



ISSN: 0975-833X

RESEARCH ARTICLE

MICROTHERMOMETRIC STUDY OF APATITE OCCURRING IN THE PROTEROZOIC ULTRAMAFIC ROCKS OF ARAVALLI SUPERGROUP, JHAROL AREA, UDAIPUR, RAJASTHAN, INDIA

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ARTICLE INFO

Article History:

Received 28th June, 2015

Received in revised form

23rd July, 2015

Accepted 10th August, 2015

Published online 30th September, 2015

Key words:

Ultramafic rocks,
Talc deposits, Apatite,
Fluid inclusions,
Jharol, Rajasthan.

ABSTRACT

Coarsely crystallized, semi precious green and yellow varieties of apatites are occurring in talcosic ultramafic rocks of the Jharol area. XRD data shows that the green variety is hydroxyl-chlorapatite while, the yellow variety is a fluor apatite. Fluid inclusion studies carried out on these apatite samples reveal that majority of primary inclusions in hydroxyl-chlorapatite are H₂O-CO₂ type while, the fluorapatite contains biphasic aqueous inclusions without visible CO₂ suggesting that the metamorphic fluids were C-O-H type in southern part and aqueous type with low salinity in northern part of the area. Microthermometric data coupled with stable mineral phases (or metamorphic index minerals) confirm that steatitisation of ultramafic rocks took place at about 400°-490 °C temperature and 2 to 3 kb pressure with metamorphic gradient increasing from north to south in the area.

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Citation: M. S. Shekhawat, M. S. Ranawat and P. S. Ranawat, 2015. "Microthermometric study of apatite occurring in the proterozoic ultramafic rocks of Aravalli Supergroup, Jharol Area, Udaipur, Rajasthan, India", *International Journal of Current Research*, 7, (8), 20845-20849.

INTRODUCTION

Apatite, an orthophosphate of calcium [Ca₅(PO₄, CO₃)₃(F, OH, Cl)] in which F, OH, and Cl ions can substitute each other and forms a complete isomorphous series ranging from fluorapatite to chlorapatite (Deer *et al.*, 1962), is present in the area. It is a common accessory mineral of most of the igneous rocks *i.e.* granites, syenites through gabbro. Apatite also occurs as green, blue, brown, orange and yellow coloured hexagonal crystals disseminated in minor amounts in gneisses, pegmatites and arenaceous sediments. Occasionally, it forms large deposits as veins in association with alkalic rocks. Samples of green and yellow apatite crystals were collected from talc deposits located near Jharol about 55km south-west of Udaipur (Fig. 1). The talc deposits are hosted by ultramafic rocks which occur as intrusive within the metasediments of Jharol Group of the Aravalli Supergroup (2500- 2000 m.y., Gupta *et al.*, 1997). Green apatite samples were collected from two working pits of talc located near the villages Khakhar and Gopir in southern part and yellow apatite samples were collected from one working quarry of talc located near the village Undithal in northern part of the area (Fig. 1).

The crystals of deep green (emerald-like) and yellow apatite (topaz-like) (Fig. 2A) occurring co-genetically with talc (Fig. 2B) have primary H₂O-CO₂ and biphasic aqueous inclusions which have been used for estimating the P-T-X parameters of metamorphism. A preliminary study on fluid inclusions in apatite reported from the area was carried out by Ranawat and Shekhawat (1986, see also Ranawat, 2007; 2009).

Geological setting

A long narrow belt of ultramafic rocks (Jharol East ultramafic belt) (Shekhawat, 1987) extending for about 35 km in length and 4 km in width is situated near Jharol about 55 km south-west of Udaipur. The ultramafic rocks are best exposed in northern and southern parts of the belt as completely serpentized and extensively steatitized bodies represented as massive serpentinite, talcose-serpentinite and talc-serpentine magnesite rock (Ranawat, 1995; Shekhawat *et al.*, 2010). These rocks occur as small lenticular to sheet-like intrusive bodies within the metasediments of Aravalli Supergroup (2500-2000 m.y.) of Palaeoproterozoic age (Gupta *et al.*, 1997). The talcose-serpentinite hosts workable deposits of talc (Ranawat, 1995; Shekhawat *et al.*, 2010) while, the green coloured massive serpentinites are extensively utilized as decorative and dimensional stones (Shekhawat, 2000; DMG, 2009).

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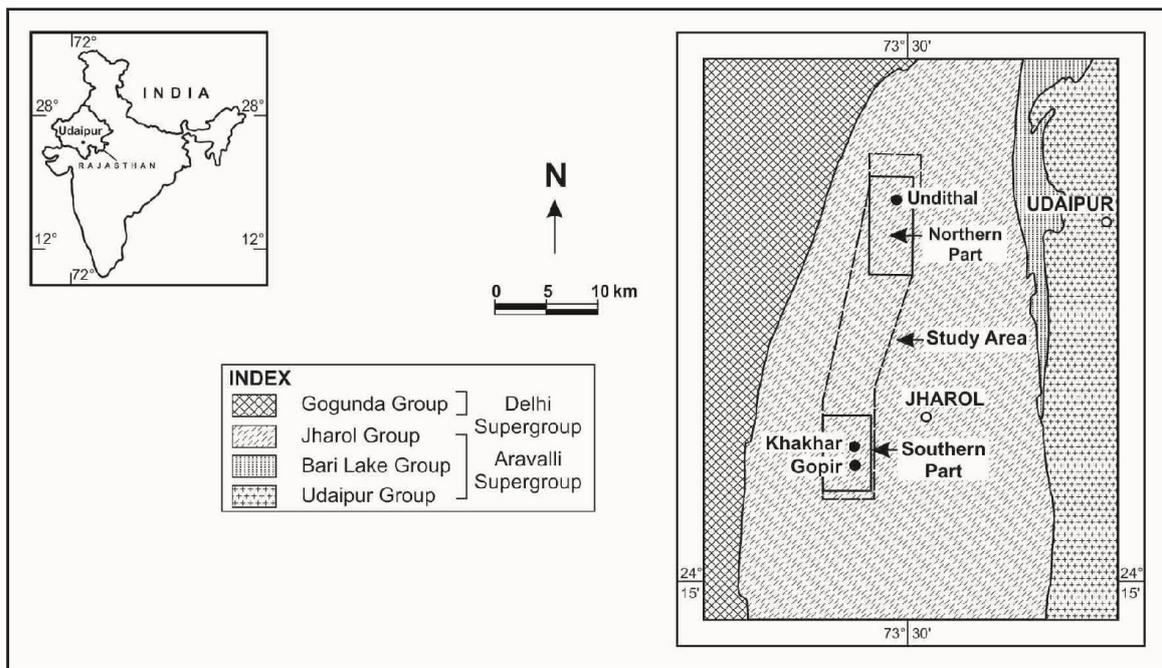


Fig. 1. Geological map showing location of the study area (based on Gupta *et al.*, 1997).

The principle metasedimentary rocks exposed in the area include quartz-mica schist and quartzite. These rocks are considered to be deposited under deep sea environmental conditions as the youngest member of the Aravalli Supergroup (Sinha-Roy *et al.*, 1998).

The talc deposits occur as pinch and swell bodies, and as bands of variable width at the contact of talcose serpentinite and the country rock quartz-mica schist. These deposits are characterized by presence of lithological zones which include outer phlogopite zone, middle chlorite zone and inner talc zone. These zones are considered to be formed as a result of metamorphic differentiation added with metasomatism (Shekhawat *et al.*, 2010). Apatite occurs as euhedral to subhedral hexagonal crystals disseminated (Fig. 2A) within the various zones of talc bodies, up to 5 cm width.

XRD STUDY

Two samples, one each of green and yellow apatite were prepared for powder XRD analyses (Cullity, 1978) using a Phillips PD 1730 diffractometer (ADP-10) with Ni-filtered $\text{CuK}\alpha$ radiation at 40 kV and 30 nA at the CRDL of Hindustan Zinc Limited, Debari, Udaipur. XRD data obtained for these samples are given in Table 1. The d-spacing values obtained for the samples were compared with the standard values given in JCPDS cards. XRD data presented in Table 1 show that the green-apatite (emerald-like, Fig. 2A), collected from southern part of the area is hydroxyl-chlorapatite whereas yellow-apatite (topaz-like, Fig. 2A) from northern part of Jharol area is fluorapatite (Table 1).

Table 1. XRD data of green and yellow- apatite from Jharol East ultramafic belt, Udaipur, Rajasthan

| S. No. | Sample No. (apatite) | No. | MSA-1 | (Green-apatite) | Sample No. (Yellow-apatite) | No. | MSA-2 | (Yellow-apatite) | |
|--------|----------------------|--------|-------|-----------------|-----------------------------|--------|-------|------------------|----|
| | | d (Å°) | I/I | I | MI | d (Å°) | I/I | I | MI |
| 1. | 3.5141 | 11 | 36 | HA | 4.6624 | 12 | 47 | FA | |
| 2. | 3.4486 | 75 | 237 | CA, HA | 3.4468 | 47 | 190 | FA | |
| 3. | 3.1675 | 15 | 47 | HA | 3.1161 | 35 | 141 | FA | |
| 4. | 2.8100 | 94 | 299 | HA | 2.8096 | 95 | 382 | FA | |
| 5. | 2.7757 | 100 | 318 | CA, HA | 2.7787 | 47 | 186 | FA | |
| 6. | 2.7120 | 52 | 164 | HA | 2.7143 | 40 | 161 | FA | |
| 7. | 2.1407 | 27 | 86 | HA | 2.6266 | 16 | 66 | FA | |
| 8. | 1.9968 | 42 | 134 | HA | 2.2740 | 4 | 18 | FA | |
| 9. | 1.9407 | 81 | 258 | CA, HA | 2.2593 | 27 | 110 | FA | |
| 10. | 1.8389 | 37 | 117 | CA, HA | 2.1497 | 3 | 14 | FA | |
| 11. | 1.8025 | 18 | 59 | CA, HA | 1.8900 | 95 | 380 | FA | |
| 12. | 1.7877 | 10 | 33 | CA, HA | 1.7777 | 6 | 25 | FA | |
| 13. | 1.7209 | 39 | 123 | HA | 1.7521 | 38 | 150 | FA | |
| 14. | 1.7128 | 5 | 17 | - | 1.5397 | 2 | 9 | FA | |
| 15. | 1.6894 | 4 | 12 | CA, HA | | | | | |
| 16. | 1.6092 | 5 | 16 | CA, HA | | | | | |
| 17. | 1.5025 | 63 | 199 | HA | | | | | |

MI – Mineral identified, HA – Hydroxyl-apatite, CA – Chlor-apatite, FA – Fluor-apatite

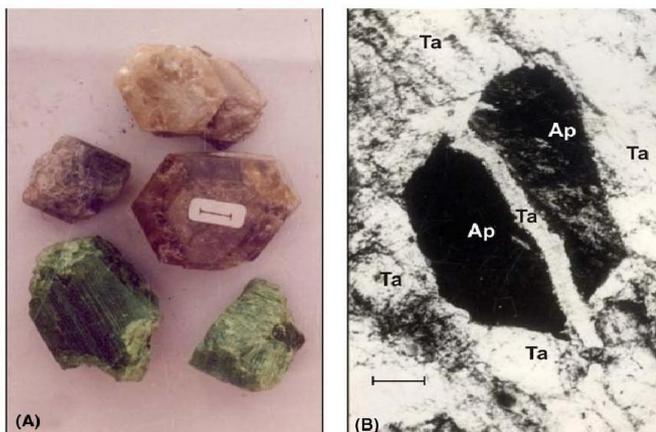


Fig. 2. (A) Hand specimen photograph showing hexagonal crystals of yellow and green-apatite occurring in talc lenses from Undithal and Gopir area respectively. Bar = 1 cm. (B) Photomicrograph of euhedral apatite (Ap) within talc (Ta) and a vein of talc (Ta) traverses the apatite (Ap) crystal. Loc. Undithal, Bar = 0.28 mm.

Fluid inclusion study

Samples of well developed crystals of both green and yellow varieties of apatite were selected for fluid inclusion studies. About 0.5 mm thick slices were cut from the crystals and both sides of the wafers were polished as per the standard procedures (Roedder, 1984). The polished wafers were studied for fluid inclusions under varying magnifications using transmitted light H.M. Pol microscope. Microthermometric experiments were carried out using Chaixmeca microthermometry apparatus, at the Fluid Inclusion Laboratory, Department of Geology, M. L. Sukhadia University, Udaipur, Rajasthan.

Phase-types and Distribution Patterns

Most of the inclusions in green-apatite from talc and chlorite zones have regular hexagonal outlines, while irregular inclusions have been observed in samples from phlogopite zone. In this variety of apatite, frequency of inclusion is high and most of them are H₂O-CO₂ type primary inclusions with perfect hexagonal outline (Fig. 3A) and have thick and distinct boundary.

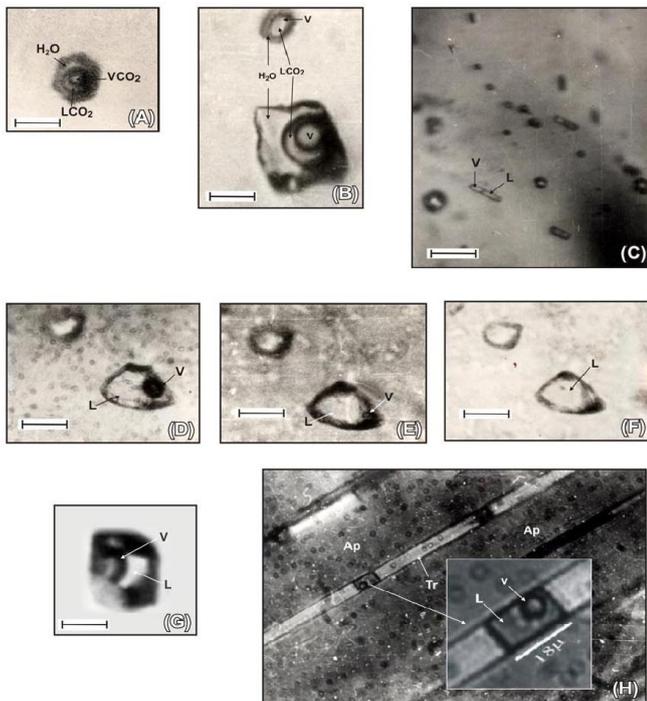


Fig. 3. Photomicrographs showing: (A) H₂O-CO₂ type primary fluid inclusion with hexagonal cavity outline in green-apatite from Khakhar area. Photographed at 0 °C having conspicuous ring of liquid CO₂ (L CO₂) around gas CO₂ (V CO₂). Bar = 7 μ. (B) H₂O-CO₂ type primary fluid inclusions with irregular cavity outline in green-apatite from Gopir area. Bar = 7 μ. (C) Primary fluid inclusion with tubular cavity in green-apatite from Gopir area. Photographed at room temperature. V = homogenised CO₂ fluid, L = Aqueous phase. Bar = 7 μ. (D, E, F) Serial photomicrographs of a primary aqueous fluid inclusion in yellow-apatite from Undithal area showing reduction in vapour bubble due to heating, (D) at room temperature, (E) at 180 °C and (F) at 212 °C homogenisation temperature (T_h). L = Aqueous phase, V = Vapour bubble. Bar = 12 μ. (G) Primary aqueous fluid inclusion having a perfect negative faceted 3-D hexagonal cavity in yellow-apatite from Undithal area. L = Aqueous phase, V = Vapour bubble. Bar = 8 μ. (H) A fairly large primary aqueous fluid inclusion confined to tremolite acicle, the latter occurs as inclusion in yellow-apatite from Undithal area. Tr = Tremolite, Ap = Apatite, L = Aqueous phase, V = Vapour bubble.

However, irregular, tabular, elliptical to sub-rectangular (Fig. 3B) and tubular (Fig. 3C) inclusions of different sizes have also been observed. The flatter inclusions with hexagonal outline have disc shaped gas bubble that makes phase ratio determination easy.

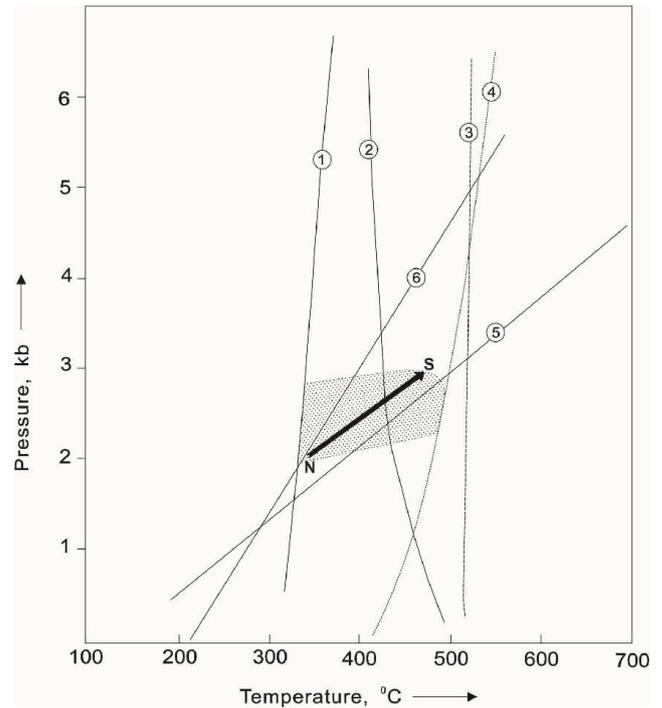


Fig. 4. Pressure-temperature diagram showing the P-T field of regional metamorphism of Jharol East ultramafic rocks based on mineral stability fields and fluid inclusion data. The shaded area represents the P-T regime and the arrow indicates rise in P-T parameters from north to south in the belt.

1. $S + 2Q = Ta + H_2O$ (Winkler, 1976)
 2. $Fo + H_2O + CO_2 = Ta + Ma$ (Johannes, 1969)
 3. $Al-Serp. = Clc$ (Yoder, 1952)
 4. $S + Dio = Tr + Fo + H_2O$ (Evans and Trommsdorff, 1970)
 5. 10% NaCl, H₂O + CO₂ isochore (Nicholls and Crawford, 1985)
 6. H₂O isochore of 0.85 gm/cm³ density (Crawford, 1981)
- S = Serpentine; Q = Quartz; Ta = Talc; Fo = Forsterite; Ma = Magnesite; Al-Serp = Aluminian Serpentine; Clc = Clinocllore; Dio = Diopside; Tr = Tremolite.

The size of H₂O-CO₂ type primary inclusions vary from 10 μ x 6.5 μ to 16 μ x 15 μ with gas bubble diameter ranging from 4 μ to 6 μ. The volume percentage of CO₂ in these inclusions ranges from 10% to 30%. Biphase and monophasic secondary inclusions of diverse shapes and sizes occur in trails and rows along the healed fractures. Yellow-apatite occurring in talcose bodies near the village Undithal in the northern part of the area contains biphase aqueous primary inclusions. They are mostly irregular to sub-regular in shape (Fig. 3D-F). However, perfect hexagonal inclusions have also been observed (Fig. 3G). A notable feature observed is the occurrence of biphase aqueous primary fluid inclusions localised along cross-fractures in needles of tremolite that occurs within yellow-apatite (Fig. 3H). The fluid inclusions are elongated parallel to the length of the tremolite needles and their width is nearly equal to the width of tremolite needle. The textural study revealed that tremolite needles were trapped in yellow-apatite during its recrystallisation growth. The space available along fracture across the needles provided favourable loci for entrapment of

metamorphic fluids to give rise to the fluid inclusions. Thus, these inclusions belong to the category of primary inclusions.

Microthermometry

For microthermometry, only large size, primary inclusions with better clarity were selected following the criteria given by Roedder (1984) and Shepherd *et al.*, (1985). Microthermometric experiments were carried out for H₂O-CO₂ primary hexagonal inclusions present in green-apatite occurring in all the three zones of talcose bodies of the southern part and primary biphasic aqueous inclusions in yellow-apatite from talcose bodies of the northern part of the Jharol area. Taking into consideration the possibility of leakage of fluids during the heating runs, freezing studies were made prior to heating experiments. Microthermometric data (Table 2) reveal that the temperature of CO₂ homogenisation (T_h CO₂) of H₂O-CO₂ inclusions varies greatly from +1.5 °C to +24.8 °C except in one inclusion which shows lower values (-0.6 °C) and the CO₂ melting temperature (T_m CO₂) ranges from -56.7 °C to -65.5 °C. The temperature of total homogenisation (T_h Total) varies from +158 °C to +306 °C.

The temperature of CO₂ homogenisation (T_h CO₂) of H₂O-CO₂ inclusions in apatite from talc zone ranges between +15 °C and +24.8 °C. This temperature recorded for inclusions in apatite from chlorite zone (+1.5 °C to +7 °C) and phlogopite zone (+4.8 °C to +9.2 °C) is much lower than those of talc zone. The temperature of CO₂ melting (T_m CO₂) in H₂O-CO₂ inclusions in apatite from phlogopite zone range from -64.9 °C to -65.5 °C. The melting temperature of CO₂ is found to be much lower than the T_m of pure CO₂ at -56.6 °C. Similarly, the CO₂ melting temperatures of H₂O-CO₂ inclusions in apatite from chlorite and talc zones are found to be in the range of -58.7 °C to -61.9 °C and -56.7 °C to -59.5 °C respectively. The temperature of total homogenisation (T_h Total) of H₂O-CO₂ inclusions in apatite from phlogopite zone varies from +186 °C to +252 °C. In apatite from chlorite zone this temperature ranges from +212 °C to +264 °C. One inclusion did not homogenise up to +242 °C and decrepitated before homogenisation. Most of the H₂O-CO₂ inclusions in apatite occurring in talc zone homogenise between +158 °C and +298 °C.

The majority of inclusions homogenise between +188 °C and +192 °C. The critical homogenisation at +290 °C was observed in H₂O-CO₂ inclusions in apatite from talc zone but in one inclusion, a faint boundary is visible up to +298 °C. The aqueous fluid inclusions in yellow-apatite homogenise to liquid phase in a fairly narrow temperature range of +187 °C to +208 °C. The secondary inclusions decrepitated at +150 °C.

Data Synthesis

Studies on fluid inclusions in apatites co-genetic with talc, chlorite and phlogopite have provided basis for the evaluation of the chemical environment of steatitisation of ultramafic rocks of the study area. Perusal of data presented in Table 2 reveals that the density of CO₂ in the H₂O-CO₂ type inclusions (Fig. 3A-C) increases from talc (0.775 gm/cm³) through chlorite (0.892 gm/cm³) to phlogopite (0.9 gm/cm³) zones. CH₄ content in CO₂ increases considerably in the phlogopite zone, which is indicated by fall of T_m CO₂ from nearly -57 °C to -64 °C. The density of carbonic fluids will be slightly less due to the presence of CH₄. However, progressive increase in the density of metamorphic fluids is indicated from the inner talc zone to the outer phlogopite zone. Because of presence of CO₂ and CH₄ in the fluids, the inclusions have high internal pressure which increases considerably during the heating runs. Therefore, in many of the cases the fluid inclusions burst open before complete homogenisation is attained. The apatite in the northern part of the area contains aqueous fluid inclusions (Fig. 3D-H) that have low salinity (~4 eq. wt% NaCl). Temperature of homogenisation is however comparable to those of C-O-H fluid inclusions. Presence of metamorphic minerals like garnet, talc, chlorite, tremolite, serpentine indicates that the pressure at the time of regional metamorphism was nearly 2 to 3 kb. This pressure will require "pressure correction" of over 200 °C on the recorded temperature of homogenisation of fluid inclusions (Roedder, 1984). Microthermometric data shows that the metamorphic fluids in the area were C-O-H type in the southern part whereas in the northern part they were mainly aqueous fluids of low salinity having temperatures of over 350 °C and density of about 0.85 gm/cm³ (*cf.* Nicholls and Crawford, 1985).

Table 2. Microthermometric data of fluid inclusions in apatite occurring in Jharol East ultramafic rocks, Udaipur, Rajasthan

| Sample No. | Phases | T _h CO ₂ Liquid °C | T _m CO ₂ °C | δ CO ₂ (gm/cm ³) | Average (gm/cm ³) | Density CO ₂ | T _h Total °C | T _m Ice °C | Salinity |
|------------|---------|--|-----------------------------------|--|-------------------------------|-------------------------|-------------------------|-----------------------|----------|
| GAT-1 | C-O-H | +19 +20.5 to 23.9 +22 to 24.8 | -59.5 | 19 = 0.776 22.2 = 0.767 23.4 = 0.787 | 0.776 | | +172 to 280 | | |
| GAT-2 | C-O-H | +15 to 20 +21 +16 to 21.5 | -56.9 | 17.5 = 0.846 21 = 0.755 18.7 = 0.856 | 0.819 | | +163 to 274 | | |
| GAT-3 | C-O-H | +16 to 18 +23 | -56.7 | 17 = 0.796 23 = 0.731 | 0.763 | | +158 to 285 | | |
| GAT-4 | C-O-H | +24.2 | -57.9 | 24.2 = 0.745 | 0.745 | | +194 to 306 | | |
| GAC-1 | C-O-H | +3 to 7 | -58.7 | 5 = 0.888 | | | +212 to 244 | | |
| GAC-2 | C-O-H | +6 | | 6 = 0.882 | | | +235 to 250 | | |
| GAC-3 | C-O-H | +1.5 to 2.6 | -59.5 | 2 = 0.906 | | | +220 to 264 | | |
| GAC-4 | C-O-H | -0.6 | -61.9 | | | | >242 | | |
| GAP-1 | C-O-H | +9.2 | | 9.2 = 0.877 | | | +197 to 245 | | |
| GAP-2 | C-O-H | +4.8 | | 4.8 = 0.942 | | | +214 to 252 | | |
| GAP-3 | C-O-H | +7 to 7.4 | -64.9 | 7.2 = 0.890 | | | +186 to 239 | | |
| GAP-4 | C-O-H | +6.8 to 7.6 | -65.5 | 7.2 = 0.890 | | | >225 | | |
| YAT-1 | Aqueous | | | | | | +193 to 208 | -2 | 3.36 |
| YAC-1 | Aqueous | | | | | | +187 to 206 | -3 | 4.91 |

GAT, GAC and GAP – Green-apatite from talc, chlorite and phlogopite zones respectively, southern part of the area; YAT and YAC – Yellow-apatite from talc and chlorite zones respectively, northern part of the study area.

DISCUSSION AND CONCLUSIONS

Petrographic study shows that small disseminated grains of apatite occur in the serpentinites of the area (Shekhawat *et al.*, 2010). These were mobilised by the metamorphic fluids and redeposited as large disseminated euhedral green and yellow apatite crystals, which entrapped part of the depositing fluids. These fluid inclusions provide constraints on the temperature and composition of the metamorphic fluids. Occurrence of large well-developed apatite crystals indicates their very slow growth from metamorphogenic hydrothermal fluids with low phosphate content. The regional metamorphic conditions for the formation of talc in the studied area have been established by using fluid inclusion data in conjunction with stable mineral phases. The fluid inclusion studies carried out on green and yellow-apatite occurring co-genetically with talc (Fig. 2B) provide evidence that the metamorphic fluids that operated in the northern part of the area were aqueous in nature with low salinity, whereas in the southern part, they were low salinity C-O-H type (Table 2). This aspect is reflected in the end product rocks of the belt. Steatitisation and simultaneous carbonatization in the southern part of the area, where carbonic fluids were operative, produced talcose-serpentine and talc-serpentine-magnesite rock. Furthermore, within the belt from north to south an increase in P-T gradient is evident as reflected by increase in presence of garnet, tremolite, magnesite and greater steatitisation of serpentine.

Petrographic studies reveal that talc and serpentine (antigorite) association is the most stable assemblage observed in Jharol East ultramafic belt (Shekhawat *et al.*, 2010). The upper stability limit of serpentine at 490 °C (Yoder, 1952) and temperature of formation of talc at above 330 °C (Johannes, 1969) indicates that the temperature of talc formation in the study area falls between 330 °C to 490 °C. As discussed in the section on data synthesis of fluid inclusion studies, the metamorphic fluids in the area were C-O-H type in the southern part, whereas in the northern part they were essentially low-salinity aqueous fluids with temperatures >350 °C and density of about 0.85 gm/cm³. The P-T estimate (Fig. 4) based on mineral assemblage (talc-serpentine-magnesite-chlorite) observed in ultramafic rocks of Jharol East belt constrained by aqueous and carbonic isochores indicates that the rocks were regionally metamorphosed under upper-green schist to lower amphibolite facies conditions in the temperature range of 400-490 °C and pressure of about 2 to 3 kb with metamorphic gradient increasing from north to south (Fig. 4).

Acknowledgements

The authors are thankful to Dr. R. N. Roy, the then Senior Manager (Geology) CRDL, HZL, Udaipur for providing XRD laboratory facilities. The Heads, Department of Geology, M. L. Sukhadia University, Udaipur are gratefully acknowledged for extending other required facilities for completing this work. The State Department of Science and Technology, Government of Rajasthan, is thankfully acknowledged for providing financial support for this work in form of an R&D project.

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