Role of hydrothermal fluids in the deterioration of pictographs and petroglyphs in rock shelters of the Gawilgarh Hills, Madhya Pradesh, India

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India has one of the largest concentrations of rock-art sites. However, these rock shelters have deteriorated due to natural agencies. The present study was carried out in one such site in Central India, which is hosted by argillaceous and ferruginous sandstones, showing prominent chemical, biological and mechanical weathering. The results based on field investigations, petrography, XRD, FE-SEM-EDS and FTIR measurements have indicated that chemical weathering is caused due to infiltration of hydrothermal fluids through innumerable zones of weaknesses formed due to neotectonic activities in the Satpura Lineament Zone, especially along Gawilgarh and Salbardi Faults in this region.

Keywords: Archeological site, chemical weathering, hydrothermal fluids, rock shelters.

EVER since the discovery of the first petroglyphs (rock engravings) in 1856 by W. G. Henwood in Almora district, Uttarakhand, India, curiosity about Stone Age culture grew in the country. This was followed by the discovery of the first Stone Age paintings (pictographs) at Sohagi Ghat in Mirzapur district, Uttar Pradesh, by Archibald Carlleyle in 1867–68, but reported much later by V. A. Smith in 1906 (ref. 1). Rock art in the form of petroglyphs and pictographs was subsequently reported from several places across India. Maharashtra and Madhya Pradesh in Central India are endowed with thick forests and distinct physiographical features formed by the Vindhyan and the Satpura mountain ranges and drained by perennial rivers such as Tapti and Narmada. The availability of sandstone cliffs together with the shelters carved out due to the weathering of surfaces and the easy availability of water were the main factors which made these ranges a preferred destination for the habitation of prehistoric man for a long time. Prominent rock-art sites along this >600 km long ridge cover the world-famous Bhimbetka[2], Jaora, Kathotia, Firangi, Mahadeo Hills, Kharwai, Bhopal, Raisen, Chaturbhujnath Nala and Daraki-Chattan[3].

In 2007, a group of amateur history enthusiasts and adventurists from nearby cities, reported the existence of rock art in a few shelters in the vicinity of a local shrine of Ambadevi[4]. Studies then began about the published and discovered rock shelters[5]. Thereafter, work was initiated by the Archaeological Survey of India's (ASI) Prehistory and later Excavation Branch I, Nagpur, Maharashtra, as an attempt to revisit the site. The intense field work by ASI, across a span of four years, yielded decorated rock shelters, comprising paintings as well as rock engravings. The repertoire of decorated rock shelters, thus discovered, numbering 247, was divided into 21 groups across an area of nearly 40 km. Apart from this, more than 500 undecorated shelters were also found. The hitherto debated geographical positioning of the shelters was ascertained and fixed at the Atner and Multai tehsils of Betul district in Madhya Pradesh. The various geographical sources helped ASI identify the hill range housing the decorated shelters as the Gawilgarh Hills[6–9].

Destruction and deterioration of pictographs and petroglyphs by natural or artificial agencies is known and studied world over. The most common agencies causing deterioration of pictographs and petroglyphs are weathering, mechanical destruction, fungus, lichens, animal excreta, insect colonization and trickling of waters inside caves. In this study, we discuss the role of hydrothermal solutions in the deterioration of pictographs and petroglyphs in a rock shelter in the hydrothermal field along the Satpura mountain range in Central India.

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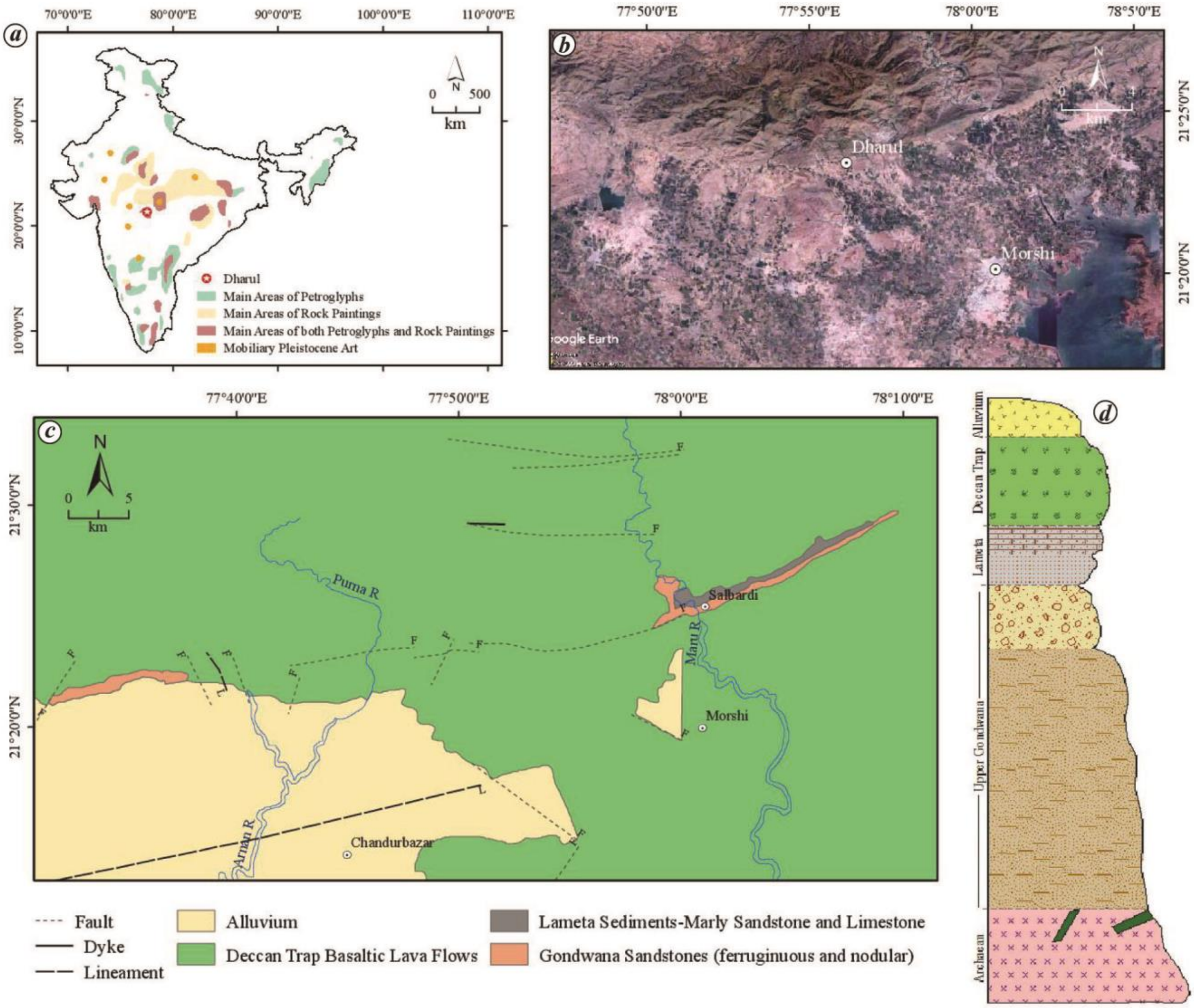

Figure 1. ***a***, Geological map of Dharul–Ambadevi and surrounding areas showing the location of Dharul and other prominent archaeological sites in India. ***b***, A Google Earth image showing the location of Dharul (accessed on 12 May 2020). ***c***, Geological and structural features of Salbardi and surrounding areas (redrawn after Srivastava and Mankar[12]). ***d***, A lithosection map showing stratigraphic succession in the study area (modified and redrawn after Srivastava and Mankar[12]).

Geology of the area

The Satpura mountain is one of the most prominent geomorphic features of the Central Indian craton. This mountain is bounded on both sides by regional-scale faults – in the north by the Son–Narmada South Fault (SNSF) and in the south by the Gawilgarh Fault Zone (GFZ) (also known as Satpura Foothill Fault, SFF)[10,11]. The Satpura hill ranges are dominantly composed of sandstone belonging to the Gondwana Supergroup of rocks. These sandstones are a part of the upper Gondwana succession along with the Lameta Group of rocks[12]. The succession shows good preservation of primary sedimentary structures indicating facies variation and the environment of deposition. The Gondwana sediments rest unconformably over the basement gneisses[12]. The Deccan trap basalt covers the top of hill ranges as capping on the Gondwana sediments and shows sub-horizontal to gently northern dips (5°–20°)[13–15].

The area is traversed by many fault systems running in the NE–SW directions with sympathetic cross-faulting. The main fault, i.e. SFF forms an escarpment in the area, and upliftment and juxtaposition the Gondwana sediments with gneisses and Quaternary sediments. This fault system is normal, bounded by the Satpura horst towards the south and the sympathetic Tapti valley, which has formed a ~1 km high scarp running for hundreds of kilometres. They are termed as the Salbardi, Gawilgarh and Ellichpur faults[16], and SFF[17]. The Tapti valley exposes Deccan basalt-derived Tertiary and Quaternary alluvium, with basalt inliers ~200 m amsl[18].

The study area comprises parts of Amravati district, Maharashtra and Betul district, Madhya Pradesh (Figure 1), covered under the Survey of India toposheets numbered

55 K/3 and 55 G/15. The area is bounded by longitude 21°25′ to 23°15′E and latitude 76°0′ to 78°5′N. The Salbardi and adjoining areas are marked by low-altitude hillocks ranging between 320 and 692 m elevation. The rocks exposed in this area are basalt, sandstone, gneiss, conglomerate and shale. The Dharul–Ambadevi area exposes a variety of rocks from Archean gneisses to Gondwana sandstone and end Cretaceous–Paleocene Deccan traps due to their tectonic disposition. A sharp contact between different litho-units is due to the ENE–WSW trending Salbardi fault crossing the area. The exposed Gondwana sediments in the area represent ~100 m thick succession, dominantly composed of medium to coarse-grained sandstone, followed by clay and pebbly horizons. The Deccan Trap basaltic lava flows are spread over the Gondwana sediments. The Archean Granitic Gneiss occurs as a basement for all the younger rocks such as Gondwana sediments, Deccan Trap, Lameta beds and the Alluvium[19,20].

Sampling and analytical techniques

Among the samples collected, six were selected for petrographic analysis and four (KR/525, KR/526, KR/527 and KR/530) for X-ray diffraction (XRD), field emission scanning electron microscope (FESEM), Fourier transform infrared spectroscopy (FTIR) analyses.

Thin-section study was carried out at the Petrology Division, Geological Survey of India (GSI), Nagpur using OLYMPUS BX 51 Trinocular Polarizing Microscope with CCD camera and basic stream software. XRD analysis of selected samples was performed (Panalytical X'Pert Pro (model-PW 3040/60 diffractometer)) with CuKα radiation (λ = 1.54 Å) at a voltage of 40 kV and current of 40 mA. Scanning was done in the 2θ angle range 5° to 80° with a scan step size and time per step of 0.01° and 15 s respectively. The morphological changes were studied using a scanning electron microscope (SEM-JEOL 6830A). Before the study of surface morphology, the material was coated with a thin layer of platinum using an auto-sputter (JOEL-JFC 1600 auto fine coater) to make the material conducting for obtaining images. FTIR spectra of samples were recorded (Perkin Elmer Spectrum One FTIR). Potassium bromide (KBr) was used for collecting the background and a total of 20 scans was taken.

Petrography

Petrographic study of the sandstones was carried out at the Petrology Division, GSI, Nagpur and the Department of Geology, RTM Nagpur University, Nagpur. A detailed description of the same is given in the Supplementary Material, and only results of the study are reported here.

Results

X-ray diffraction study

Four samples were analysed for characterizing the minerals present in the rocks. However, the results were far from simple and needed more detailed characterization comparing with the results obtained using other methods. The peak intensities corresponding to 2θ angles were located at 20.76°, 26.57°, 36.51°, 39.41°, 42.35°, 50.05°, 59.86° and at 68.05° which represents quartz as the most commonly occurring phase. However, presence of minerals such as antigorite (alteration of minerals such as olivine) at 25°, zinnwaldite (possibly muscovite) represented from 26°, 35° and 44° and graphite (a polymorph of carbon) at 26°, 42°, 45°, 51°, 55°, 60° and 80° peaks are also conspicuous. Figure 2 shows the XRD peaks for samples KR/525, KR/526, KR/527 and KR/530.

FESEM aided with energy dispersive X-ray spectroscopy

The FESEM studies have been useful in predicting the micro-morphologies of the rocks and energy dispersive X-ray spectroscopy (EDS) was useful in predicting the chemical composition of the minerals studied. The host rock (sandstone) is dominantly composed of sand grains (mainly quartz, but feldspar is also present). The micro-morphology of the rocks indicates the presence of anhedral, broken crystals of quartz over which several microcrystals have formed (Figure 3). Well-developed (idiomorphic) six-sided grains of quartz are also common (Figure 3 *a–c*). The clay minerals form platy and stacked aggregates, which are either straight or curvilinear (Figure 3 *d–f*).

Apart from these, several other forms have also been noticed. However, their composition remains largely unidentified, except for some general approximations. Such forms notably include: (i) a flower-like aggregate of hexagonal flakes in which some of them have curved inwards forming a bowl-shaped structure (Figure 3 *g*); (ii) tiny spherules (possibly moganite, a polymorph of quartz having monoclinic symmetry) and cryptocrystalline silica which represent neo-mineralization after dissolution–precipitation (Figure 3 *h*), and (iii) mineral fibres, possibly antigorite as predicted in XRD analyses (Figure 3 *i*). The EDS analysis indicated overwhelmingly abundant silica (Figure 3 *e* and *f*) with minor quantities of aluminosilicates (Figure 3 *g* and *h*), and the presence of carbon and possibly iron.

Fourier transform infrared spectroscopy

The same samples (used for XRD study) were analysed for FTIR. The results show similarity for the Si–O

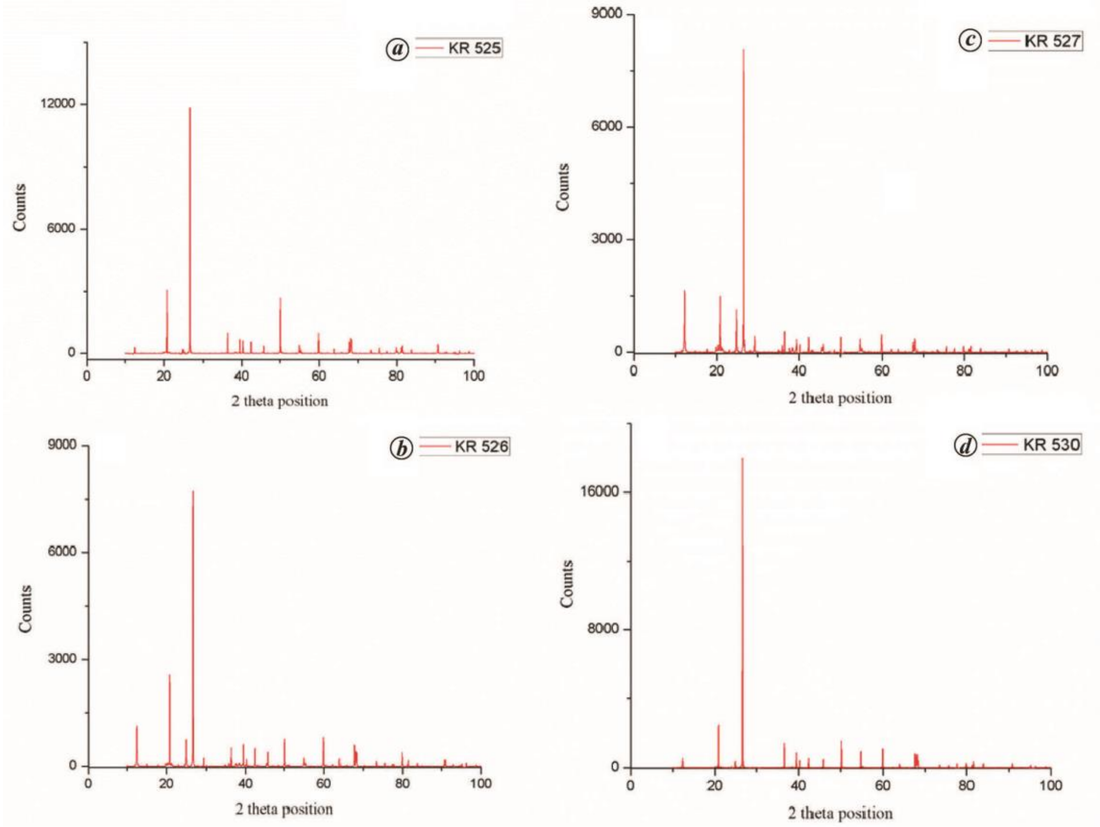

Figure 2. X-ray diffractograms of the studied samples. ***a***, KR/525 showing peak for quartz. ***b***, KR/526 showing peak for antogorite (a serpentine group mineral) and quartz. ***c***, KR/527 showing peak for muscovite and calcite. ***d***, KR/530 showing peak for antogorite, quartz and graphite.

functional group indicating the presence of quartz (Figure 4). In the mid-infrared region quartz shows peaks at 511, 694, 777, 796, 1057, 1079 and 1163 cm^{-1}. Corresponding to these peaks in the fingerprinting region of FTIR, the common peaks shown by the samples include 695, 779–781, 794–795 and 1033–1040 cm^{-1}, whereas peaks equivalent to 1079 and 1163 cm^{-1} are missing. Nevertheless, the following peaks are significant: 1111 cm^{-1} showing Si–O–Si asymmetric stretching and/or Si–O–C band, 1113 cm^{-1} showing bending mode of C–C stretching, 1319–1320 cm^{-1} representing the vibration peak of C–H bends, 1494–1504 cm^{-1} showing asymmetric O–H stretching and 3696–3697 cm^{-1} showing stretching of O–H bond. Whereas in the FTIR spectrum of pristine graphite no significant peaks relevant to any functional groups were found (Figure 4), but weak bands may appear which can be assigned to adsorbed water molecules[21].

Discussion

Agencies responsible for the deterioration of pictographs and petroglyphs

The pictographs were drawn using the material available in the surrounding areas. The pigments used here might have been completely inorganic or mixed with organic pigments in certain cases. The main pigment across the various cultures in the Gawilgarh Hills shelters appears to be hematite or some of its derivatives. However, at a few places, green, white, black and yellow pigments were also used.

The climatic, environmental and geological conditions and changes in vegetation pattern over time influence the relative deterioration of pictographs and petroglyphs. Also, the panel orientation and pigment mineralogy are factors which otherwise influence the physical and chemical conditions of individual rock-art panels. According to studies, avian guano can chemically consolidate pigments and ensure their sustainability, as seen at the Walga Rock in Western Australia[22]. Also, the shelter projections to some extent may have preserved the rock art and protected it from various weather conditions. However, the shelters and in some cases the art too, have been subjected to deterioration by combined chemical and mechanical weathering. The seepage of subsoil water through weak planes such as fractures and faults planes in the shelter wall causes chemical weathering. The pictographs are also being abraded due to tricking fluids (Figure 5 *a*–*c*), all of which is not rainwater, but a mixture of dissolved material such as atmospheric CO_2 and

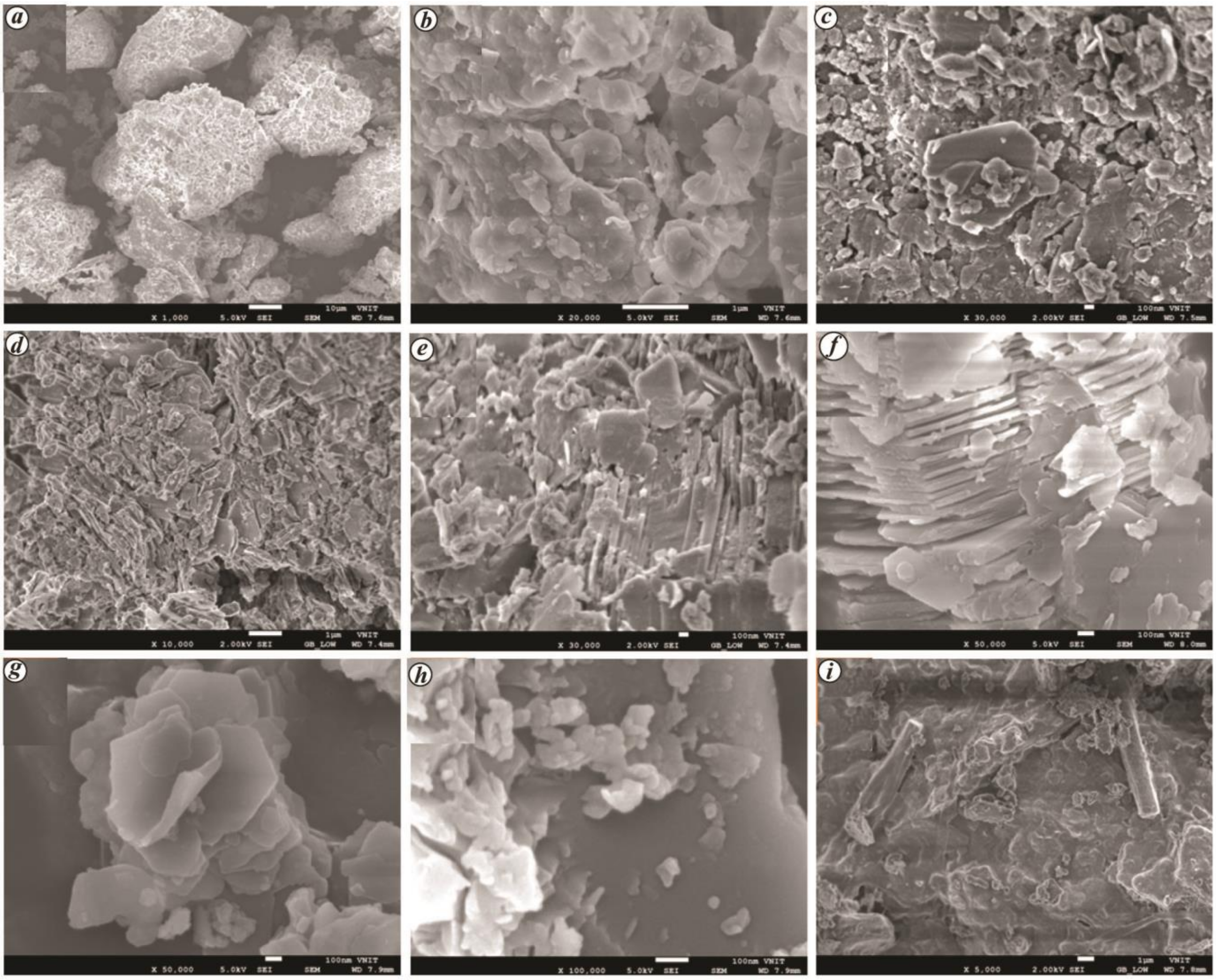

Figure 3. Secondary electron (SE) images showing a variety of morphological features, textures and microstructures of the host rock. ***a***, Pulverized rock samples at lower magnification (×1000). Some of these grains have been magnified. ***b***, Typical morphology of hexagonal-shaped quartz crystals. More than 90% of this rock (sandstone) is composed of quartz (×20,000). ***c***, Further magnification (×30,000) showing hexagonal quartz microcrystal in the middle. ***d***, An aggregate of clayey minerals in the sample (KR/525) collected from inside the cave (×10,000). ***e***, Magnified view of the clayey aggregate in another sample (KR/527) showing stacks of phylosilicates (×30,000). ***f***, Highly magnified view of the same minerals (×50,000). ***g***, Flower-like or inverted bowl-like morphology formed due to curvature in the hexagonal flakes (×50,000). ***h***, Small spherulitic structure of moganite (monoclinic polymorph of quartz) at very high magnification (×100,000). ***i***, Fibrous morphology observed in some parts (×5,000).

silica-rich hydrothermal fluids. Petroglyphs (pecking, engraving, carving, bruising and cupules), on the other hand, have been affected more by mechanical weathering of the host rock (Figure 5 *d* and *e*). However, the role of chemical as well as biological components also appears significant. Chemical weathering processes triggered by hydrothermal fluids at the geological surface caused decomposition and discolouration, and altered the rocks. Besides, biological growth has penetrated the rocks and covered the pictographs and petroglyphs. It also acts as a catalyst for other weathering processes by retaining water and disintegrating the underlying layer[23]. As rock surfaces contain biotic substances and organic carbon, these may account for the significant colour changes. It, therefore, appears that all three components, namely physical (or mechanical), chemical (active fluids) and biological (e.g. fungi) have been responsible for natural destruction and deterioration of the pictographs and petroglyphs.

Hydrothermal alteration of the host rock

The Gawilgarh Hills are a part of the SFF zone, which is marked by low-altitude hillocks ranging from 320 to 692 m facing the southern Deccan peninsula. It forms small ridges with the development of depressions in the adjacent area, which act as a water-holding structure. These faults also provided loci for the genesis of hot

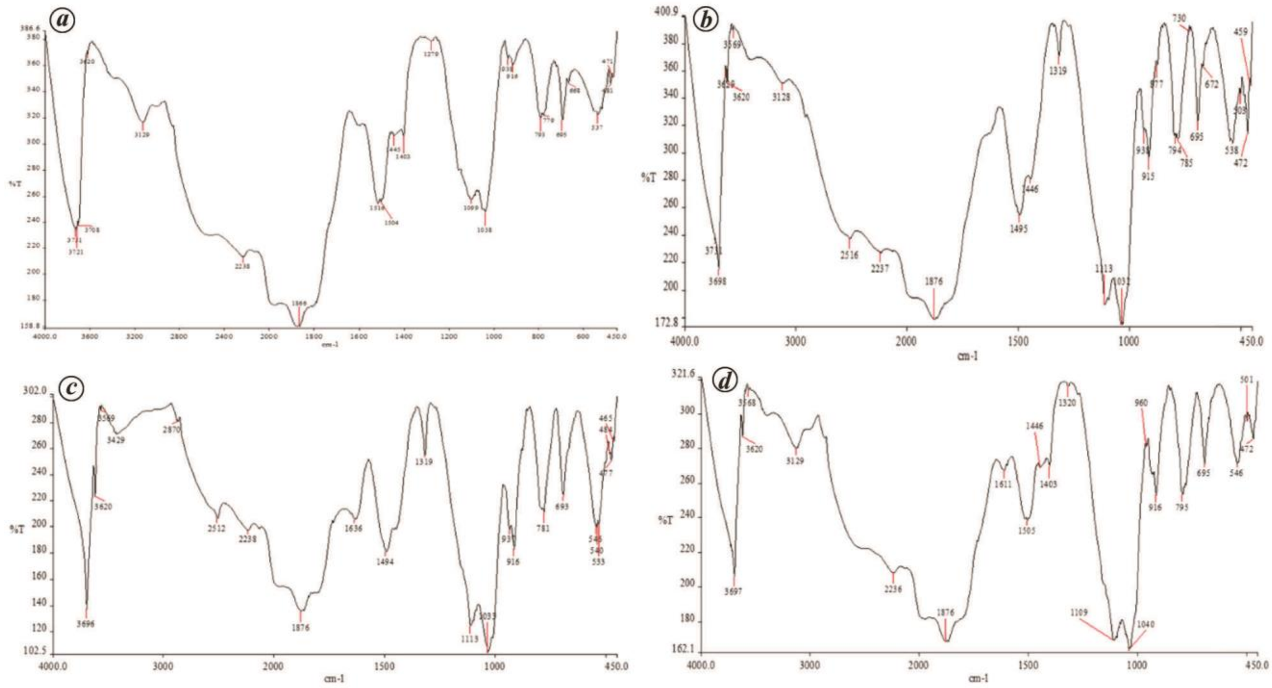

Figure 4. Fourier transform infrared spectra of samples: ***a***, KR/525; ***b*** KR/526; ***c***, KR/527; ***d***, KR/530.

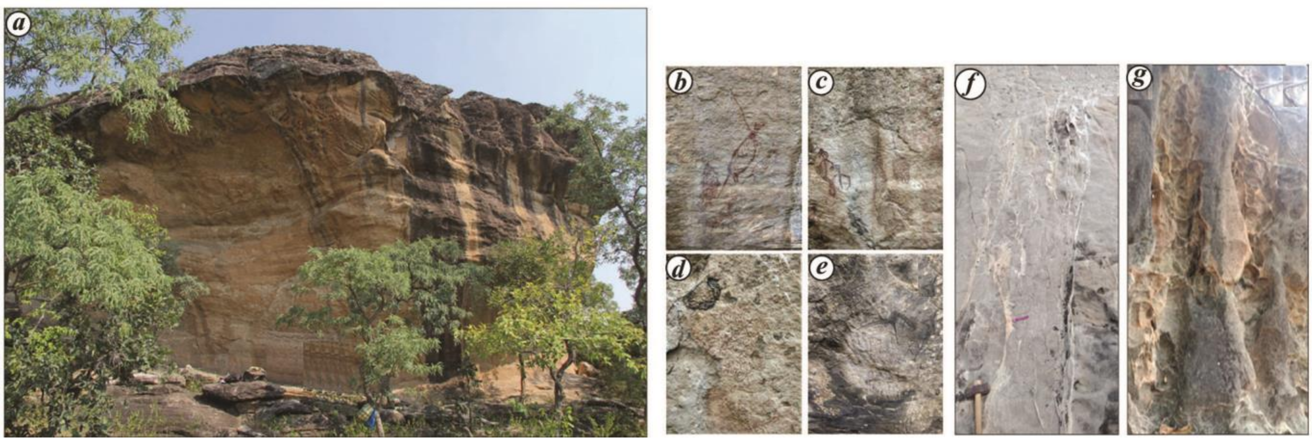

Figure 5. ***a***, View of one of the rock shelters in Ambadevi area. ***b***, Pictograph showing pangolins and other animals; some paintings have been destroyed due to hydrothermal fluids. ***c***, Another set of pictographs showing a part faded out due to tricking fluids. ***d***, Part of a pictograph showing a honey comb that is broken due to fragile surface. ***e***, A petroglyph mechanically fragmented along the rock cleavages, and also attacked by chemical and biotic components (dark-coloured material). ***f***, Quartz-rich vein system in a sandstone-hosted fault network/vein-fault network criss-crossing veins filled with secondary silica-rich fluids. ***g***, Chemical weathering of rock surface due to dissolution of minerals forming of pits, cavities, hollows and box work patterns of indurated joints (view roughly covering 1.5 × 0.5 m).

springs at Salbardi, on the left bank of River Maru[19,24]. Due to neotectonic activities in the study area, a fracture system has developed. This has also activated the hydrothermal system in the region evidenced by the presence of thermal springs[16,25]. A network of meso- to micro-scale veins, often crisscross, either open or sealed, occurs throughout the area. Majority of them are resistant to erosion; they are hard, brittle and break with conchoidal fractures, and are composed of quartz and/or chalcedonic silica (Figure 5 *f*). Another indication of hydrothermal alteration is the presence of ovoid cavities and pits in the host sandstone (Figure 5 *g*), which is a feature related to dissolution and precipitation of the constituent material in the rocks. Since the country-rock sandstone has marly, clayey and calcareous cementing material, this cement would have been removed by dissolution due to interaction with the hydrothermal solutions. It is chemically charged water saturated with bicarbonate and hot having thermal gradients ranging from 38°C to 42°C (ref. 24). The pathways of infiltration of fluids through the walls

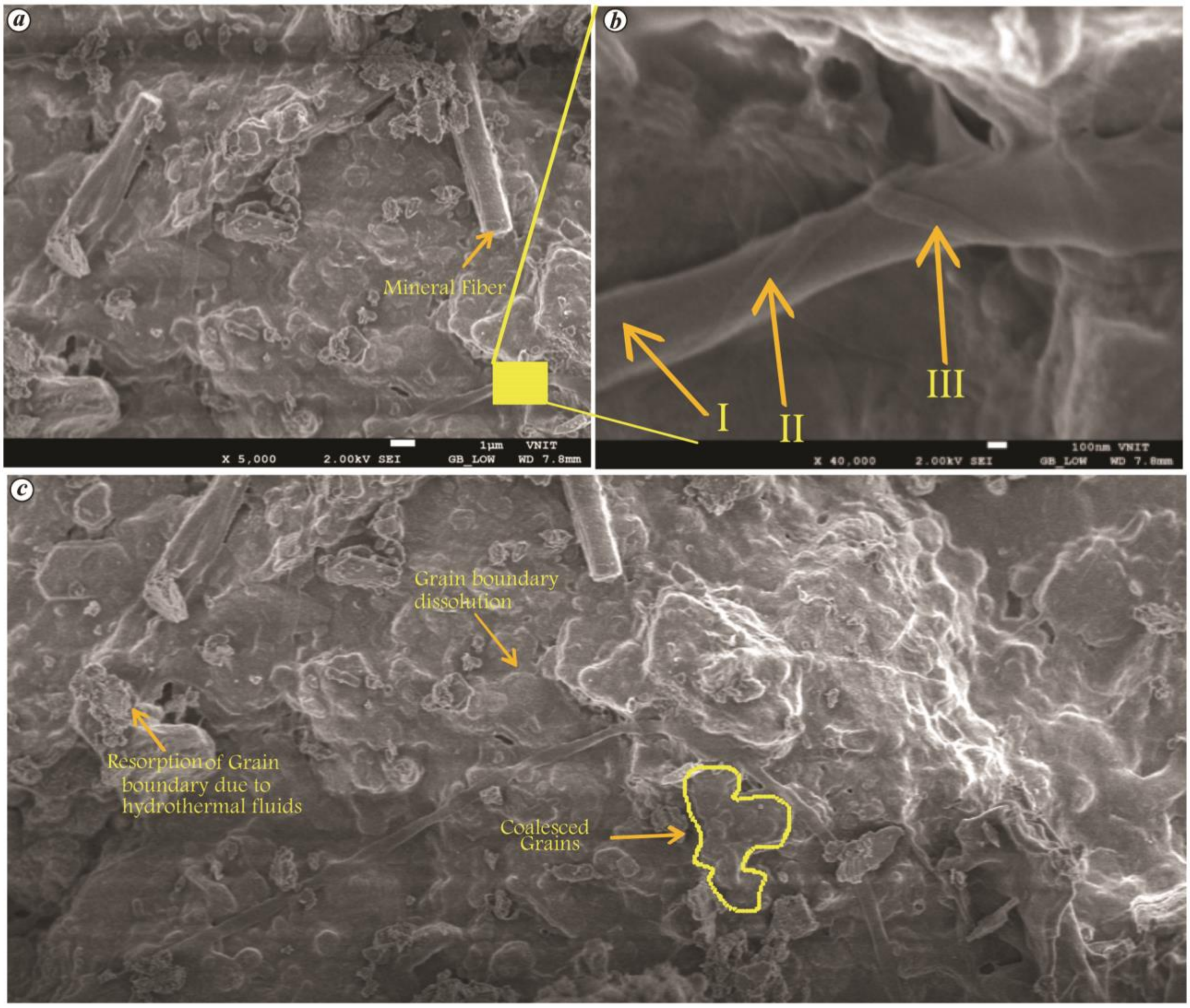

Figure 6. SEM image showing the influence of hydrothermal fluids and subsequent changes in morphology and texture of the original rock. ***a***, Magnified (×5000) image of sample KR/525 showing the host rock for wall paintings inside the cave. Highly magnified (×40,000) image of the hydrothermal veins is shown in inset (***b***). ***c***, Lower-magnified image (×2500) of the same sample. The highly magnified image (***b***) shows the main vein (I) and another smaller vein (II) cutting across it, and a third vein (III) cutting both the veins implying multiple generations of veins influence the area due to neotectonic activities. The grain boundary dissolution and transfer of components into the veins, resorption of grain boundary and coalescence of multiple grains resulting into formation of single grain can be seen in (***c***).

and roofs of the shelters (vertical bands descending on the surfaces in Figure 5 *a*), also indicate a strong imprint of hydrothermal activity and solution transfer in the area.

Altered portions of the roofs and walls of the shelters hosting pictographs and where dissolution, decolourization and impregnation of other liquids caused damage to the original paintings were collected to ascertain their composition. The results of XRD analysis show prominent peaks of quartz, which is an abundant mineral, dominating the overall spectra. Similarly, several bands in the FTIR spectra show Si–O functional groups indicating quartz. However, additional peaks of C–C and C–H stretching might be related to biotic components present on the altered surfaces inside the shelters. More evidences emerge from FESEM images in which well-developed, idiomorphic, hexagonal grains of quartz are present (composition ascertained from EDS). However, the quartz grains are often fragmented, deformed (forming 'flower-like' or 'bowl-shaped' morphology; Figure 3 *g*), and a polymorphic transformation to the monoclinic structure has been indicated by moganite (Figure 3 *h*). The 481, 695, 779–781 and 793–795 cm^{-1} bands also confirm the presence of moganite. The FTIR spectra further show asymmetric as well as normal O–H stretching, which forms Si–OH bonds, confirming the presence of cryptocrystalline silica or chalcedony[26]. However, the presence of calcite might indicate that this mineral developed due to precipitation of hydrothermal solutions coming from the limestone of upper Lameta Formation[12].

Antigorite is a Serpentine Group mineral which is formed during metamorphism of hydrous ultramafic rocks; its presence in the altered milieu is not expected. It could have been formed due to the *in situ* hydrothermal alteration of detrital pyroxenes in the Gondwana sandstone. Traces of zinnwaldite are indicated by the XRD spectra; however, there is no further indication of this mineral in either FTIR or SEM–EDS. However, it is known that muscovite can be erroneously interpreted as zinnwaldite if the sample is not properly oriented[27]. In a composite sample being studied here, there is possibility of such misorientation. Therefore, we prefer muscovite over zinnwaldite as a mineral in the given assemblage. However, charcoal might be original; it was commonly used for paintings by the Palaeolithic people.

Figure 6 demonstrates the influence of hydrothermal solutions at the nanoscale. At lower magnification (×2500), the micro veins have induced grain-coalescence, dissolution and resorption of grain boundaries (Figure 6 *c*). Whereas at higher magnification (×5000), the surface morphology indicates the presence of different generations of microveins (Figure 6 *a*). At much higher magnification (×40,000), three prominent veins crossing each other can be seen. A major vein, designated as vein-I, appears to be the oldest. It is cut across by vein-II, and both these veins are cut across by vein-III (Figure 6 *b*). This indicates that (i) there is a profound influence of hydrothermal solutions in the dissolution of the wallrocks, and (ii) there is also the influence of different episodes of the hydrothermal solutions. These observations, therefore, indicate continuous activity in this region related to neotectonism. Consequently, these evidences show that hydrothermal solutions have a major role in the destruction of the rock art in the Gawilgarh Hills area. However, further analytical study is needed to elucidate the relationship between the role of hydrothermal fluids and the conditions for rock-art preservation.

Conclusion

The study of the Gawilgarh Hills rock shelters has revealed that pictographs and petroglyphs in the shelters hosted by the Gondwana sandstones are under severe threat of environmental destruction together with human activities. There are innumerable fractures which form a dense network of veins, the presence of hot springs and neotectonics activities such as active faulting indicate an active hydrothermal system in this area. It is therefore evident that this heritage in the wilderness of the Satpura mountain range in Central India is under severe threat by human and natural agencies. Although there are several known factors such as weathering, trickling water, fungus, animal excreta, etc., hydrothermal fluids were not so far known as the probable source of destruction of these pictographs and petroglyphs.

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ACKNOWLEDGEMENTS. This study was carried out as a collaborative research programme between Indira Gandhi National Centre for the Arts (IGNCA), New Delhi, as the funding agency and RTM Nagpur University, Nagpur, under an MOU signed in 2016–17 for a project titled 'Multidisciplinary documentation of rock art and its allied subjects in Maharashtra and adjoining regions' awarded to P.S. We thank IGNCA, New Delhi and RTM Nagpur University, Nagpur for the necessary permissions, facilities and funds to carry out this study.

Received 25 October 2020; revised accepted 31 May 2021

doi: 10.18520/cs/v121/i2/255-263