

A Modified Placer Origin for Refractory Gold Mineralization Within the Archean Radioactive Quartz-Pebble Conglomerates from the Eastern Part of the Singhbhum Craton, India

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Abstract

Archean quartz-pebble conglomerates are one of the most extensively studied stratigraphic horizons and are globally well known for their gold and uranium prospects of economic grades. Similar quartz-pebble conglomerate horizons have been delineated within the Neoarchean-Paleoproterozoic Gorumahisani-Badampahar schist belt and Dhanjori volcano-sedimentary basin lying along the northeastern fringe of the Singhbhum-Orissa craton in eastern India. Gold and uranium occurrences have been reported previously from the quartz-pebble conglomerates of the Singhbhum craton; however, no detailed report on gold occurrences and genesis is available. This work focuses on the nature and genesis of gold mineralization hosted by radioactive quartz-pebble conglomerates from the eastern Iron Ore Group and the Dhanjori basin within the Singhbhum-Orissa craton. The quartz-pebble conglomerates reported in this work are predominantly matrix supported and oligomictic in nature. Petrography, microprobe data, and scanning electron microscopy studies reveal the presence of chromite, zircon, rutile, magnetite, monazite, pyrite, chalcopyrite, pyrrhotite, stibnite, and brannerite. Laser ablation-inductively coupled plasma-mass spectrometry analyses on various generations of arsenian pyrites reveal significant concentrations of Au in solid solution. The gold was initially incorporated in its detrital form onto growing pyrite surfaces, possibly due to microbial mediation. The trace element data coupled with back-scattered electron images and X-ray compositional maps suggest remobilization of detrital Au and associated trace elements from the diagenetic pyrites and their precipitation onto the younger hydrothermal pyrites. The sulfide-hosted gold mineralization, trace element signatures, and zoning patterns studied in this work mimic the mineralization style reported from sediment-hosted gold deposits such as Spanish Mountain and Carlin. However, in broad terms, the origin of Au within these quartz-pebble conglomerates resembles the modified placer model of gold deposition within the Witwatersrand conglomerates. This is the first report of refractory gold mineralization within arsenian pyrites from the radioactive quartz-pebble conglomerates of eastern India and has important implications for gold exploration in this region and analogous areas.

Introduction

Sediment-hosted gold deposits account for the majority of annual gold production of the world. The Archean Witwatersrand basin in South Africa hosts the world's largest gold reserves and has produced over 48,000 metric tonnes of gold since 1886 (Phillips and Law, 2000). Gold within the Witwatersrand basin is hosted in fluvial quartz-pebble conglomerates deposited between 2.9 and 2.84 Ga (Frimmel et al., 2005). Similar types of quartz-pebble conglomerate-hosted gold mineralization have also been reported from various parts of the world, mostly within the Archean cratons, forming siliciclastic placer deposits resting unconformably on the Archean granite-greenstone terrains (Chakrabarti et al., 2013). In addition to gold occurrences, the Archean-Proterozoic quartz-pebble conglomerates are also favorable horizons for uranium mineralization. The presence of gold, silver, platinum group elements, uraninite, and other radioactive minerals has also been reported in similar types of conglomeratic horizons (Kumar et al., 2017). In addition to the economic significance of Archean-Proterozoic quartz-pebble conglomerates, understanding their genesis provides useful insights into their depositional history, paleogeography, and paleoclimate (Cox et al., 2002; Kumar et al., 2017).

Different workers have proposed various theories regarding the origin of gold within the quartz-pebble conglomerates

of the Witwatersrand basin from time to time. However, the debate remains unresolved as to precisely how the gold was incorporated into the conglomerates. According to the placer model, gold was mechanically transported from older granite-greenstone source areas and subsequently concentrated within the conglomerates as detrital gold particles by fluvial-deltaic processes (Pretorius, 1974; Hallbauer and Utter, 1977; Kirk et al., 2001). As per the metamorphic-hydrothermal model, gold was incorporated into the basin by hydrothermal-metamorphic fluids. Gold was initially transported in solution from outside the basin, and the fluid infiltration occurred after basin sedimentation was complete (Phillips and Law, 1994; Barnicoat et al., 1997). The modified placer model proposes that gold was incorporated within the basin, as per the placer model, but subsequently mobilized and reprecipitated along with other associated sulfides at deposit scale by infiltrating hydrothermal fluids (Frimmel and Gartz, 1997; Minter, 1999).

However, Horscroft et al. (2011) pointed out that the Witwatersrand basin contains more gold than any probable source rock in the vicinity from which the gold might have been derived, as previously suggested by the modified placer hypothesis. Horscroft et al. (2011) suggested a modified syngenetic model wherein the gold and uranium were transported in solution due to complex and aggressive chemical reactions in a low-temperature environment. The development of microbial mats over paleosurfaces during hiatuses in sedimentation acted as a catalyst for further concentration of gold and

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uranium. The biochemical concentration of gold is thought to be a result of extracellular fixation by living microbes or due to dead microbes, which provided activated carbon for passive adsorption of gold. These gold particles were subject to further reworking and downslope mobilization due to locally active fluvial processes. Hence, the syngenetic model resolves the problem of the presence of an enormously endowed gold-rich source area for the Witwatersrand gold ores.

Heinrich (2015) postulated that the Witwatersrand ores could have formed only during the Archean, after the occurrence of continental life but before the emergence of oxygen in the Earth's atmosphere. This model suggests the chemical transport of dissolved gold in Archean river waters from a gold-rich source region onto the Witwatersrand basin. The presence of reduced sulfur in Archean river waters played the primary role in gold dissolution and transport. Based on thermodynamic modeling and mass balance calculations, Heinrich (2015) established that the sulfur from basaltic eruptions significantly modified the rainwater composition and the chemistry of land surface during the deposition of Witwatersrand gold. The sulfur-rich basaltic eruptions resulted in acid rain that acted as a catalyst for the transport and dissolution of gold as sulfur complexes in surface waters. Subsequent precipitation of gold was initiated by the reduction of the dissolved gold onto organic matter in shallow water bodies.

Large et al. (2013) suggested a model wherein detrital gold and pyrite were sourced from a fertile basement hinterland followed by sedimentary reworking and a subsequent basin-wide hydrothermal fluid infiltration, resulting in the final upgradation of gold within the Central Rand Group of the Witwatersrand basin. The model suggests that the gold and pyrite were derived from the underlying intrabasinal carbonaceous shales and sandstones by sedimentary reworking, rather than from basement granites or metamorphic rocks in the hinterland. This was followed by a hydrothermal event that resulted in remobilization of the synsedimentary Au, As, Ni, Te, and other trace elements through the Central Rand Group, finally resulting in the enrichment of gold. The hydrothermal event is thought to be a consequence of the emplacement of the Bushveld Igneous Complex.

In India, quartz-pebble conglomerate-type gold and uranium mineralization was previously reported from the Dharwar and Singhbhum cratons (Rama Rao, 1974; Chakrabarti et al., 2013; Mukhopadhyay et al., 2014). In the Dharwar craton, the Bababudan and Chitradurga Groups are considered as good prospects for quartz-pebble conglomerate-type uranium mineralization (Srinivasan and Ojakangas, 1986; Fareeduddin et al., 1988; Radhakrishna, 1988). Uranium occurrences have been reported from conglomerates of the Dhanjori basin and Bonai Group within the Singhbhum craton (Rao et al., 1988; Kumar et al., 2012). The Mahagiri-Keonjhar-Mankarchua siliciclastic rocks in the western part of the Singhbhum craton also host quartz-pebble conglomerate-type gold and uranium mineralization (Mukhopadhyay et al., 2015).

In this work, quartz-pebble conglomerates from the eastern Iron Ore Group and the basal succession of the Dhanjori basin have been studied. Gold values of 0.1 to 2 g/t are reported from the quartz-pebble conglomerates of the Suleipat area within the Gorumahisani-Badampahar belt (Sarangi and Mohanty, 1987; Iyenger, 1995) and 0.4 to 0.8 g/t are reported

from quartz-pebble conglomerates of Turliga Parbat (part of study area) (Das et al., 1988). Average whole-rock gold values of 1,200 and 2,400 ppb have been previously reported from the lower Dhanjori quartz-pebble conglomerates, which are also a part of the present study (Chandra et al., 1995, in Ghose, 1996; Singh et al., 1992, in Ghose, 1996). Detailed mineralogical characterization, scanning electron microscopy (SEM), energy dispersive spectrometry (EDS), electron probe microanalysis (EPMA), laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS), and whole-rock analyses using ICP-MS were carried out to understand the mineralogy, nature of the conglomerates, occurrence of Au and U, nature of Au mineralization, and the overall economic potential of the quartz-pebble conglomerates. We report the presence of lattice-bound Au within arsenian pyrites of diagenetic as well as hydrothermal origin. The Au values are very promising, and this is the first time lattice-bound gold is being reported from the quartz-pebble conglomerates of eastern India. The conglomerates are fairly similar to those of the Witwatersrand basin; however, the nature of mineralization and the trace element characteristics are similar to some sediment-hosted gold deposits. The enrichment of Au in the diagenetic and sedimentary pyrites point toward microbial fixation of gold onto the pyrite surfaces. This makes the work all the more significant, especially in understanding the primordial life forms vis-à-vis gold precipitation, the paleoenvironment of deposition of metals like Au and U, and clues to early life on Earth (Sahoo and Venkatesh, 2014; Tashiro et al., 2017). The detailed description of the mineral phases within the Archean quartz-pebble conglomerates also has implications for the paleoredox conditions. In addition to gold, we also report the presence of brannerite and minor uranium (whole-rock ICP-MS data) from the studied conglomerates.

Regional Geology

The Singhbhum craton constitutes a complex geologic assemblage of Archean and Proterozoic age in eastern India (Fig. 1). It is among the five Archean cratonic nuclei within the Indian shield. The Singhbhum cratonic part includes tonalite-trondhjemite-granodiorite (TTG) batholiths and middle- to late-Archean supracrustal greenstone sequences, termed the Iron Ore Group, intruded by late-Archean granites (Bhattacharya et al., 2014; Mukhopadhyay et al., 2015). The craton in its northern part is traversed by a crustal-scale shear zone, which marks the boundary between the North Singhbhum mobile belt and the Singhbhum cratonic nuclei. The granite-greenstone terrains within the core of the craton are collectively represented by the Singhbhum granite (Ghosh et al., 2016), which is a suite of TTG granitoids with three phases of granite emplacement (SBG I, II, III) and the Older Metamorphic Tonalite Group. The Older Metamorphic Group comprises metasedimentary and metavolcanic rocks that occur as enclaves within the Older Metamorphic Tonalite Group.

The Older Metamorphic Group forms the largest supracrustal enclave within the Singhbhum craton. It includes amphibolites, calc-silicate rocks, mica-schist, and sparsely distributed fuchsite quartzite (Roy and Bhattacharya, 2012). The intrusive granitoids within the Older Metamorphic Group are termed the Older Metamorphic Tonalite Group. They are mostly tonalitic-trondhjemitic and granodioritic in

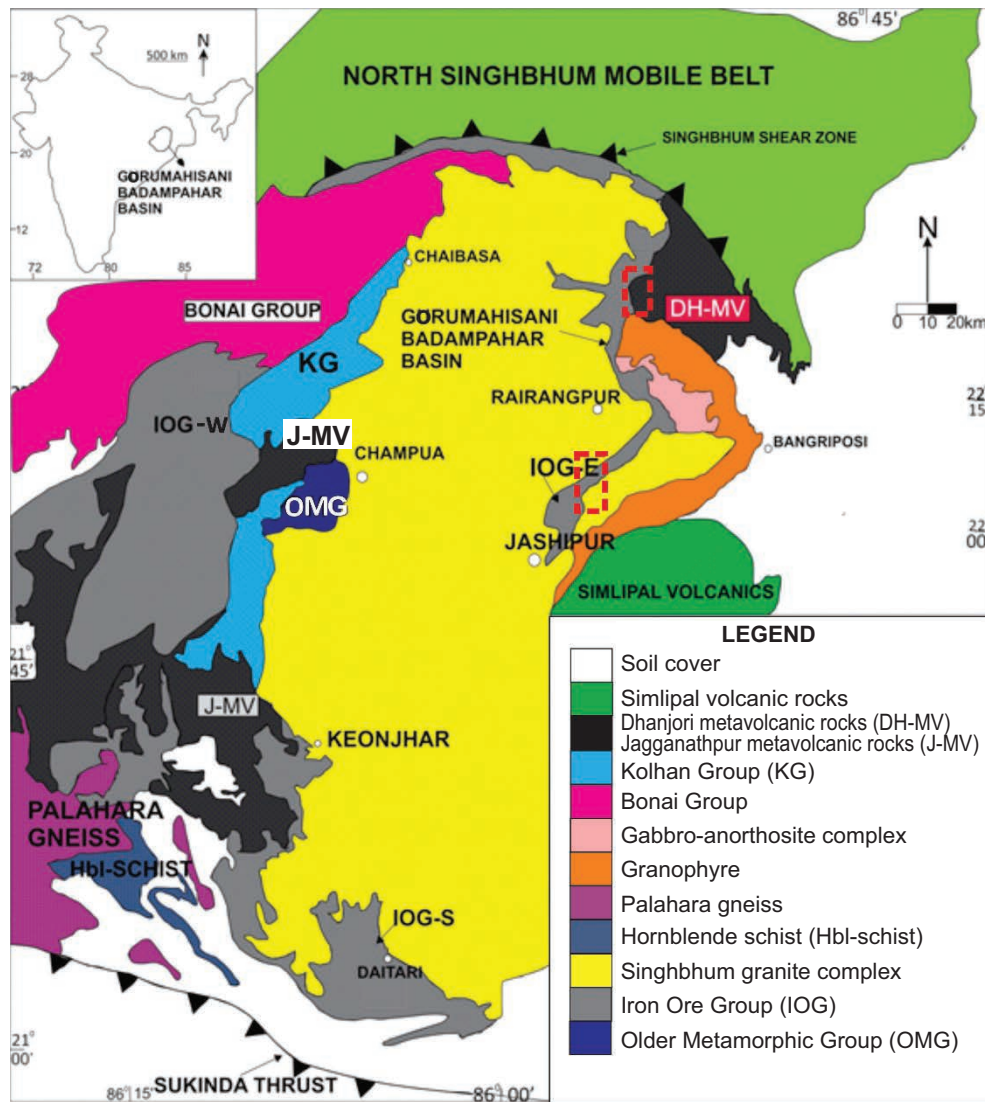


Fig. 1. Regional geologic map of the Singhbhum-Orissa craton, eastern India. Modified after Saha (1994).

composition. Based on its higher metamorphic grade, intense deformation, and intrusive relationship with the tonalitic gneisses, the Older Metamorphic Group is considered to be the oldest stratigraphic unit (Mukhopadhyay et al., 2015). On the basis of available data, it is inferred that majority of the Older Metamorphic Group and Older Metamorphic Tonalite Group was emplaced at around 3.4 to 3.3 Ga. The basement of the Older Metamorphic Group remains unidentified so far, but detrital zircons suggest a sialic crust. Mukhopadhyay (2001) suggested that the basaltic parent rocks of the Older Metamorphic Group orthoamphibolites were derived from a depleted mantle source. The Older Metamorphic Tonalite Group, on the other hand, is probably the result of partial melting of the Older Metamorphic Group amphibolites and older crustal sources. The Singhbhum granite was emplaced in three successive phases during 3.44, 3.32, and 3.31 Ga. Xenoliths of older tonalitic gneisses and mafic-ultramafic rocks have been observed within the Singhbhum granite. Apart from age and field relationship, the Singhbhum granite phases are also differentiated on the basis of their geochemical characteristics.

SBG-I rocks are K-poor granodiorite-trondhjemites, whereas SBG-II and SBG-III rocks grade from granodiorite to adamellite granites. The SBG-I and II rocks have a gently sloping rare earth element (REE) pattern with moderately enriched light REEs (LREEs) without Europium (Eu) anomaly similar to those of the Older Metamorphic Tonalite Group. SBG-III rocks show a relative enrichment of LREEs and a flat heavy REE (HREE) pattern with negative Eu anomaly (Saha and Ray, 1984; Mukhopadhyay, 2001).

Archean supracrustal sequences, i.e., Iron Ore Groups, occupy the western, eastern, and southern flanks of the Singhbhum craton. The three Iron Ore Groups (Fig. 1) are named the Noamundi-Jamda-Koira basin (western Iron Ore Group), Gorumahisani-Badampahar basin (eastern Iron Ore Group—study area), and the Tomka-Daiteri basin (southern Iron Ore Group). These successions consist of banded iron formations (BIFs), quartz-pebble conglomerates, quartzites, phyllites, tuffaceous shales, local dolomite, and volcanics (Saha, 1994). All of the three iron ore successions begin with a conglomerate-quartzite base, followed upward by shale, tuffs, lava, and BIFs.

In all of these belts, there is a close association of the BIFs and mafic lavas, which show pillow structures and tuffs. The felsic volcanic rocks from the western Iron Ore Group yielded a U-Pb (LA-ICP-MS) age of 3.29 Ga (Basu et al., 2008). Dacitic lava from the southern part of the craton yielded a U-Pb zircon age of 3506 ± 2.3 Ma (Mukhopadhyay et al., 2008). The eastern Iron Ore Group has been designated a minimum age of 3.1 Ga based on intrusive relationships within granitoids, as direct age data is unavailable (Saha, 1994; Mukhopadhyay et al., 2015). The Dhanjori and Simplipal volcano-sedimentary basins overlie the Iron Ore Group rocks. Both these basins have quartz-pebble conglomerates and quartzite at their base followed by layers of mafic rocks, tuffs, and volcanic breccias. The Simplipal basin forms an oval-shaped outcrop in the southeastern part of the craton. The Dhanjori basin lies

unconformably over the Singhbhum granite complex and marks the eastern flank of the Singhbhum craton. The other older supracrustals occur on the northern margin of the craton and bear impressions of intense deformation. The age of both of these basins is around 2.8 Ga (Roy and Bhattacharya, 2012). The youngest stratigraphic unit within the Singhbhum craton is the Kolhan Group, which is believed to be a carbonate platform succession formed in shelf to deep-water conditions (Mukhopadhyay et al., 2015).

Geologic Setting of the Study Area

The quartz-pebble conglomerates of the study area belong to the Gorumahisani-Badampahar schist belt (eastern Iron Ore Group) and the basal formation of the Dhanjori volcano-sedimentary basin within the Singhbhum craton (Fig. 2).

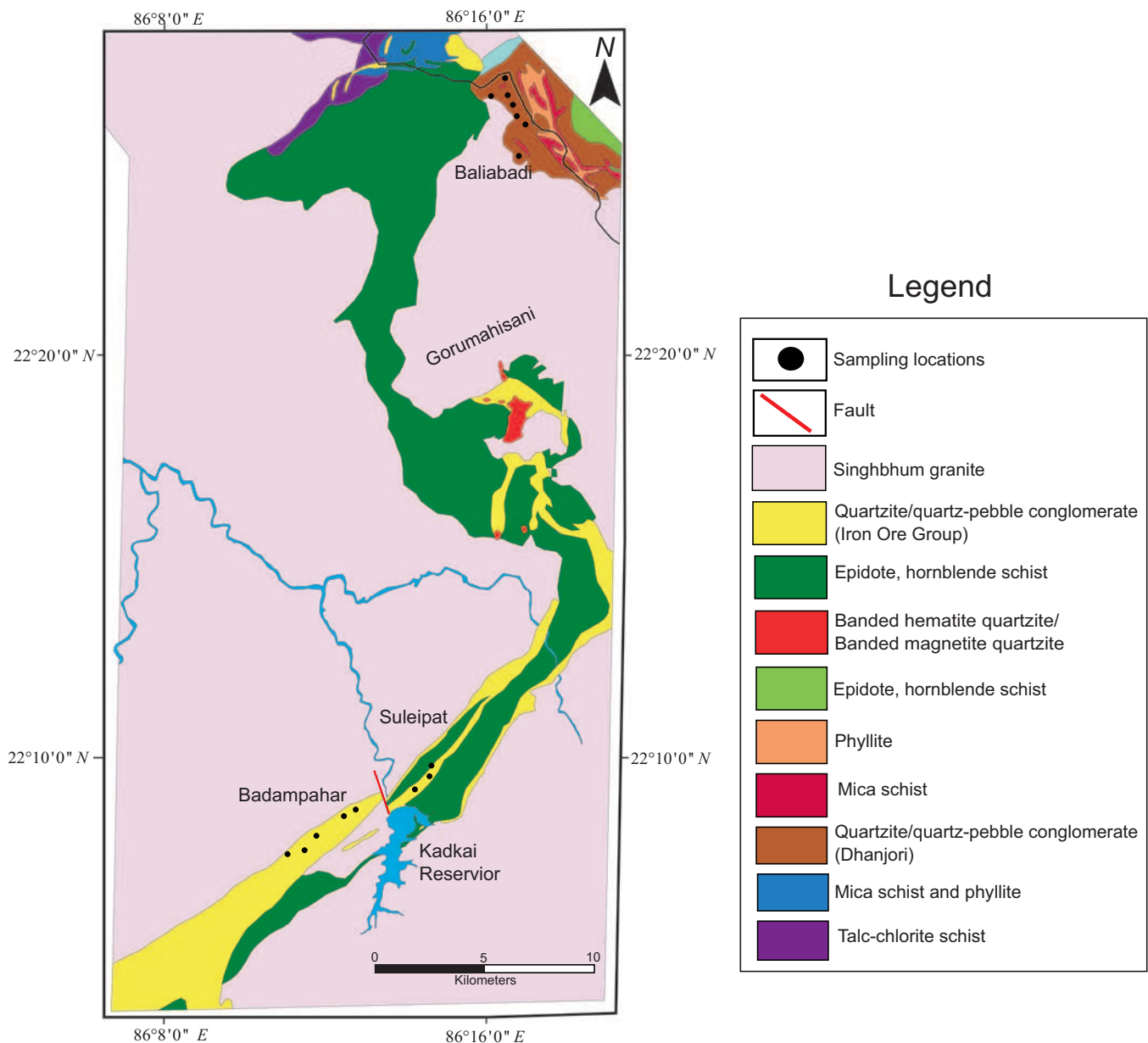


Fig. 2. Geologic map of study area indicating sampling locations. Modified after Geological Survey of India (unpub. data, 2006, 2007).

Gorumahisani-Badampahar schist belt

The eastern Iron Ore Group/Gorumahisani Badampahar schist belt trends north-northeast to south-southwest and turns to the northwest, forking into two arms (Fig. 2). It consists of a basal cherty quartz arenite, conglomerate, and quartzite overlain by one or two distinct BIF horizons. The thickness of the conglomerate quartzite package varies from 5 to 85 m. The associated metavolcanic rocks are locally pillowed and amygdular. The basalts are olivine tholeiites, and the composition varies from picrite to basaltic andesite (Mahadevan, 2002). Ultramafic rocks such as komatiites have also been reported from the belt. The entire belt is a long, thin band surrounded by the Singhbhum granite complex. In the southern part, between Badampahar and Suleipat, bedding is steep and subvertical. Within the eastern Iron Ore Group, conglomerates from the Suleipat dam section have been examined. A small NS-trending fault near Suleipat is observed, which has affected the BIF band.

Dhanjori volcano-sedimentary basin

The Dhanjori Formation is an EW-trending, northerly dipping linear belt, flanked by the Singhbhum shear zone at its northern and eastern periphery. The eastern Iron Ore Group rocks mark the northwestern boundary of the Dhanjori Formation (Ghosh and Sengupta, 1987; Mazumder and Sarkar, 2004). The Dhanjori basin unconformably overlies the Singhbhum granite and underlies the mica-schist quartzite assemblages

of the Chaibasa Formation (Mazumder and Sarkar, 2004). There is a clear predominance of siliciclastic assemblages within the Dhanjori Formation, but the presence of mafic and minor felsic rocks along with volcanoclastic rocks has also been reported from the basin (De et al., 2015). The Dhanjori basin has been classified into the lower and upper Dhanjori Formation (Fig. 3). The lower Dhanjori Formation unconformably overlies the Singhbhum granitoid and is further subdivided into two members. The lower member is primarily composed of phyllites, quartzite, and conglomerates, whereas the upper member is dominated by volcanic rocks, volcanoclastic rocks, phyllites, and quartzites. The upper Dhanjori Formation primarily consists of mafic volcanic rocks. The age of the Dhanjori basin is poorly constrained, and the basin is believed to be deposited at around 2.6 Ga (Acharyya et al., 2010a, b; De et al., 2015), hence assigning it a Paleoproterozoic status.

Characterization of Conglomerates

Quartz-pebble conglomerates were sampled from the peripheral margin of the Dhanjori basin, overlying the Singhbhum granitoid. The 2.3-km-long conglomerate horizon and lenses in Suleipat within the Gorumahisani-Badampahar belt were examined for their mineralogy and Au-U values.

The conglomerate exposure at Suleipat (eastern Iron Ore Group) rests on volcanic rocks and is intricately associated with compositionally banded quartzites. The conglomerates are mostly clast supported, are oligomictic in nature, and have

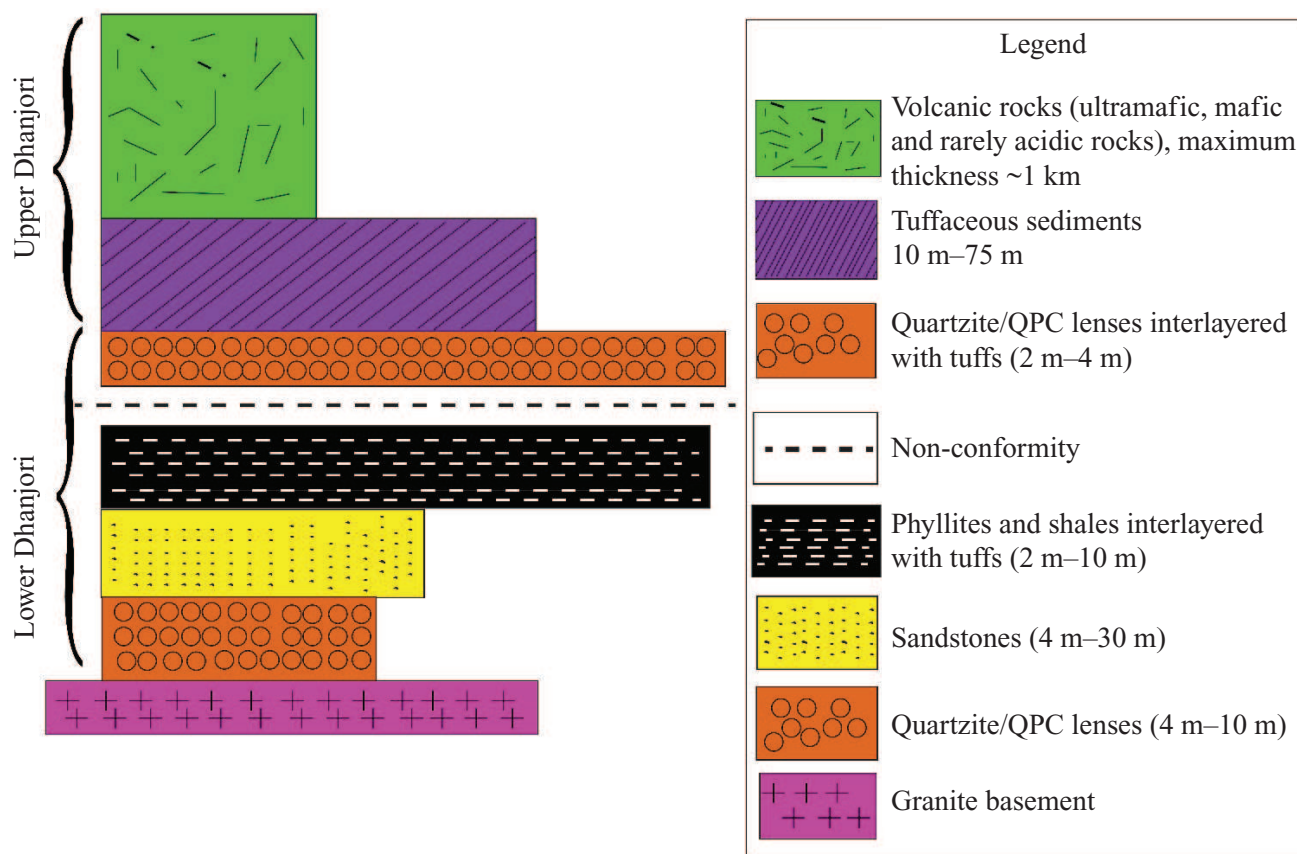


Fig. 3. Generalized stratigraphic succession of the Dhanjori volcano-sedimentary basin (after Mazumder and Sarkar, 2004). QPC = quartz-pebble conglomerate.

a variable thickness of 5 to 85 m. Clasts are made up of vein quartz, white quartz, smoky quartz, quartzite, and chert. The smoky nature of the quartz pebbles is partially a consequence of their exposure to radioactive minerals, and many workers are of the opinion that these pebbles were irradiated after deposition (Robinson and Spooner, 1982). The grain size varies from pebbles to boulders, and there is a complete lack of sorting (Fig. 4C). The clast size of the conglomerates at Suleipat varies according to their composition, viz. quartzite clasts (2–8 cm), chert clasts (0.5–6 cm), vein quartz (>1 cm), and white quartz pebbles (0.5–2 cm).

The clasts are rounded to subrounded and do not exhibit any preferred orientation. The lack of sorting, absence of sedimentary structures, and lack of preferred clast orientation suggest a debris flow mode of deposition for the Suleipat conglomerate. Numerous chert and quartz veins have intruded the conglomerate horizon, and the intrusion is believed to be facilitated by the NS-trending dextral fault observed at Suleipat (Fig. 4D). The matrix is predominantly made up of

recrystallized silica, and an overall silicification of the conglomerate body is observed. The silicification is thought to be associated with the widespread occurrences of quartz and chert veins.

The conglomerates examined from the lower Dhanjori Formation are different from the conglomerate exposures at Suleipat. The Dhanjori conglomerates are intercalated with quartzite units and unconformably overlie the Singhbhum granitoid. They are overlain by nonuniform outcrops of mafic volcanic rocks. The conglomerate outcrops are nonuniform in nature and occur as localized lenses ranging in thickness between 5 and 10 m. The clasts are mostly made up of vein quartz, smoky quartz, and quartzite. Quartzite clasts are of larger dimensions than the clasts of different composition. The size of quartzite clasts varies between 1 and 4 cm and that of the smoky and white quartz pebbles varies from 0.4 to 2 cm. The clast morphology varies from rounded to subrounded with well-abraded edges and bedding-parallel alignment (Fig. 4A). The majority of these conglomerates are clast supported



Fig. 4. A: Field photograph of smoky and white quartz pebbles within quartz-pebble conglomerates showing weak preferred orientation. B: Field photograph of Bhatgora quartz-pebble conglomerates with fuchsite matrix and overturned quartzite pebbles. C: Field photograph showing unsorted nature of clasts from quartz-pebble conglomerates at Suleipat. D: Field photograph of chert veins cutting across the Suleipat quartz-pebble conglomerates close to the NW-SE-trending fault.

and show moderate sorting. The presence of current bedding and trough crossbeds along with a fining-upward sequence is observed in a few outcrops. The matrix is predominantly of arenaceous nature and exhibits lower greenschist facies metamorphism. The bedding-parallel clast orientation, moderate sorting, and sheet-like geometry indicate a sheet-flow mechanism for the formation of these conglomerates (Blair, 1999a, b; Mazumder and Sarkar, 2004). At Bhatgora, the fuchsite matrix shows evidence of mild alteration and deformation (Fig. 4B). The schistosity is presumably governed by the mineralogy of the matrix and not by the directed pressure. The clast mineralogy is dominated by quartzite with minor quartz vein occurrences. The Bhatgora conglomerate is intercalated with lenses of fuchsite quartzite, which is reflected in the high Cr values. The poor sorting, lack of sedimentary features, and chaotic clast orientation suggests a debris flow mode of origin

for the Bhatgora conglomerate (Dasgupta, 2003; Mazumder and Sarkar, 2004).

Mineralogical Characterization of Quartz-Pebble Conglomerates

The conglomerates are characterized by the presence of detrital heavy minerals and a weakly foliated matrix. The clasts are entirely made up of rounded to elongated quartz pebbles and minor chert occurrences. Apart from recrystallized quartz, the framework grains are composed of ferruginized biotite, sericite (altered plagioclase feldspar), chlorite, and fuchsite. The diagenetic modifications are manifested in the form of pressure-solved grain boundaries in quartz pebbles and cracked detrital grains. The detrital minerals include rutile, chromite, zircon, sphene, magnetite, monazite, and pyrite (Fig. 5). Detrital gold occurrences have also been reported

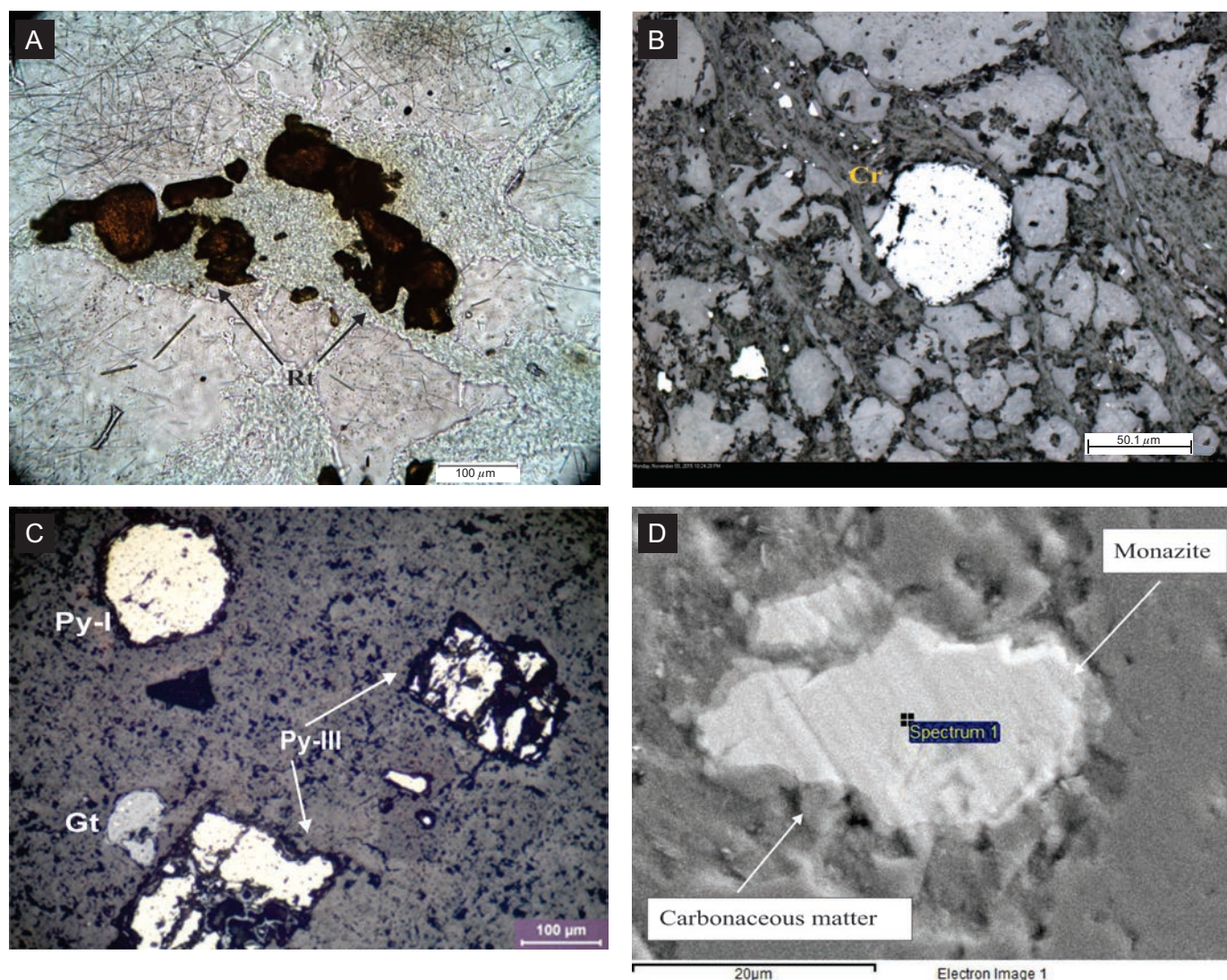


Fig. 5. A: Photomicrograph showing detrital rutile (Rt) within fuchsite matrix. B: Photomicrograph of detrital chromite (Cr) within quartz-pebble conglomerate matrix. Note mild deformation due to postdepositional burial diagenesis. C: Py-I and Py-III within quartz-pebble conglomerate. Note rounded nature of Py-I. Goethite (Gt) seen in the photomicrograph is a consequence of late oxidative alteration of pyrite. D: Backscattered electron image of monazite grain within possible carbonaceous matter from quartz-pebble conglomerates.

from these conglomerates by previous workers (Bose, 1904; Sarangi and Mohanty, 1987; Iyenger, 1995). Detritus was presumably derived from the Singhbhum granite complex and mafic-ultramafic enclaves therein. The other minerals include hematite, goethite, chalcopyrite, pyrrhotite, stibnite, brannerite, and other secondary uranyl minerals. Pyrite and chalcopyrite exhibit replacement textures along with pyrrhotite.

Occurrences of gold nuggets have been previously reported in the vicinity of Ruansi and Gohaldungri within the study area in the Gorumahisani-Badampahar belt along the Godia stream. Nuggets weighing between 23.25 and 31.1 g were obtained from a gravel bed in the neighborhood of Gohaldungri. (Geological Survey of India, 2012). Uraninite occurs as fine grains and veinlets in the sulfide-rich matrix within the conglomerates of the Gorumahisani-Badampahar belt (Das et al., 1988). Uraninite occurs as rounded to subrounded and muffin-shaped detrital grains (0.06–0.2 mm) within the quartz-pebble conglomerates of the Dhanjori basin. Stringers of pitchblende and secondary uranium minerals occur as fracture fillings (Rao et al., 1988). The majority of the uraninite grains are oxidized and altered due to the susceptibility of this phase to oxic environments (Kotzer and Kyser, 1995; Bergen and Fayek, 2012). The mineral phases such as chromite, zircon, rutile, monazite, pyrite, arsenopyrite, stibnite, and brannerite were confirmed by EPMA (Table 1) and SEM studies (Figs. 6–9).

Analytical Techniques

LA-ICP-MS analyses of various sulfides were carried out for trace element analysis at Department of Geology and

Geophysics, Indian Institute of Technology, Kharagpur, India, with a New Wave ESI 193-nm solid state laser ablation system coupled to a TermoFisher Scientific iCAP quadrupole ICP-MS. Spot analysis was conducted with 5-Hz laser repetition, 4.5-J/cm² energy, and He used as carrier gas. Analyses were performed in peak-hopping mode, with each analysis consisting of a 30-s background measurement with the laser turned off and 40-s peak signal measurement with the laser turned on. Dwell time for each mass was set to 25 ms. Data were collected in the time-resolved mode. Spot size ranges from 25 to 40 µm. NIST 610 and NIST 612 were used as reference material. The data were reduced offline with the help of Glitter© software (Griffin et al., 2008). Measured isotopes are ⁵²Cr, ⁵⁵Mn, ⁵⁹Co, ⁶⁰Ni, ⁶³Cu, ⁶⁶Zn, ⁷⁵As, ⁹⁵Mo, ¹⁰⁷Ag, ¹¹⁸Sn, ¹²¹Sb, ¹²⁵Te, ¹⁸²W, ¹⁹⁷Au, ²⁰⁸Pb, ²³²Th, and ²³⁸U.

Quartz-pebble conglomerate samples were studied to infer their textural features and paragenetic relationships. Compositional zoning and microtextures of pyrites were studied by EPMA. The analysis was carried out using CAMECA SXFive instrument at DST-SERB National Facility, Department of Geology, Centre of Advanced Study, Institute of Science, Banaras Hindu University, India. Polished thin sections were coated with a 20-nm-thin layer of carbon for EPMA using a LEICA-EM ACE200 instrument. The CAMECA SXFive instrument was operated by SXFive software at a voltage of 15 kV and current of 10 nA with a LaB₆ source in the electron gun for generation of the electron beam. The natural silicate mineral andradite was used as an internal standard to verify positions of crystals (SP1-TAP, SP2-LiF, SP3-LPET,

Table 1. Electron Probe Microanalysis Data of Various Mineral Phases from Gorumahisani-Badampahar and Dhanjori Basin Quartz-Pebble Conglomerates (in wt %)

Mineral phase	S	Fe	As	Ni	Co	Sb	U	Th	Ti	Pb
As-bearing pyrite	53.10	45.80	1.03	0.33	-	-	-	-	-	-
As-bearing pyrite	52.39	45.90	0.60	-	-	-	-	-	-	-
As-bearing pyrite	52.87	46.16	0.42	-	-	-	-	-	-	-
As-bearing pyrite	52.90	46.46	0.73	-	-	-	-	-	-	-
As-bearing pyrite	53.08	45.52	0.50	-	-	-	-	-	-	-
As-bearing pyrite	51.89	45.93	0.51	-	-	-	-	-	-	-
As-bearing pyrite	54.36	45.02	0.79	-	-	-	-	-	-	-
As-bearing pyrite	53.27	46.22	0.66	-	-	-	-	-	-	-
As-bearing pyrite	52.26	45.99	0.97	-	-	-	-	-	-	-
As-bearing pyrite	53.19	45.70	0.74	-	-	-	-	-	-	-
As-bearing pyrite	52.63	45.87	0.58	-	-	-	-	-	-	-
As-bearing pyrite	52.86	45.93	0.86	-	-	-	-	-	-	-
As-bearing pyrite	50.80	46.30	1.41	-	-	-	-	-	-	-
As-bearing pyrite	53.18	44.84	1.80	-	-	-	-	-	-	-
As-bearing pyrite	53.31	44.79	0.72	-	-	-	-	-	-	-
As-bearing pyrite	52.82	44.63	0.72	-	-	-	-	-	-	-
As-bearing pyrite	53.09	45.57	0.86	-	-	-	-	-	-	-
As-bearing pyrite	52.77	45.92	0.87	-	-	-	-	-	-	-
As-bearing pyrite	52.34	44.54	3.00	1.52	0.40	-	-	-	-	-
Arsenopyrite	20.58	35.65	45.02	-	-	-	-	-	-	-
Arsenopyrite	20.51	33.10	44.96	1.00	-	-	-	-	-	-
Arsenopyrite	20.77	34.70	45.42	-	-	-	-	-	-	-
Arsenopyrite	21.16	36.02	43.57	-	-	-	-	-	-	-
Arsenopyrite	21.57	31.60	39.80	1.71	2.30	-	-	-	-	-
Arsenopyrite	21.77	33.09	42.43	0.56	0.35	-	-	-	-	-
Stibnite	29.37	-	0.75	-	0.31	72.25	-	-	-	-
Stibnite	26.97	0.11	0.49	-	-	68.01	-	-	-	-
Stibnite	5.92	0.11	0.48	-	-	67.89	-	-	-	-
Brannerite	-	4.98	-	-	-	-	39.58	0.24	18.19	11.57

- = below detection limit

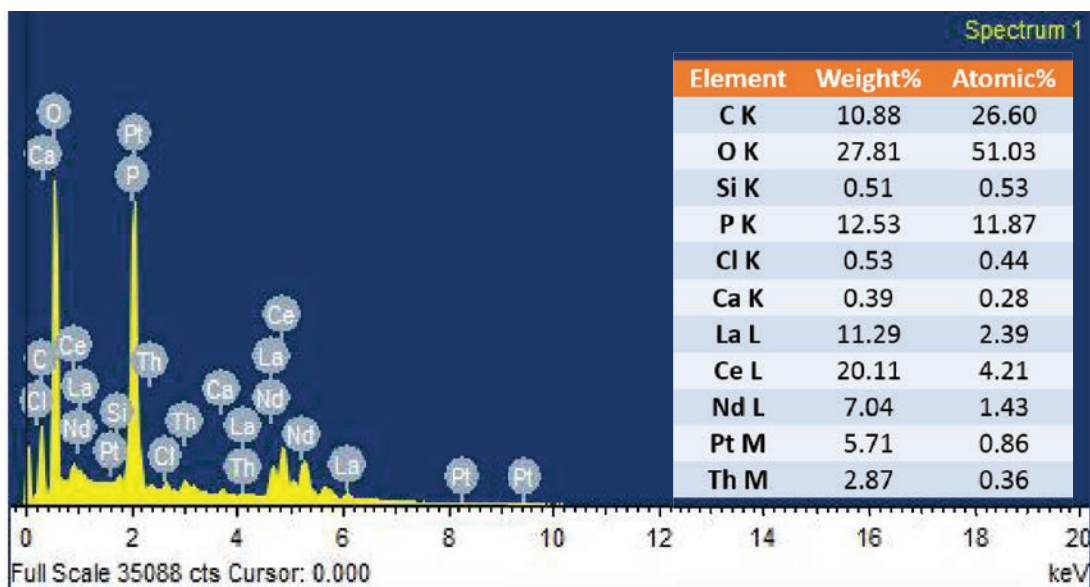


Fig. 6. Scanning electron microscopy energy dispersive spectrometry data of monazite within carbonaceous matter.

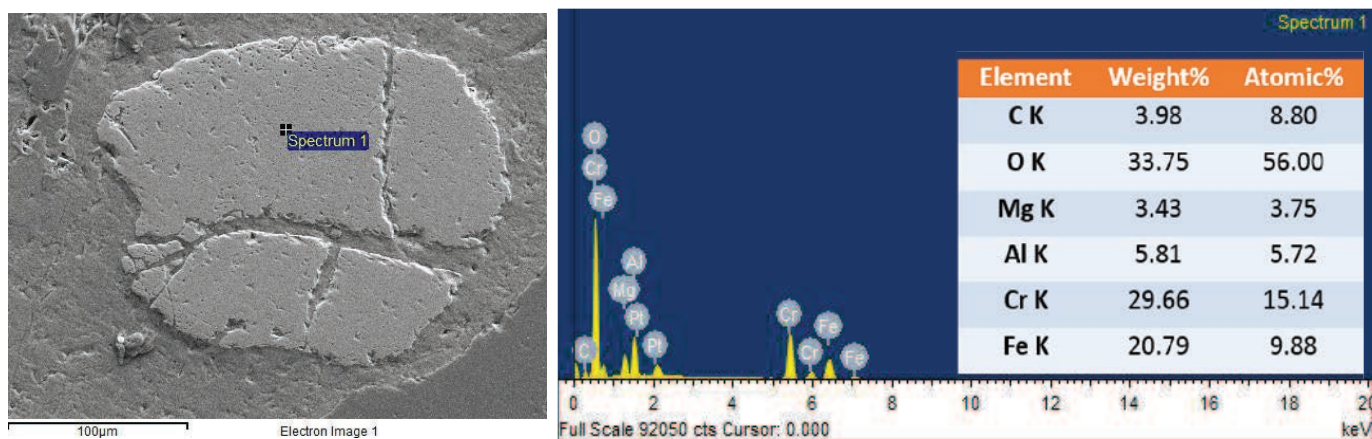


Fig. 7. Backscattered electron image and scanning electron microscopy-energy dispersive spectrometry data of detrital chromite in quartz-pebble conglomerate matrix.

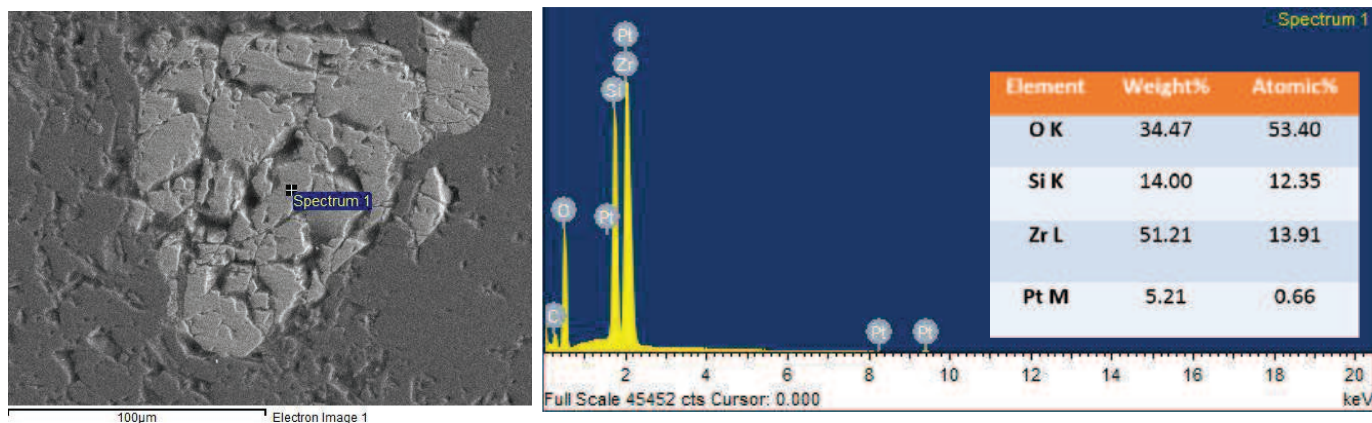


Fig. 8. Backscattered electron image and scanning electron microscopy-energy dispersive spectrometry data of detrital zircon in quartz-pebble conglomerate matrix.

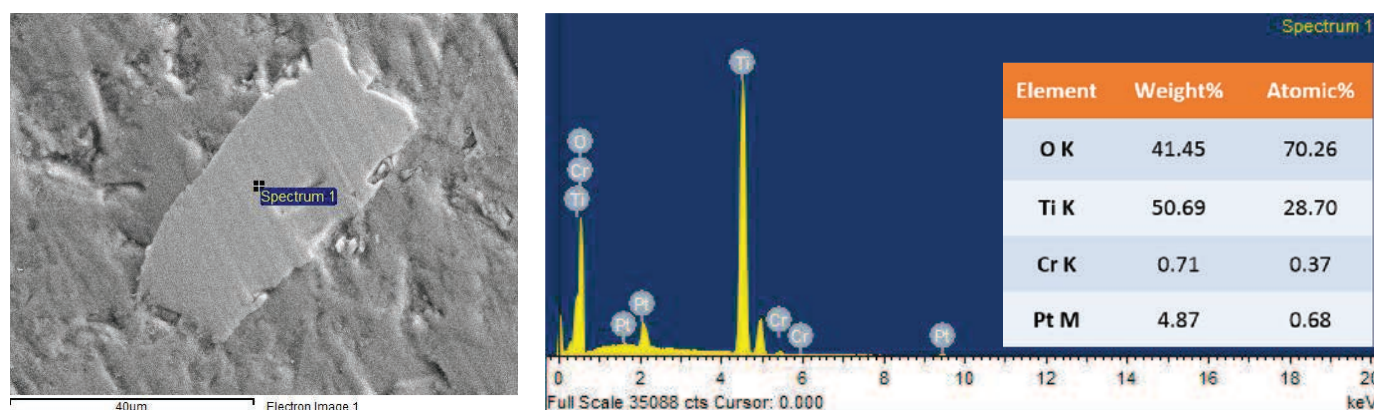


Fig. 9. Backscattered electron image and scanning electron microscopy-energy dispersive spectrometry data of detrital rutile within quartz-pebble conglomerate matrix.

and SP4-TAP) with respect to corresponding wavelength dispersive (WD) spectrometers in the CAMECA SXFive instrument. The following X-ray lines were used in the analyses: S- $K\alpha$, Ti- $K\alpha$, Fe- $K\alpha$, Zn- $K\alpha$, Cu- $K\alpha$, Co- $K\alpha$, Ni- $K\alpha$, As- $L\alpha$, Ag- $L\alpha$, Sb- $L\alpha$, Pb- $M\alpha$, Au- $M\alpha$, Hg- $M\alpha$, and Bi- $M\alpha$. Natural mineral standards pyrite, rutile, hematite, galena and cinnabar and synthetic indium arsenide (InAs), indium antimonide (InSb), Ni, Co, Cu, Zn, Au, bialloy, and metal supplied by CAMECA-AMETEK were used for routine calibration and quantification. Routine calibration, acquisition, quantification, and data processing were carried out using SxSAB version 6.1 and SX Results CAMECA software. The precision of the analysis is better than 1% for major element oxides and 5% for trace elements from the repeated analyses of standards.

Pyrite Textures and Morphology

Based on petrography, microprobe data, and backscattered electron images, six major textural varieties of pyrite have been identified in the studied quartz-pebble conglomerate samples. Rounded detrital, microcrystalline, small euhedral, small anhedral, large anhedral, and large euhedral pyrite varieties are the major textural varieties observed. For simplicity, we group the detrital pyrites as Py-I; the small anhedral, small euhedral, and microcrystalline pyrites as Py-II; and the large anhedral and large euhedral pyrites as Py-III. The detrital pyrites (Py-I) were the first to be incorporated within the conglomerates and are disposed along well-defined bedding planes (Fig. 10A). They are characterized by hydrothermal overgrowths around the detrital core. The microcrystalline, small anhedral, and small euhedral grains (Py-II) (Fig. 10B, C) are interpreted to be of diagenetic origin (Large et al., 2007). The large euhedral and anhedral crystals (Py-III) are of hydrothermal origin (Fig. 10D-F) seen as overgrowths around the microcrystalline and diagenetic pyrites. According to Gregory et al. (2015), such coarser textures might be a consequence of migration and recrystallization of Fe and S from preexisting smaller pyrite crystals. Based on experimental evidence, it has been determined that low-temperature pyrites (<150°C) that formed rapidly are fine grained and microcrystalline in shape. However, pyrites that precipitate slowly from higher-temperature (>200°C) hydrothermal or metamorphic fluids tend to be coarse grained (Butler and Rickard, 2000).

Distribution of Trace Elements in Pyrite

Economic geologists have long recognized that pyrite contains a wide variety of trace elements (Abraitis et al., 2004). As far as ore deposits are concerned, As is the most significant trace element, as it facilitates the substitution of other trace elements within the pyrite lattice, and As-bearing pyrite is a well-known host for Au in a wide range of Au deposits (Deditius et al., 2014; Sahoo and Venkatesh, 2015). Co and Ni are also significant, as the Co/Ni ratios have been used to distinguish between hydrothermal and diagenetic or syngenetic pyrite (Gregory et al., 2015). Co/Ni ratios >2 suggest precipitation of the pyrite crystals from hydrothermal fluids. In addition, the Co and Ni values have implications for the depositional setting of pyrite formation. Pyrites formed in organic-matter-rich environments generally exhibit high Ni values and low Co/Ni ratios (Dill and Kemper, 1990; Guy et al., 2010). It has also been observed that higher reducing conditions increase the uptake of Ni into pyrite (Maslennikov et al., 2009; Scott et al., 2009).

LA-ICP-MS spot analyses on selected pyrite grains were carried out to understand their trace element systematics and confirm the presence of Au within the system (Table 2). Arsenic values vary from 14,186 to 68,481 ppm, Co values vary from 285 to 4,517 ppm, and Ni values vary between 223 and 2,925 ppm. Nickel is consistently enriched in the diagenetic pyrites (Co/Ni ratios <2), and there is an overall enrichment of trace elements, especially As, in the diagenetic pyrites compared to the hydrothermal pyrites (Fig. 11). In the case of hydrothermal pyrites, the higher temperature and slower growth rate facilitate partitioning of trace elements into separate sulfide phases rather than uptake only within pyrite as solid solution or microinclusions, hence an overall depletion is observed (Large et al., 2009). The recrystallization of preexisting pyrite by metamorphic-hydrothermal fluids may also result in the loss of trace elements due to dissolution of the pyrite crystal and subsequent release of the trace elements. However, hydrothermal pyrites exhibit enriched trace element values in certain cases, where trace elements are released by either organic matter or Fe oxides and incorporated within the pyrite crystal (Gregory et al., 2015). X-ray compositional maps obtained from the microprobe show that

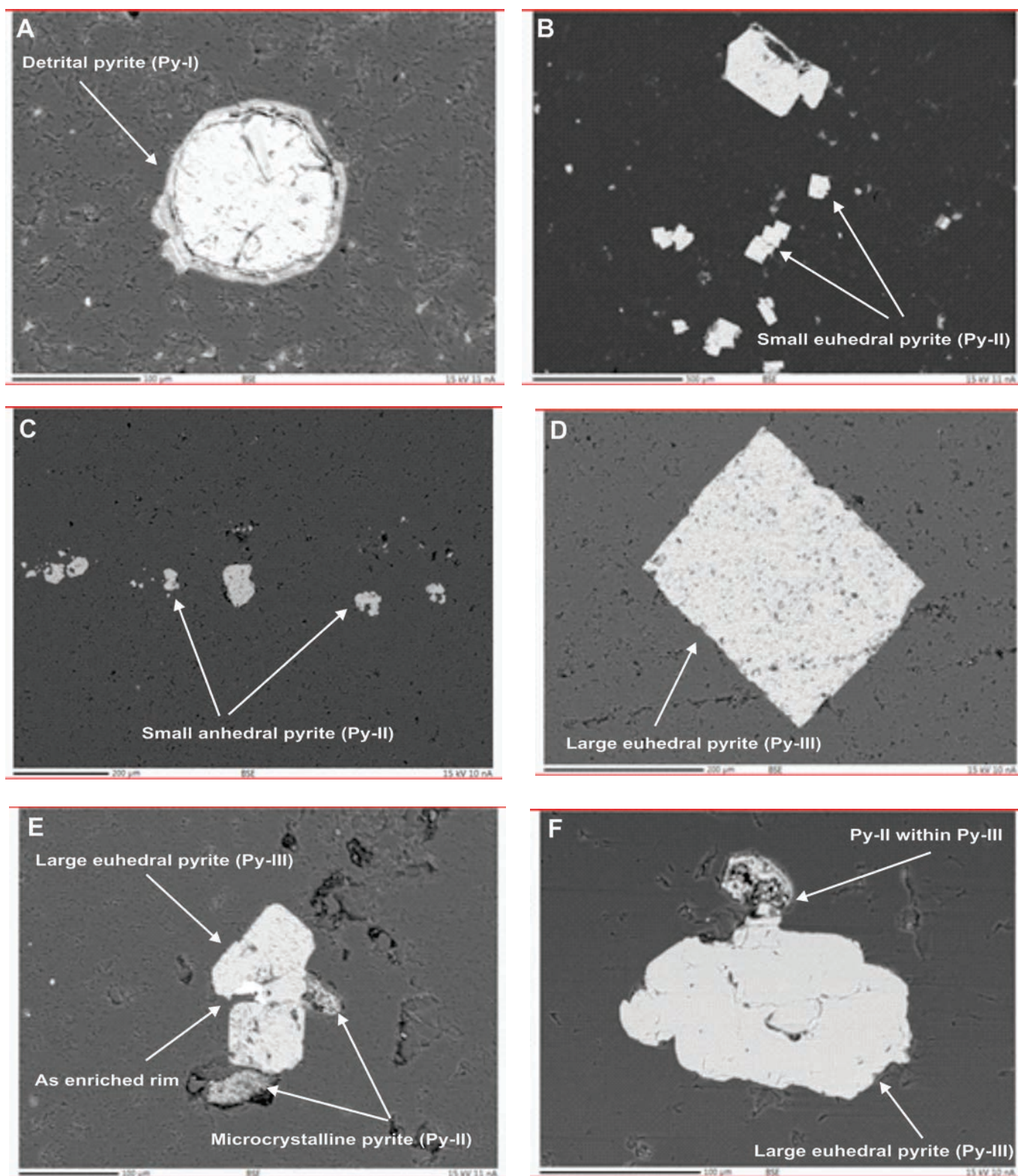


Fig. 10. A: Detrital pyrite (Py-I) with hydrothermal overgrowth around detrital core. B: Small euhedral pyrite (Py-II) of diagenetic origin within quartz-pebble conglomerates. C: Diagenetic small anhedral pyrite (Py-II) within quartz-pebble conglomerates. D: Large euhedral pyrite of hydrothermal origin (Py-III). E: Large anhedral pyrite (Py-III) of hydrothermal origin. Note the presence of relict microcrystalline pyrite (Py-II). Also As-rich rim is observed, indicating As enrichment around the edges of Py-III. F: Large euhedral pyrite (Py-III) precipitating over microcrystalline (diagenetic) pyrite (Py-II).

Table 2. Laser Ablation-Inductively Coupled Plasma-Mass Spectrometry Trace Element Values from Different Generations of Pyrite (in ppm)

Mineral phase	Mo	Cu	Co	Ni	Ag	Zn	As	Au	Sb	Bi	Pb
Py-II	0.099	23.79	953.9	1,271.03	0.31	4.21	34,011.7	6.73	42.65	4.62	16.62
Py-II	111.77	256.98	285.36	2,177.84	1.43	5.18	37,712.22	17.24	1,792.86	24.67	217.1
Py-II	1.49	186.96	474.85	2,925.57	0.91	5.63	68,481.43	51.87	960.31	12.51	185.2
Py-II	0.82	18.32	465.77	255.7	0.57	4.44	14,186.1	3.4	188.1	14.2	68.2
Py-II	<0.61	157.18	499.34	223.13	0.22	4.57	59,024.81	9.14	80.37	2.12	17.31
Py-III	<0.45	6,248.61	2,643.54	715.62	1.23	23.1	18,734.3	3.34	93.13	13.93	79.54
Py-III	5.29	20.09	4,517.53	540.82	1.64	4.31	30,165.87	8.71	425.71	22.4	96.02
Py-III	1.15	49.93	2,754.88	832.73	1.33	6.17	20,177.79	5.24	411.77	13.79	118.6
Py-III	0.99	30.27	1,645.47	471.19	0.3	4.81	22,048.19	3.7	161.29	15.12	135.2
Py-III	0.43	299.91	2,106.65	295.54	0.82	5.65	27,675.83	5.51	164.11	12.53	45.74

the hydrothermal (large euhedral and large anhedral) pyrites are characterized by cyclic growth zones of As (Fig. 12B) and As enrichment around the grain boundaries as evidenced from the As-rich rims (Fig. 12C).

Nature of Au Within Pyrite

Gold is commonly associated with sulfides in a wide range of ore systems, including sediment-hosted disseminated gold, Carlin-type (Arehart et al., 1993; Cline et al., 2005), orogenic (Mumin et al., 1994; Saha and Venkatesh, 2002), epithermal (Griffin et al., 1991), intrusion-related (Marsh et al., 2003), and Witwatersrand modified placer Au (Frimmel et al., 2005; Agangi et al., 2015). Submicroscopic gold or invisible gold can be hosted within arsenian pyrite and arsenopyrite in solid solution within their crystal lattices and/or as discrete minute particles (Cook and Chrysosoulis, 1990; Fleet and Mumin, 1997; Morey et al., 2008; Deditius et al., 2009; Jha et al., 2015). The maximum amount of Au that can be incorporated into pyrite is primarily a function of As concentration and is governed by the equation $C_{Au} = 0.02 \times C_{As} + 4 \times 10^{-5}$ (Reich et al., 2005). This equation is described by a line, known as the gold saturation line for pyrite. Below this line, Au occurs in the form of solid solution or lattice bound Au (invisible Au) within the pyrite crystal lattice. Above this line, gold occurs in the form of tiny blebs, Au nanoparticles, free Au, or Au tellurides within the pyrite.

LA-ICP-MS point analyses on the pyrite cores and rims from the quartz-pebble conglomerate samples reveal gold values between 3.7 and 51.8 ppm (Table 2). Gold shows a strong positive correlation with As in the diagenetic as well as in the hydrothermal pyrites (Fig. 13A). However, trace elements such as Ni, Mo, and Sb exhibit a moderate positive correlation with Au only in the diagenetic pyrites (Fig. 13B-D). On the Au vs. As plot, the pyrites cluster below the Au saturation line, which indicates the presence of Au in solid solution within the pyrite lattice (Fig. 13A). The Au saturation line also provides information about the saturation state of solutions that precipitated Au (Reich et al., 2005). All pyrites from our study area fall below the Au saturation line, which suggests that the pyrites precipitated from solutions that were undersaturated with respect to gold.

Our data suggest that the Au was transported and accumulated as fine detrital particles during diagenetic pyrite growth within the conglomerate horizon. The positive correlation of Au with redox-sensitive elements such as Ni and Mo in diagenetic pyrite (Fig. 13) further supports the idea that Au was incorporated within pyrite during diagenetic growth (Large et al., 2013). The fixation of gold onto diagenetic pyrite is believed to be microbially mediated or due to the presence of activated carbon from the remains of dead microbes (Horscroft et al., 2011). This theory is supported by the presence of carbonaceous matter adsorbed onto the

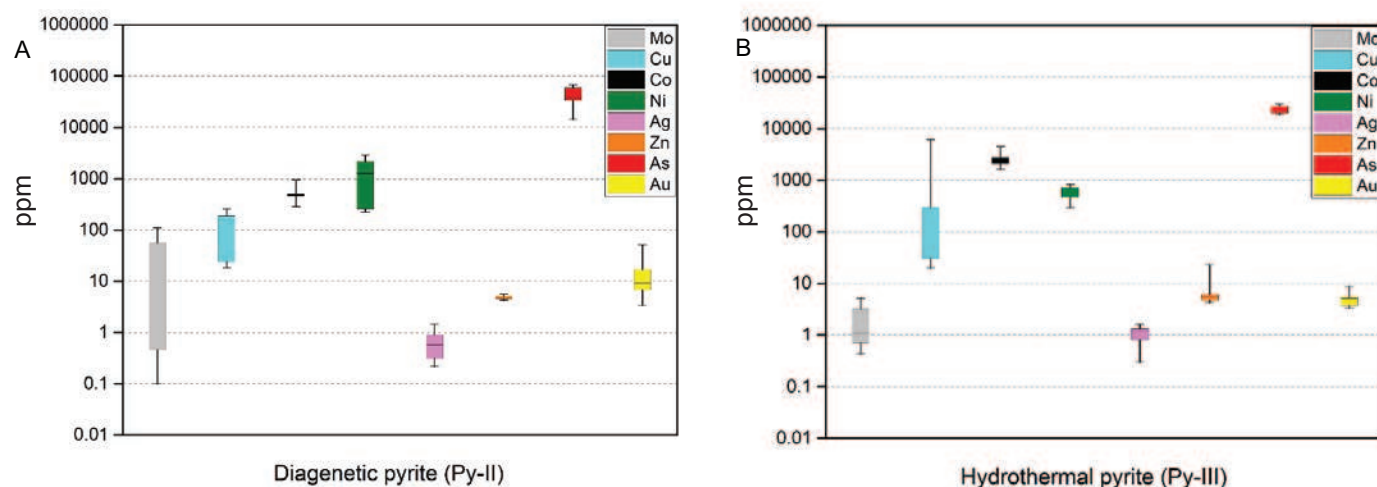


Fig. 11. A: Box plot of trace element distribution in diagenetic pyrite (Py-II). B: Box plot of trace element distribution in hydrothermal pyrite (Py-III).

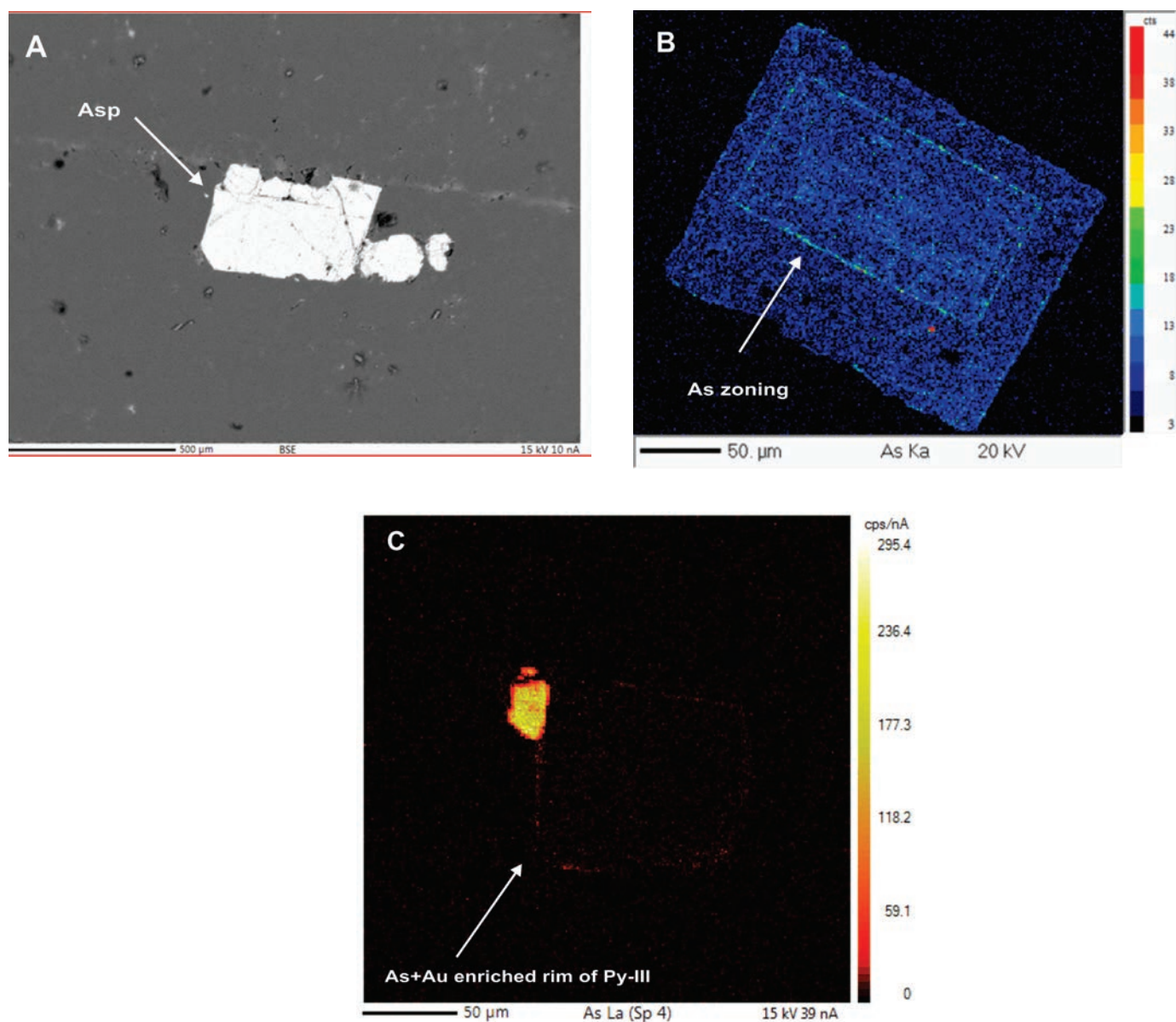


Fig. 12. A: Backscattered electron image of an arsenopyrite (Asp) grain along a vein. B: Electron probe microanalysis (EPMA) X-ray compositional map of Py-III (hydrothermal) showing cyclic growth zone of As. C: EPMA X-ray compositional map of Py-III showing As enrichment around the rim of the pyrite crystal, indicating As enrichment around the edges.

surface of diagenetic pyrite of the study area, as seen in Figure 14. A subsequent hydrothermal event precipitated the large euhedral and the anhedral pyrite types (Py-III) along with solid solution Au. These fluids remobilized Au along with other associated trace elements and precipitated them onto the younger generation pyrites (Py-III). This is evidenced by the depleted trace element values and cyclic growth zones in the younger pyrites. The infiltrating fluid was definitely rich in S but undersaturated with respect to gold, as evidenced from the Au vs. As plot (Fig. 13A). There are very few evidences of remobilization of the preexisting pyrites, hence the formation of the coarser hydrothermal pyrites and associated Au cannot be attributed completely to the remobilization and reprecipitation of the earlier pyrites. A hydrothermal input is further supported by the numerous crosscutting veins within

the conglomerate horizon. Hence, it is inferred that Au was incorporated in its detrital form within the conglomerates followed by a subsequent bout of hydrothermal infiltration, which remobilized some of the earlier Au and precipitated hydrothermal Au. The NS-trending fault facilitated the movement of S-rich fluids, which subsequently deposited hydrothermal pyrite, chalcopyrite, pyrrhotite, and invisible Au in favorable horizons such as unconformity surfaces, bedding planes, and fractures.

Discussion and Conclusions

In this work, quartz-pebble conglomerate samples have been collected from the Archean Gorumahisani-Badampahar schist belt and the Paleoproterozoic lower Dhanjori volcano-sedimentary succession. Petrography, whole-rock geochemical

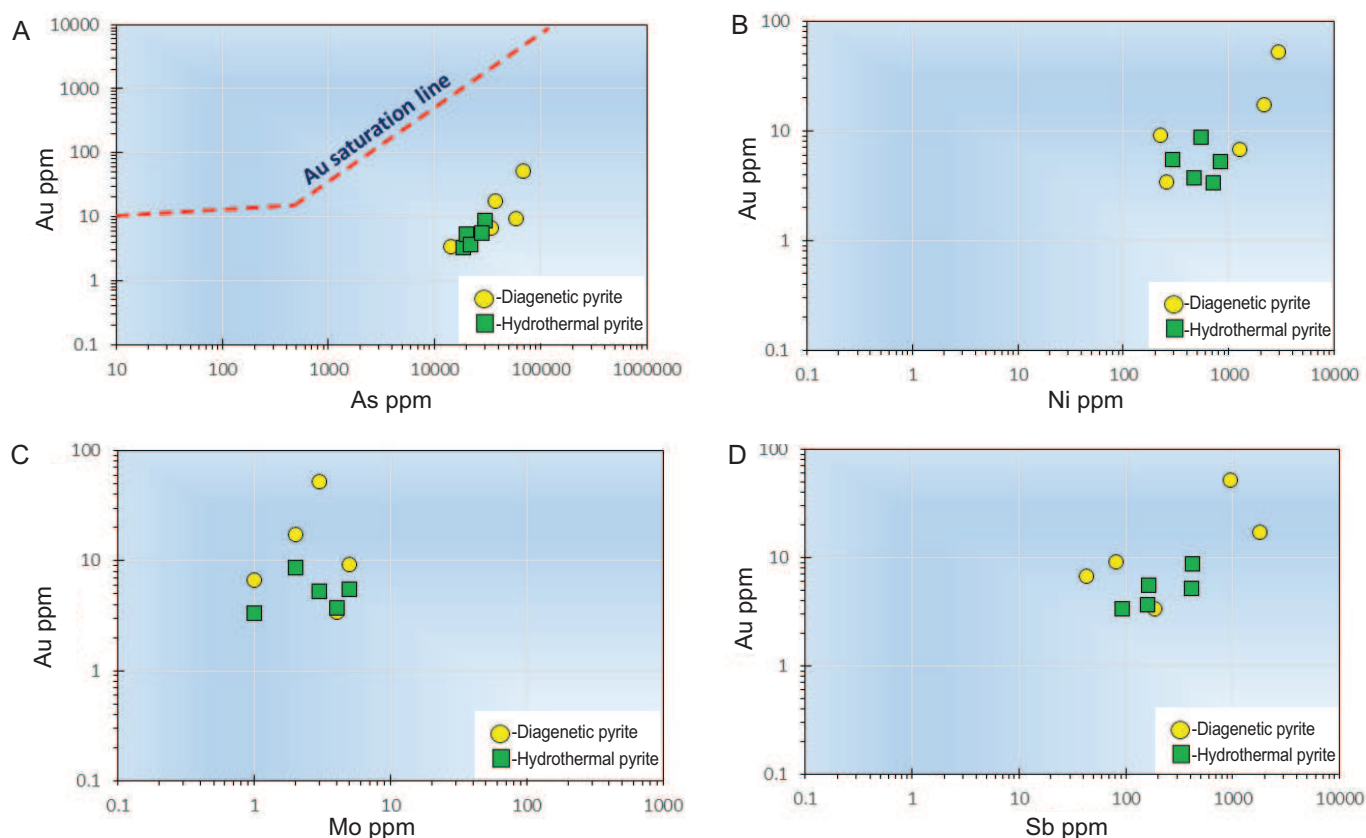


Fig. 13. A: Plot of Au vs. As. Note the positive correlation of Au and As. Note all pyrites fall below the Au saturation line (after Reich et al., 2005), suggesting presence of Au in solid solution. B: Plot of Au vs. Ni. Note the positive correlation of Au and Ni in diagenetic pyrite. C: Plot of Au vs. Mo. Note the positive correlation of Au and Mo in diagenetic pyrite. D: Plot of Au vs. Sb. Gold shows a moderate positive correlation with Sb in all pyrite types.

analyses (ICP-MS), EPMA, SEM, and LA-ICP-MS studies were carried out to understand the mineralogy, mineral paragenesis, trace element characteristics, ore genesis, and economic potential of the quartz-pebble conglomerate horizons. Significant values of gold were obtained from As-bearing pyrites using LA-ICP-MS. The presence of uranium is confirmed by whole-rock geochemical analysis using ICP-MS. Surficial samples show whole-rock uranium values varying from 0.45 to 22.9 ppm (Table 3). We also report the presence of brannerite, as confirmed by microprobe analysis. Brannerite generally forms due to the affinity of rutile to absorb or adsorb uranium from sediments, hence we infer that the brannerite within the studied quartz-pebble conglomerates is a pseudomorphic replacement of rutile (Rao and Rao, 1983; Udayakumar and Phillip, 1997; Bergen and Fayek, 2012).

The conglomerates studied in this work have some similarities when compared with the Witwatersrand conglomerates. The Archean age; disposition of the siliciclastic sequence over a granite-greenstone terrain; presence of detrital chromite, rutile, zircon, sphene, apatite, pyrite, and magnetite; presence of biotite, chlorite, and fuchsite in the conglomerate matrix; and the presence of secondary sulfides and gold are the major similarities observed. However, the differences of these conglomerates with respect to the Witwatersrand cannot be overlooked. Gold within the conglomerates of the study area is entirely hosted in the crystal lattices of arsenian

pyrite, and it shows a strong positive correlation with As, similar to the Witwatersrand conglomerates (Large et al., 2013). However, all the hydrothermal pyrites from the studied conglomerates fall below the gold saturation curve, unlike at Witwatersrand, where the hydrothermal pyrites plot above the gold saturation curve (Large et al., 2013). There are no strong evidences of metamorphism within the conglomerates of the study area, unlike the Witwatersrand conglomerates, which have undergone widespread regional metamorphism (Phillips

Table 3. Whole-Rock Geochemical Data (inductively coupled plasma-mass spectrometry) of Quartz-Pebble Conglomerates from Study Area Indicating U, Th, and Pb Values (all values in ppm)

Sample	Pb	Th	U
BTG 1	4.22	2.63	1.12
BTG 2	2.49	5.47	1.37
BTG 3	0.65	1.24	0.45
BTG 4	7.71	3.71	1.35
BTG 5	5.03	9.01	2.99
BTG 6	7.82	13.62	4.19
END 1	1.89	1.60	0.57
END 2	12.08	1.20	0.53
END 3	26.62	7.13	22.90
BLB 1	5.95	12.13	5.97
SLP 1	2.71	4.23	1.09

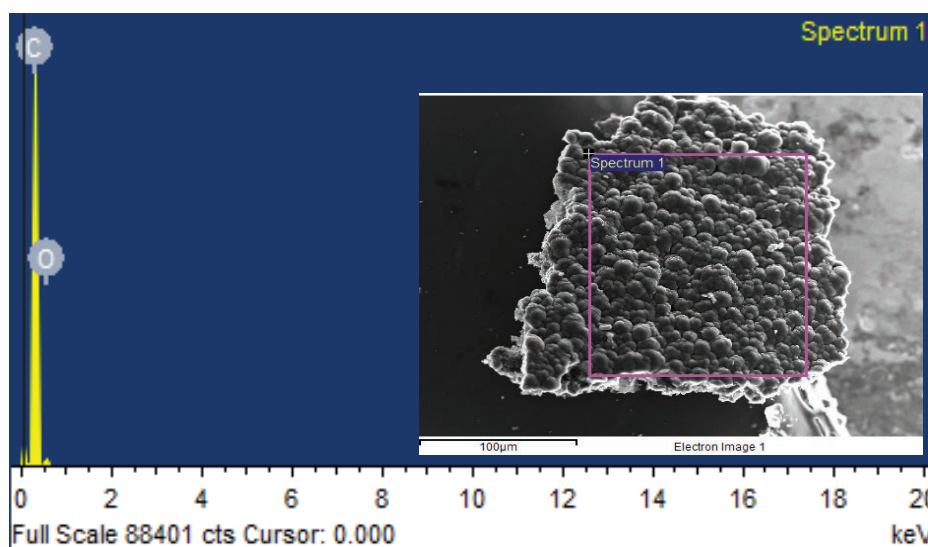


Fig. 14. Backscattered electron image of carbonaceous matter adsorbed on a pyrite surface.

and Meyers, 1989). The hydrothermal input in the study area is fault aided, unlike at Witwatersrand, where fluid circulation occurred due to regional metamorphism: two phases of hydrothermal infiltration and the Vredefort meteoritic impact (Frimmel, 1997).

These conglomerates exhibit striking similarities in mineralization style with certain sediment-hosted gold deposits like Carlin, Bendigo, Spanish Mountain, and Sukhoi Log. The significant enrichment of As along with elevated trace element contents in the diagenetic pyrites (Large et al., 2009), a strong positive correlation between As and Au (Cline, 2001; Reich et al., 2005), the moderate positive correlations shown by Au with Ni and Sb, the presence of solid solution gold within arsenian pyrites, cyclic zones of As in the hydrothermal pyrites, and As-enriched rims of the hydrothermal pyrites (Large et al., 2009) are the similarities observed in the mineralization style between the sediment-hosted gold deposits and the quartz-pebble conglomerates of the study area.

The source of Au in the studied conglomerates, however, remains a puzzle. The pervasive occurrences of detrital chromite and fuchsite suggest ultramafic source rocks in the vicinity of these quartz-pebble conglomerates. The detrital gold is presumed to be sourced from a similar ultramafic source rock. We infer that the gold within the diagenetic pyrites was incorporated from fine detrital particles during pyrite growth. The presence of finely dispersed gold within diagenetic/sedimentary pyrite indicates a genetic link between biological activity and Au deposition (Horscroft et al., 2011; Agangi et al., 2015). The possible role of microbial mats as traps for initial gold deposition (Frimmel, 2015; Tashiro et al., 2017) cannot be rejected altogether, given the similarities these quartz-pebble conglomerates have with the Witwatersrand basin. Besides, the elevated Ni values within the diagenetic pyrites further suggest an organic-matter-rich environment. Frimmel (2014) suggested that anomalous gold concentrations observed in the diagenetic pyrites is a consequence of trapping of Au from river waters onto the sedimentary pyrite surfaces due to microbial fixation or by complex chemical processes. Based

on our studies, we infer that the fixation of Au onto the diagenetic pyrites was microbially mediated. The conglomerate horizon was subsequently infiltrated by S-rich hydrothermal fluids. These fluids precipitated younger pyrites, chalcopyrite, pyrrhotite, and lattice-bound refractory gold. Based on previous studies, it has been established that an increase in As concentration in pyrite and an Fe-depleted surface enhances the uptake of Au in solid solution within pyrite (Fleet and Mumin, 1997; Reich et al., 2005; Deditius et al., 2014). The precipitation of pyrite and lattice-bound gold was facilitated by the Fe in the biotite-arsenopyrite matrix of the conglomerates. The presence of pyrite near the altered biotite matrix might be evidence of Fe replacement during hydrothermal infiltration. We infer that Fe from biotite played a key role in precipitation of pyrite and gold.

The Au and As enrichment on the outermost rim of the hydrothermal pyrites (Fig. 12C) suggests a climax in gold-arsenic transport and deposition (Large et al., 2009). The fluid migration took place through the bedding planes, unconformity surfaces, and fracture networks that developed due to the major fault at Suleipat (Fig. 2). The source of the fluid possibly could be a consequence of devolatilization of mafic-ultramafic rocks during greenschist-amphibolite facies transition (Phillips and Law, 2000). The nearby Archean ultramafic litho units at Kunderkocha gold deposit compose a major portion of fuchsite quartzite, suggesting a hydrothermal imprint on the precursor ultramafic rocks. These rocks could be the possible source of the infiltrating fluids, considering the similar composition and metamorphic grade (Sahoo and Venkatesh, 2015).

The early incorporation of detrital Au followed by its remobilization and reprecipitation along with associated trace elements onto younger pyrites suggests a modified placer model for the origin of gold within these quartz-pebble conglomerates. This is the first report of refractory gold within arsenian pyrites from Archean quartz-pebble conglomerates of eastern India. In addition to encouraging gold values, the surficial samples of these quartz-pebble conglomerates also

reveal minor uranium occurrences. These quartz-pebble conglomerates bear some similarities to the Witwatersrand basin and have a mineralization style that is similar to a few sediment-hosted gold deposits. Overall, these quartz-pebble conglomerate horizons show good potential and generate great interest in the research and exploration of gold within the area and in analogous areas.

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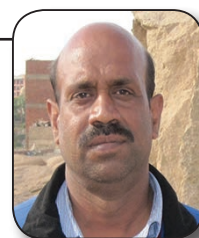
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