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# Raman Microspectrometry of Fluid Inclusions from Quartz Veins of Eastern MahakoshalBelt, Central India

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***Abstract:*****Gold mineralization occurs in the PaleoproterozoicMahakoshalSupracrustalBelt (MSB) of the Central India Tectonic Zone (CITZ). The MSB comprises metavolcanics and metasedimentary rocks,where gold mineralization is predominantly hosted by quartz veins traversing in these rocks. In the present study,Laser Raman Microspectrometry(LRM) technique is used for characterization of various types of fluid inclusion linked with gold mineralization in the eastern part of the MSB. The gold mineralization at Parsoi, Sonapahari and GurharPahar is associated with the compositionally indistinguishable fluids.Involvement of $CH_4$ during the transportation and deposition of the ore fluid significantly favors gold mineralization. Ore fluids with composition of $H_2O$-NaCl-$CO_2$-$CH_4$ ± $N_2$ was accountable for gold mineralization in the eastern MSB. The genetic aspects of auriferous ore fluids in the quartz veins of eastern MSB, have implications to further exploration of gold in this mineral belt.**

***Index Terms:*****Gold mineralization, Mahakoshal, Fluid inclusion, Laser Raman microspectrometry, Ore fluid.**

## I. INTRODUCTION

Fluid inclusions are microscopic pore spaces or bubbles or dropletswithin a natural crystal (minerals) and contain materials in different phases such as solid, liquid and vapor (Roedder, 1984; Alderton, 2021). These microscopic bubbles usually occur in wide variety of rocks and minerals formed during hydrothermal activities, sedimentation and metamorphism (Roeder, 2003).However, fluid inclusions <10 µm in size are commonly found inquartz, calciteand fluorite crystals (Rankin, 1989). Genetic classification of fluid inclusions in minerals should be established with the study of petrographic microtextural relationship (Van den Kerkhof and Hein, 2001). Generally, various generations of fluids occurring in minerals require the determination of compositional properties of discrete inclusions (Burke, 2001). Numerous microanalytical techniques have been developed for characterization of individual fluid inclusions in minerals (Roedder, 1990). Qualitative and semiquantitative study of fluid inclusions can be performed using Laser Raman Microspectrometry (LRM;Delhaye and Dhamelincourt, 1975) and Fourier Transform Infra-Red (FTIR) spectroscopy (Barres et al., 1987), which is a non-destructive analytical technique.

Laser Raman microspectroscopy is a versatile technique to study fluid inclusions and characterization of earth materials (Chou and Wang, 2017). LRM technique is applied for the identification of gaseous mixtures present in the fluid inclusions and estimation of fluid density (Frezzotti et al., 2012) as well as characterization of structural and compositional order - disorder study in minerals(Mernagh et al., 1993). The intensity of characteristics vibrational modes of species or molecules or phases corresponding to the peaks in the Raman spectrum are used for identification(Roedder, 1990; Burke, 2001). Gold

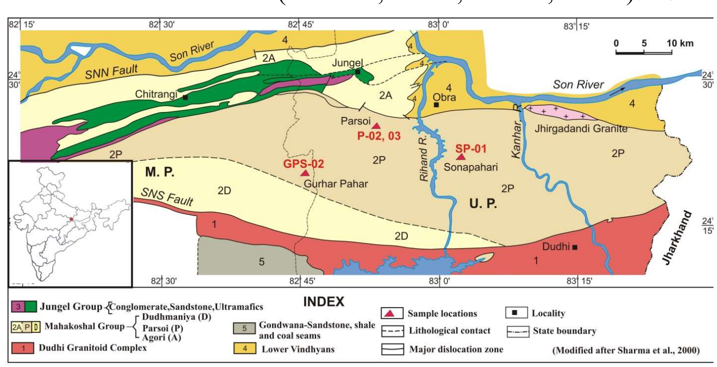


Fig. 1.Regional geological map of eastern Mahakoshal belt showing the sample locations of quartz veins.

mineralization has been reported in quartz veins from various parts of eastern Mahakoshal belt of Central India (Devarajan et

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al., 1998; Khan, 2013; Misra et al., 2021, 2022).The investigation by the Geological Survey of India has led the discovery of thirty two gold occurrences in the MahakoshalSupracrustal Belt (MSB) among which Sonkorwa, Imaliya, Chakariya of western part (Baswani et al., 2023) and GurharPahar, Sonapahari, Gulaldih and Parsoi in the eastern part of the belt, are the important prospects (Misra et al., 2022). Fluid inclusion studies of quartz veins have been carried out from GurharPahar, Gulaldih, Sonapahari and Phaphrakund areas (Prasad et al., 2000; Tiwari and Singh, 2009; Dubey and Shankar, 2017). The present study emphasizes on comparative study of fluid inclusions in quartz veins from the eastern Mahakoshal belt, using LRM technique to identify various species present in the fluid inclusionsfrom the auriferous quartz veins and resolve the issues related to theevolution of the gold bearing ore fluid.

Table 1. Details of samples of quartz veins collected from eastern Mahakoshal belt.

| *S. No.* | *Location* | *Lithology* |
|---|---|---|
| P-2 | N 24° 25' 59" ; E 82° 54' 05" Parsoi, Sonbhadra district, U.P | Quartz vein in chlorite schist |
| P-3 | N 24° 25' 59" ; E 82° 54' 05" Parsoi, Sonbhadra district, U.P | Quartz vein in phyllite |
| SP-1 | N 24° 22' 10" ; E 83° 01' 30" Sonapahari, Sonbhadra district, U.P | Quartz vein in phyllite |
| GPS-02 | N 24° 19' 49" ; E 82° 45' 32" GurharPahar, Singrauli district, M.P. | Quartz vein in phyllite |

## II. Geological Framework

The Proterozoic Central India Tectonic Zone (CITZ) is a major crustal feature represented by a collage of different lithotectonicterranes, which divides the peninsular Indian Shield (Acharyya and Roy, 2000; Naganjaneyulu and Santosh, 2010).It is developed by polyphaseProterozoic tectonothermal events necessitate several successions of volcano-sedimentary deposition (Bandyopadhyay et al., 2001), deformation, metamorphism (Roy and Prasad, 2003) and magmatism (Wani and Mondal, 2018; Khanna et al., 2020). The PaleoproterozoicMahakoshalsupracrustalbelt is one of the important supracrustal belt of CITZ, which extends towards east (Nair et al., 1995; Talusani, 2001; Roy et al., 2002; Srivastava, 2013). The eastern Mahakoshalsupracrustal belt is enslaved by two major lineaments viz., the Son-Narmada South Fault (SNSF) and Son-Narmada North Fault (SNNF) demarcate the southern and northern limit, respectively (Fig. 1).

The MahakoshalSupracrustal Belt (MSB)comprising metavolcanic and metasedimentary rocks,in study area, consists of the lower Agori, middle Parsoi and upper Dudhamaniya formations (Nair et al., 1995; Misra et al., 2022) and belong to PaleoproterozoicMahakoshal Group (Roy and Bandyopadhyay, 1990; Roy and Devarajan, 2000). The MSB is intruded by ultramafic, mafic, alkaline and carbonatite rocks mainly derived from tholeiitic magma (Srivastava, 2012; 2013). Revised lithostratigraphy of the Mahakoshal Group (Nair et al., 1995; Roy and Devarajan, 2000) divides the MSB into two sectors viz. Sleemanabad area (western MSB) and Chitrangi-GuraharPahar-Dudhamaniya area (eastern MSB). The western part of MSB is dominated by stromatoliticcarbonates, metasedimentaries (chert, quartz-arenite, greywacke, conglomerate), metavolcanics, intruded by syenite bodies and alkaline rocks (Talusani, 2001; Shukla et al., 2021).However, the eastern part of MSB mainly comprises BIF, metasedimentaries (argillites, greywackes, carbonates), metavolcanics intruded by quartz veins, dolerite dykes and granitoids (Devarajan et al., 1998; Sharma et al., 2000; Misra et al., 2022). In the western part of the MSB, the metasedimentary rocks, in general shows predominance of carbonate and other chemical precipitates over the clastics, whereas, in the eastern part of the MSBdisplays predominance of clastics sediments over carbonates (Roy and Devarajan, 2000). In the eastern MSB, theAgori Formation comprises tuffs with metabasic lenses, dolomitic marble, BIF and quartzite (Mathur and Narain, 1981; Devarajan et al., 1998). The Parsoi Formation is represented by phyllites with intercalations of quartzite bands and metabasalt (Deshmukh et al., 2017). The contact between Parsoi Formation and the Agori Formation is noticeable by a major syn-depositional fault knownas the Obra-Amsi-Jiyawan fault, which is observed in the eastern part of the MSB (Nair et al., 1995; Misra et al., 2022). The Dudhmaniya Formation comprises chloriticphyllite with thin bands of BIF (mixed oxide-sulphide-silicate facies) and garnetiferous amphibole (Mathur and Narain, 1981; Roy and Devarajan, 2000). These lithounitsof MSB are intruded by numerous quartz veins and mafic dykes. The quartz veins are cryptocrystalline, fractured, of varying thickness from a few millimeters up to 2 metres, extend along the strike for 30-35m, and are observed mainly along the pervasive foliation of the phyllite (Misra et al., 2021). Mineralized quartz veins occurred at several locations viz.Arangi, Phaphrakund, Parsoi, GurharPahar, Gulaldih and Sonaparahi (Fig. 1).

The sulphide mineralization in the form of occasional specks of arsenopyrite, pyrrhotite, pyrite, galena and lumps of scorodite ($FeAsO_4.2H_2O$) is predominantly observed in quartz veins. The thin layers and lumps of bluish-brownish grey arsenate occur along the cracks and fractures of quartz veins. The arsenopyrite and scorodite is associated with gold. Spot samples from these lumps of scoroditehave yielded values up to 20.5 ppm Au (Misra et al., 2022, Murmuet al., 2012, 2014).

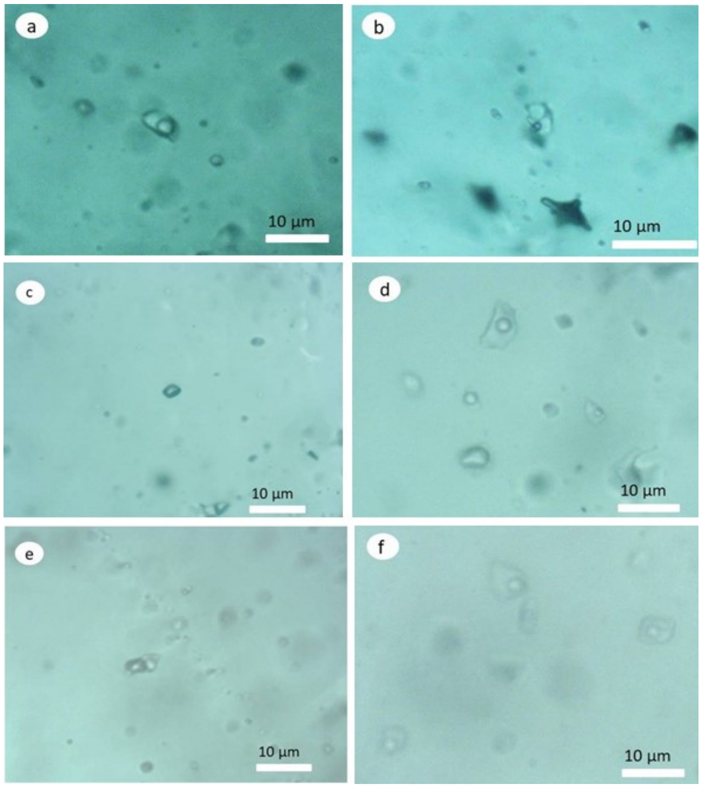


Fig. 2.Photomicrographs of polished wafers of quartz veins showing: (a) primary bi-phase inclusions ($CO_2$ + $H_2O$); (b) bi-phase inclusion showing solute crystal (NaCl+ $CO_2$) in sample no. P-2; (c) primary monophase ($CO_2$) inclusion in sample no. P-3; (d) primary bi-phase inclusion ($CO_2$+ $CH_4$ +/-$N_2$) in sample no. SP-1; (e) solid bearing secondary bi-phase inclusion ($CO_2$ + $CH_4$+ NaCl) in sample no. SP-1; (f) primary bi-phase ($CO_2$ + $CH_4$) inclusion in sample no. GPS-02.

## III. Sampling and Analytical Technique

Systematic regional traverses and detail geological field investigation were carried out in the eastern MSB during March 2022.Representative bed rock samples of mineralized quartz veins were collected from the Parsoi, Sonapahari areas of Sonbhadra district, U.P. and GurharPahar area of Singrauli district, M.P.(Table 1). The study area is located in parts of Survey of India Toposheet Nos. 63L/15 and 63P/3. Doubly polished thin wafers of 0.3mm thickness were prepared from the collected samples and studied at Geology Department, BHU and Petrology Division, Geological Survey of India, Jaipur.

Laser Raman Microspectrometry(LRM) study was conducted using Horiba JY Lab RAM HR microRaman spectrometer at the Fluid Inclusion Lab of Wadia Institute of Himalayan Geology, Dehradun (India). This instrument is fitted with an Olympus BX41 microscope, confocal arrangement, 514.4 nm argon-ion laser, 600 and 1800 lines mm grating, 1024 × 256 pixels multichannel CCD detector, motorized stage, Labspec 6.1 software, and has better than 1 $cm^{-1}$ spectral resolution (Kharya et al., 2020). The power used for the laser source was kept at ~15 mW, and the time for each run was 10-15 seconds for different fluid inclusion phases (Frezzotti et al., 2012). The uncertainties in the Raman shift were <1 $cm^{-1}$ as the instrument was calibrated and verified using silicon standard, which showed a Raman Band at 520.6 $cm^{-1}$.

## IV. Results

Fluid inclusions of different shapes and size <10 µm have been identified in double side polished wafers of quartz veins samples. The petrographic study of quartz wafers suggests that the fluid inclusions are primary as well secondary types which consists ofboth monophase and bi-phasevarieties.In this study, six types of fluid inclusions are observed. The first type (Group I) of primary bi-phase inclusions are liquid rich with gas/vapor (LV) (Fig. 2a). Second type (Group II) are primary multi-phase inclusions and contain liquid, solute crystal with vapor (Fig. 2b). Third type (Group III) are primary monophase inclusions with vapor only (Fig. 2c). Fourth type (Group IV) are primary bi-phase inclusionsand comprise carbonic fluid, mixed carbonic-aqueous fluid and aqueous fluid (Fig. 2d). Fifth type (Group V) are solid crystal bearing secondary bi-phase inclusions (Fig. 2d). Sixth type (Group VI) are primary bi-phase inclusions with mixture of vapors (Fig. 2f). The fluid inclusions have limited populations in the mineralized quartz veins with variable forms ranges between regular to irregular shapes. These inclusions occur in isolation, small clusters and randomly distributed in the quartz samples. Most of the fluid inclusions are primary in nature, however less common and aligned secondary inclusions are also observed in this study.Some of the fluid inclusions are pseudosecondary types, which are formed along healed fractures in the quartz vein samples (Fig. 2e).

The LRM spectra of the samples are characteristic and comprise narrow bands corresponding to $CO_2$, $CH_4$ and $N_2$ molecule spectra in sample GPS-02 (Fig. 3a); $CO_2$, $CH_4$ spectra in sample SP-1 (Fig. 3b); $CO_2$, $H_2O$ and $N_2$ spectra in sample P-2 (Fig. 3c); and $CO_2$, $CH_4$ spectra in sample P-3 (Fig. 3d).

Barren milky white quartz vein from Parsoi area (P-2) consists of primary inclusions, mostly biphase ($CO_2$ + $H_2O$) with very small population ofmonophase ($H_2O \pm N_2$) inclusions. However, the auriferous brownish white quartz veins from Parsoi (P-3) bears primary inclusions of monophase ($CO_2$, $CH_4$) and biphase($H_2O$ + $CO_2$) type.Creamy white quartz vein from Sonpahari (SP-1) comprises mostly biphase($CO_2$-$CH_4$) primary inclusions with very strong intensity of $CH_4$. Secondary inclusions comprising mostly $H_2O$ with minor $CH_4$ are characteristically aligned along healed fractures and occur as clusters and trails (Fig. 2e). The auriferous brownish white quartz vein from GurharPahar (GPS-02) comprises mostly bi-phase $N_2$-$CH_4$-$CO_2$rich primary inclusions.

Methane Raman spectrum includes on band at 2913.18 $cm^{-1}$ corresponding to symmetric stretching of C-H (Dubessy et al., 2001). The Raman spectra of $CO_2$ consist of two intensive bands at 1387.20 $cm^{-1}$ and 1282.91 $cm^{-1}$, which corresponds to symmetric stretching and symmetric bending modes (Caumon et al., 2019). The Raman spectra ofgas phase $N_2$be made up of band occur at 2325.85 $cm^{-1}$ (Mamedov, 2017; Caumon et al., 2019).

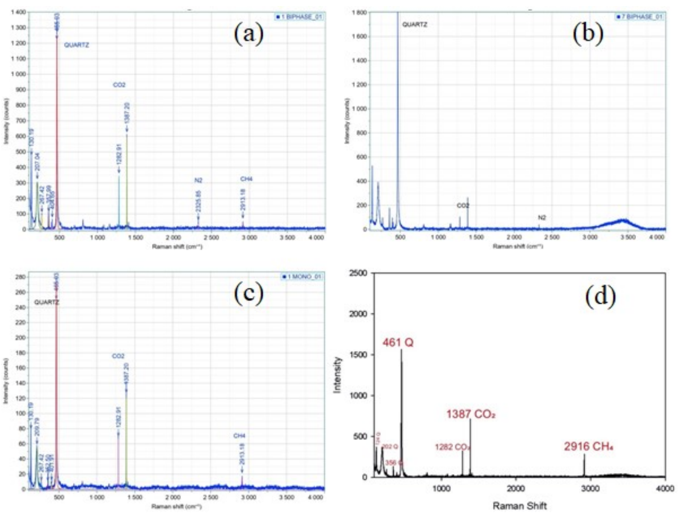


Fig. 3.Raman spectra of fluid inclusions in samples of quartz veins: (a) $CO_2$ rich inclusion with $CH_4$ and $N_2$ in sample GPS-02, (b) biphase aqueous inclusions with minor N2 in sample P-2, (c) $CH_4$ rich aqueous inclusion in sample P-3, (d) $CO_2$ rich inclusion with $CH_4$ in sample SP-1.

## V. Discussions

The auriferous quartz veins of the eastern MSB are enriched in $CH_4$ inclusions, besides $CO_2$ and water ($H_2O$) and minor traces of $N_2$. However, the barren milky white quartz vein intruding chlorite schist from Parsoi area containsonly aqueous carbonic ($H_2O + CO_2$) inclusions. The sample of quartz vein from SonaPahari is significantly enriched in the inclusions with $CH_4$. Secondary inclusions are observed only in quartz vein from Sonapahari. Quartz vein samples from GurharPaharcarrysignificant amount of $CH_4$ and $N_2$rich fluid inclusions. Fluid inclusion petrography and microthermometry was performed on mineralized quartz veins from GurharPahar and Guladih gold prospect by Prasad et al (2000). Earlier studies suggests that the homogenization temperature for the carbonic aqueous fluids is 318°C and 322°C for GurharPahar and Guladih quartz veins, respectively. The average $CO_2$ density of the fluids is 0.8 gm/$cm^3$, which indicates an average pressure of about 2.5kbar and temperature of 300°C is inferred from the carbonic aqueous fluids responsible for the gold mineralization in the GurharPaharand Guladih prospect (Prasad et al., 2000). The presence of $CH_4$ in the fluids can be inferred from the depression in the melting point of carbonic and carbonic aqueous inclusions (Hurai, 2010).

Previous fluid inclusion microthermometric studies on gold bearing quartz veins from the Sonapahari area was conducted by Tiwari and Singh (2009). Foregoing investigation suggest that the homogenization temperature for aqueous-carbonic inclusions varies in range of 178°C to 397°C and for aqueous inclusions homogenization temperature varies from 127°C to 435°C in mineralized quartz samples from the Sonapahari area. There are two compositional types of mineralizing fluids present ($H_2O$-$CO_2$-$CH_4$ ± $N_2$ and $H_2O$-NaCl ± $CaCl_2$ ± $MgCl_2$), which was trapped in the temperature range of approximately 275°C-300°C. The gold mineralization in the SonaPahari has been due to mixing of these two types of fluids (Tiwari and Singh, 2009).

In recent times, fluid inclusion microthermometric studies have been carried out on quartz veins from Phaphrakund areaDubey and Shankar (2017).These quartz veins have intruded in phyllitewhich reveal the bi-phase liquid-rich fluid inclusion possibly originated between the temperature range from 169°C to 256°C and derived from a magmatic moderately saline fluid (3.7 to 18.29 wt. % NaCl equiv.). The final ice-melting temperatures from Phaphrakund quartz veins ranges from -14.6°C to -2.2°C which indicate that the aqueous fluids are mainly contains $H_2O$-NaCl. The gold bearing fluids appear to be entrapped between pressure 1.6 to 2.1 kbar at depth of 200m, which possibly inferredan epithermal source (Dubey and Shankar 2017).

Fluid inclusion microthermometric and LRM investigations suggested that the gold mineralization in the eastern MSB linked within the temperature range of 250°C-300°C and pressure of 1.6-2.5 kbar. The auriferous ore fluid may be derived due to devolatilization of carbonatedsupracrustalmetvolcanics (Phillips and Powell, 2010; Bhattacharya and Panigrahi, 2015; Patten et al., 2020). Devolatilizationof carbonated supracrustalmetavolcanics released $CO_2$ rich fluids, which resulted in the precipitation of calcite and / or siderite in quartz veins (Fyfe et al., 1978). Availability of $N_2$rich fluid inclusions in mineralized quartz veins is caused by decomposition of $NH_4^+$ bearing silicate minerals during metamorphic processes of host rocks (Andersen et al., 1993; Moine et al., 1994; Poter et al., 2004). There are incidences of $CH_4$bearing fluid inclusions in the mineralized quartz veins, which may be due to contamination of the $CO_2$ with the metasedimentary rock during deposition (Guha et al., 1991; Ho et al., 1992)or as aeffect of continuous phase dissociation from the $H_2O$-NaCl-$CO_2$-$CH_4$ ± $N_2$ fluid (Naden and Shepherd, 1989; Pal et al., 2019). According to Mishra and Panigrahi (1999),the aqueous and carbonic phase immiscibility has played a vital role in the origin of mineralized fluid related processes. The homogenous ore fluid, rich in carbonic component with low salinity possibly favor transportation of gold during the deformation and metamorphism along the shear zones (Beach, 1976; Hodgson, 1989; Carter et al., 1990; Nassif et al., 2022). In the eastern MSB, the SNNF and SNSF have acted as channels for the transportation of ore fluid and quartz veins have been the relocation and entrapment sites for gold (Misra et al., 2021).

Thus, the compositional variation in fluid inclusions indicate the evolution and phase separation of the ore forming fluid during transportation through shear zone (Thompson and Connolly, 1990).Interaction of the original ore forming fluid with host rock, phase dissociation,and gangue mineral compositions favored in the gold deposition at suitable locales of shear zone(Petrella et al., 2021).

## Conclusion

The comparison of LRM data of fluid inclusions in quartz veins shows the variation in fluid environment of the prospects and represents the genetic aspects of gold mineralization. The gold mineralization at Parsoi, Sonapahari and GurharPahar is related to compositionally similar fluids, with enrichment of $CH_4$ during the evolution and mixing of the ore fluid. Ore fluid with composition of $H_2O$-NaCl-$CO_2$-$CH_4$ ± $N_2$ was responsible for gold mineralization in the eastern MSB. The quartz veins which possess $CH_4$bearingfluid inclusions are directly linked withauriferous mineralization. The presence of secondary aqueous carbonic inclusions indicates the syn-kinematic entrapment of the ore fluid along the fractures of auriferous quartz veins.

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