have moved to lower wavelengths Haefeker (2012). pure

mg-cordierites, as shown by the bending vibrations of the M site from the peak at 260 cm^{-1} . The bending of $T_2 1$, $T_2 3$, and $T_2 6$ sites is associated with the band at 456 cm^{-1} . The stretching vibrations of M and $T_2 6$ are represented by the bands at 595 cm^{-1} and 530 cm^{-1} . This kind of band division is consistent with earlier research on synthetic cordierites. The low-temperature orthorhombic structure replaces the hexagonal structure when the symmetry varies with temperature. Consequently, peaks at 553 cm^{-1} and 595 cm^{-1} have separated from the strong band at 566 cm^{-1} . The splitting of the 566 cm^{-1} band clearly shows the change from hexagonal to orthorhombic transformation, which is based on investigations on synthetic indialite and its transformation into cordierites Haefeker et al. (2013)(Fig. 12)

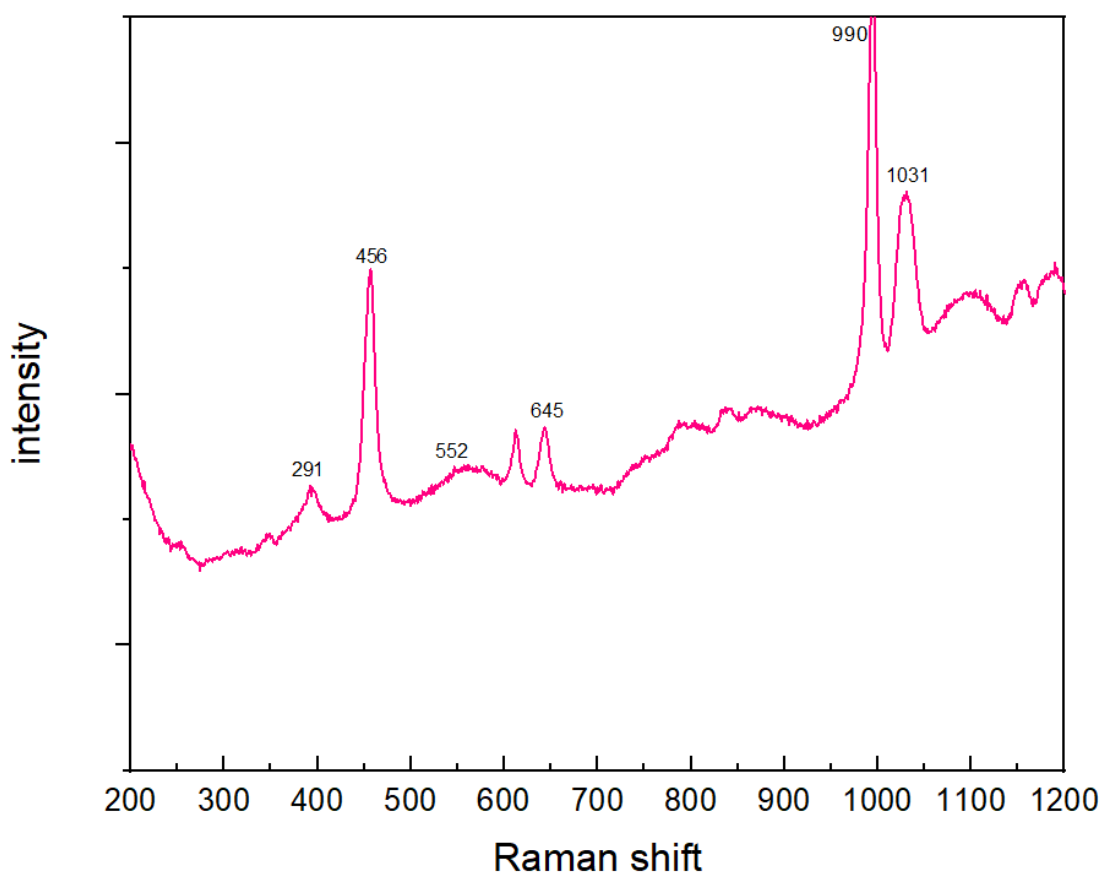


Figure 12: Raman spectra of orthorhombic cordierite in the 1100–1200 cm^{-1} band are shown.

The Raman spectrum characteristics of Indialite, a rare polymorph of cordierite, have been examined in migmatites and hornfelses. M and T sites are typically linked to bands below 380 cm^{-1} to 300 cm^{-1} . Two medium-intensity bands (283 cm^{-1} and 105 cm^{-1}) and two strong bands below 300 cm^{-1} have been seen; the bending vibration of the octahedral M, T₁₆, T₂₆ sites, which results from the bedding mode of tetrahedral M sites, is likely responsible for the strong bands at 283 cm^{-1} and 105 cm^{-1} . Bending and rotation at the T and M sites result in a strong band at 105 cm^{-1} Kanidile et al.(2011). The octahedral and tetrahedral T₂ sites' bending vibrations are linked to the peak at 342 cm^{-1} . Several medium- to low-intensity peaks in the 444.908 cm^{-1} region are visible in the Raman spectrometry of Fe-Mg-containing indialite samples. These peaks are linked to a bending group in the crystallographic sites of the cordierites structure. In general, research on the bands at 555 cm^{-1} and 573 cm^{-1} is related to very complex bending and stretching modes and the unclassifiable movements of the octahedral and tetrahedral T₂ 1, T₂ 3, T₂ 6 sites. The band at 543 cm^{-1} is likely associated with the stretching of the tetrahedral T₂ 6 and M sites and the bending of the tetrahedral T₁₆ site, along with the presence of Si in T₁₆ and the presence of Al in T. The spectrum has a band at 543 cm^{-1} that can be utilized to identify cordierites because it is not present in cordierite (fig13).

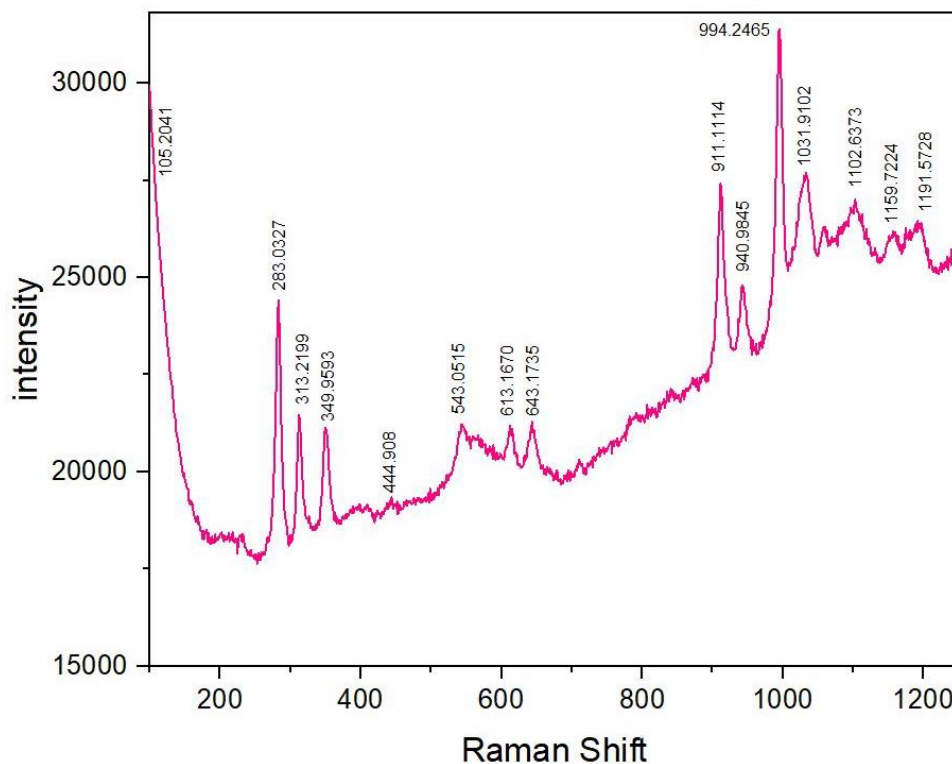


Figure 13: Figure 13: Raman spectrum of indialite from the hornfels aureole in the range of 1100 cm^{-1} to 1200 cm^{-1} .

In the range of 100 cm^{-1} to 1200 cm^{-1} , the analysis and comparison of Mg-bearing Indialite from Germany with Indialite from the Misho metamorphic aureole reveal that Misho Fe and Mg indialite display stronger bands at 283 cm^{-1} and 994 cm^{-1} than German Fe and Mg indialite (fig. 14).

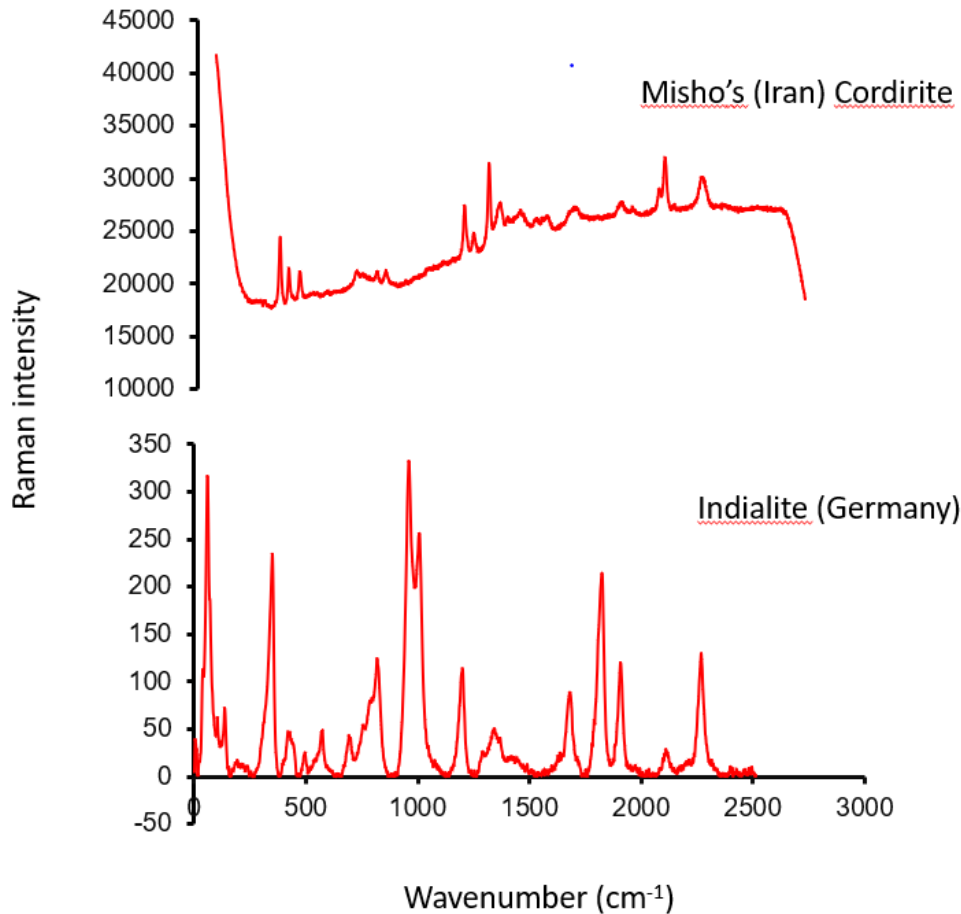


Figure 14: Investigation and comparison of German magnesium-bearing indialite and Misho's indialite in the 1100–1200 cm⁻¹ range.

Conclusion

considering that the strong band at 566 cm⁻¹ is divided into peaks at 553 cm⁻¹ and 595 cm⁻¹ and according to research on synthetic indialites (Haefeker et al. 2013) and how they turn into cordierite, at the distance from the intrusive cordierites between 500 and 350 m, The breaking of the 566 cm⁻¹ band makes the transition from a hexagonal to an orthorhombic transformation more visible. Additionally, the strong band at 1031 cm⁻¹ and 990 cm⁻¹ are attributed to the stretching vibrations of the tetrahedral T₂ 1 + T₂ 3 of SiO₂, which are connected to M site, which is characteristic of orthorhombic cordierite. Therefore, it can be concluded that these cordierites,

which are located at greater distance from the intrusive mass, form at lower temperatures, and their Raman spectra indicate their orthorhombic characteristics. In proximity, at a distance of 50 m from the intrusive, butterfly twins are observed in the studied cordierites, which are 2.5 mm in size, well-formed, and exhibit a poikiloblastic texture. The cordierite contain abundant inclusions of muscovite, quartz, and dark minerals. These rocks have undergone migmatization and are non-foliated. The micas outside the cordierite are usually more well-formed and exhibit random orientation. This feature indicates their formation and recrystallization during contact (thermal) metamorphism. Base on petrographic studies and evidence, the crystallization of hexagonal indialites likely occurred due to the melting of cordierites in the temperature range of 700 to 900 C°. This melting caused a distortion in the structure of the cordierites, leading to the formation of indialite (Miyashiro,1957) . cordierite whose Raman spectra show orthorhombic characteristics are mainly located at distances of 1 km to 500 m from the intrusive massive and are stable at temperatures lower than the melting range.

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