Uranium in ancient slag from Rajasthan

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Anomalous radioactivity was recorded in two ancient slag dumps spread on the surface near Bansda (24°35′N lat., 70°09′E long.) and Dhavadiya (24°30′N lat., 70°05′E long.) villages, Udaipur District, Rajasthan. The slag, with a range of high to low radioactivity levels, is the remnant of ancient smelting in the area, probably for copper. Six samples showing low radioactivity in Bansda contain an average of 0.030% U_3O_8, while five moderately radioactive samples analysed contain 0.225% U_3O_8 and four highly radioactive samples analysed contain 1.15% U_3O_8. The 15 samples contain on an average 0.627% copper, 719 ppm zinc, 329 ppm cobalt and 133 ppm vanadium. Fifteen samples from Dhavadiya slag assayed on an average contain 0.040% U_3O_8, 0.297% Cu, 292 ppm Zn and 250 ppm Co. The extent of crystallization seen in the slag is intriguing because an over-cooled melt generally forms glass. The high rate of crystal formation may be attributed to high amounts of volatiles, particularly CO_2 and SO_4, released during the breakdown of limestone (added as flux during smelting) and sulphide minerals in the ore. The high order of radioactivity recorded in the slags of Bansda and Dhavadiya points to the presence of ore-grade uranium concentration associated with sulphide mineralization in the vicinity of the basement Banded Gneissic Complex, intrusive granites and the cover sequence of the Bhinder basin.

Keywords: Ancient slag, mineralization, radioactivity, uranium.

RADIOACTIVITY in slag dumps of unknown age near Bansda and Dhavadiya villages, Udaipur District, Rajasthan have been reported in 1961 and 1962 respectively (P. S. Nikam and D. K. Seth, unpublished of RMD). The present authors studied the occurrences in detail in the field, followed by chemical and petrographic analysis of thirty samples. The results of this study are given here so as to bring this unique occurrence to the notice of the scientific community. Radiometric survey of these ancient slag dumps shows that the radioactivity is not uniformly distributed; rather it is enriched to varying degrees in different pieces. Gamma-radiometric and chemical assays of the samples show that the radioactivity is due to uranium, the content of which varies from a few parts per million (ppm), i.e. the background concentration of rocks, to as much as 1.15% U_3O_8. Although the chemistry of the slags cannot be extrapolated to that of the parent ore, it indicates a high-grade base metal – uranium ore source.

The Aravalli mountain belt in Rajasthan exposes rocks of two Supergroups, namely Aravalli and Delhi. Both the successions form a cover on the basement Banded Gneissic Complex (BGC) and host potential uraniferous horizons by virtue of their geological history in terms of the temporal nature of uranium mineralization as well as the typology of uranium deposits. Another set of geological units equivalent to the Aravallis in Rajasthan are the metasediments of ‘pull-apart basins’[1]. Owing to their peculiar genetic conditions, rocks in these basins host a variety of multi-metallic mineralizations. Exploration by the Atomic Minerals Directorate for Exploration and Research (AMD), Jaipur, in recent years led to identification of uranium anomalies in most of the seven pull-apart basins, while attempts are being made to locate viable deposits in them. The radioactive slag occurrences of Dhavadiya and Bansda are located in the BGC adjacent to one of these belts – the Bhinder belt – and also to the intrusive granites of Untala/Tekan. The BGC comprises TTG gneisses, biotite schist, quartzites, limestones and intrusive granitic and basic rocks (Figure 1)[2]. The Bhinder–Dariba belt comprises quartzites, dolomitic limestone and carbonaceous phyllite/graphitic schist. No clear-cut unconformity could be observed between the basement and cover rocks in this area due to lack of exposed rocks at the contact zone.

Ancient slag occurrences are numerous in Rajasthan. Almost all the base-metal occurrences were discovered by detailed exploration of areas having ancient slag. Hundreds of slag occurrences are present in Udaipur, Dungarpur and Banswara districts of southern Rajasthan, many of which have been reported and explored[3].

A number of old workings and slag dumps have been reported from the Bhinder basin at Wari, Bansda, Nangauli, Dariba and Bhinder. Ancient slag dumps are observed in the basement also, like Bansda and Dhavadiya. Extensive malachite staining is observed both in the Bhinder basin and in the basement rocks. Highly anomalous radioactivity was recorded in the two slag dumps spread on the surface near Bansda (24°35′N lat., 70°09′E long.) and Dhavadiya (24°30′N lat., 70°05′E long.) villages. Thirty samples, 15 from each of the two occurrences, were studied in an attempt to decipher the nature of the slag, extent and source of radioactivity and possible provenance. The following are the field observations pertaining to these slags.

1. The slag is the remnant of ancient smelting in the area, probably for copper. Malachite stained gneiss and quartz vein fragments are present near the slag heaps.
2. The slag is not uniformly radioactive, rather it has high to low radioactivity.

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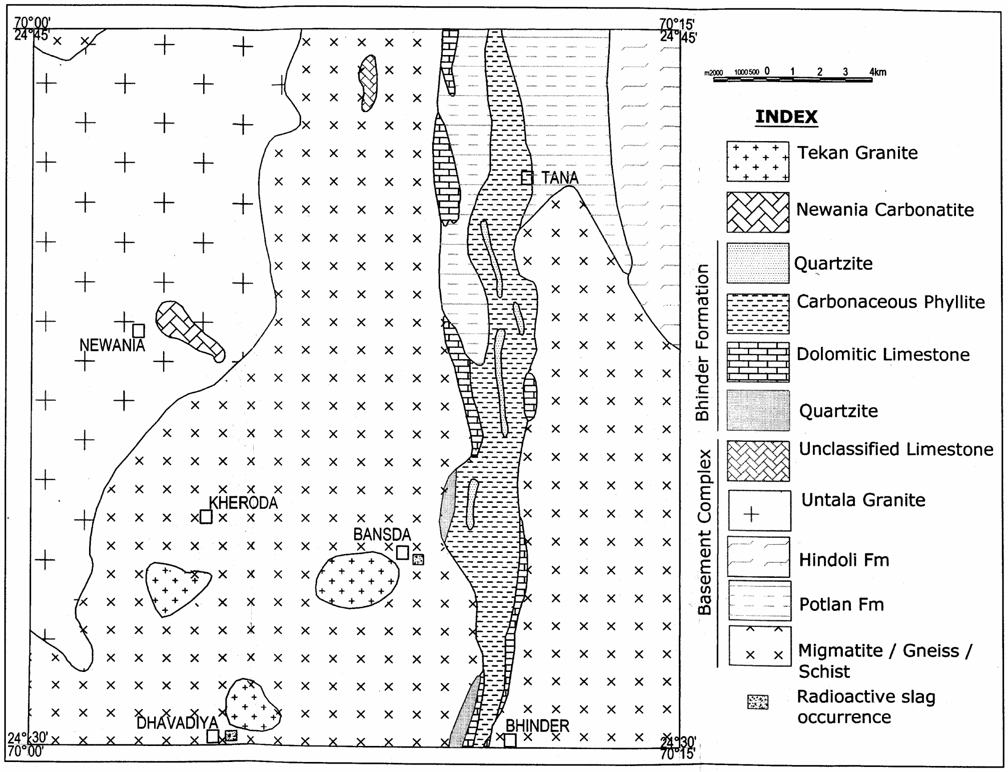

Figure 1. Geological map showing location of slag occurrences near Bansda and Dhavadiya (modified after Gupta *et al.*[2]).

Table 1. Mean chemical composition* of slag from Bansda and Dhavadiya, Udaipur District, Rajasthan

Location	Bansda				Dhavadiya
Parameter	SLA	SSA	SRA	Mean	SSA
SiO_2	34.51	35.94	39.21	36.37	41.57
TiO_2	0.31	0.39	0.26	0.33	0.2
Al_2O_3	5.01	3.83	4.33	4.36	5.69
Fe_2O_3	1.71	1.33	1.97	1.63	2.46
FeO	37.91	37.88	32.79	36.47	26.53
MnO	0.19	0.16	0.14	0.16	0.21
MgO	1.34	0.8	0.9	1.01	2.22
CaO	14.97	16.41	15.75	15.75	17.38
Na_2O	1.06	1.08	1	1.05	1.8
K_2O	0.69	0.85	1.12	0.87	0.89
P_2O_5	0.89	0.78	0.61	0.77	0.51
LOI	0.97	0.9	1.42	1.07	0.32
Total	99.57	100.35	99.48	99.85	99.77
U_3O_8	0.033	0.225	1.15	0.407	0.04
Cu	0.637	0.348	1.035	0.627	0.297
Zn	716	698	753	719	292
Ni	61	38	64	52	<50
Co	303	339	348	329	250
Pb	<10	<10	101	N.A.	50
V	132	134	135	133	N.A.

*Zn, Ni, Co, Pb and V in parts per million, and the rest in percentage. SLA, Low-radioactive (six samples); SSA, Moderately radioactive (five samples from Bansda and 15 from Dhavadiya); SRA, highly radioactive (four samples).

3. The ore which is the source of the slag, does not seem to have been transported over large distances. This is because the distance between slag heaps in the area is of the order of 1 or 2 km only. The distance between the two radioactive slag occurrences is about 10 km.
4. The radioactive material seems to have been part of the Cu ore. However, the entire Cu ore body is not radioactive, since radioactivity is variable.
5. Although the source of radioactivity could have been limestone that was added to the ore as a fluxing agent, the chances for the same are less owing to the variable order of radioactivity and the lack of inherent radioactivity in limestones available in the locality. Had the source been the fluxing material, radioactivity may be expected in practically all slag pieces. Moreover, the slag occurring in the vicinity of locally radioactive Newania carbonatite further northwest of the present area was found to be non-radioactive.

East of Bansda village the slag is spread on the surface over an area of about 300 m × 300 m. The maximum depth of the slag-heap from the surface does not seem to be more than 2 m. Radioactivity over the slag-heap increases up to more than 20 times the background. Radioactivity is not uniform over the slags. Six samples showing low radioactivity in the field contain average

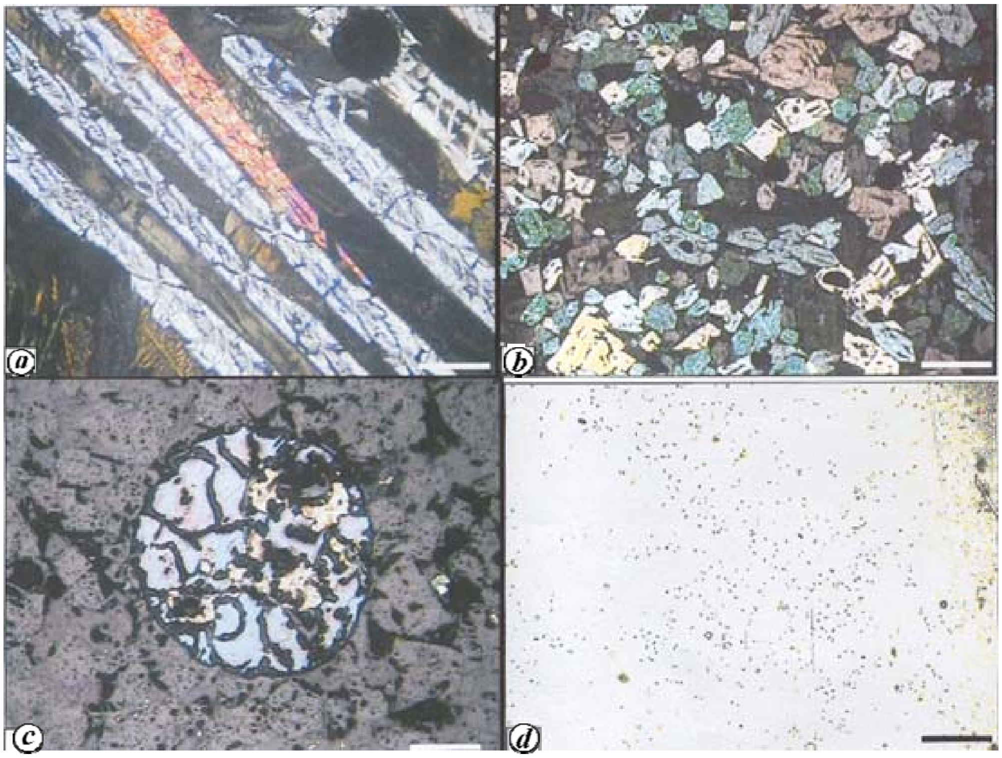

Figure 2. Textures and radioactivity in slag. ***a***, Highly elongate, bladed crystals of olivine in a groundmass of olivine, pyroxene, feldspar and devitrified glass forming spinifex texture. Groundmass at the left bottom shows dendritic olivine crystals. An opaque mineral globule is seen at the top right (TL, XN, bar = 170 μm). ***b***, Equant crystals of olivine in opaque matrix. Opaque minerals are also seen in the core of olivine crystals forming intrafasiculate texture (TL, XN, bar = 850 μm). ***c***, Globular grain consisting of chalcopyrite (yellow) and copper (shades of pink and blue) in the slag (RL, ON, bar = 340 μm). ***d***, α-tracks on cellulose nitrate film (TL, bar = 170 μm) (refer to (***b***)).

0.033% U_3O_8, while five moderately radioactive samples analysed contain 0.225% and four highly radioactive samples analysed contain 1.15%. The fifteen samples contain on an average 0.627% copper, 719 ppm zinc, 329 ppm cobalt and 133 ppm vanadium (Table 1). Attempts to detect *in situ* radioactivity failed, as the area is covered by soil and fragments of slag.

In Dhavadiya, slags are spread over an area of 30 m × 10 m, having depth of the order of half a metre only. In comparison to the slags of Bansda, Dhavadiya slags are less radioactive. In fact, compared to Bansda, highly radioactive slags are totally absent in Dhavadiya. Fifteen samples from the Dhavadiya slag assayed on an average 0.04% U_3O_8, 0.297% Cu, 292 ppm Zn and 250 ppm Co.

Petrographically, samples from both Bansda and Dhavadiya appear similar to ultramafic rocks with major minerals of fayalite (Fe-olivine), clinopyroxenes, amphiboles and chlorite, and subordinate amounts of feldspar and devitrified glass. The most penetrative texture observed is spinifex, which consists of highly elongate bladed crystals of olivine, set in a groundmass of olivine, pyroxene, feldspar and devitrified glass (Figure 2 *a*). A large number of skeletal grains of olivine form dendritic texture that represent very rapid growth during quenching. Apart from this, aggregates of equant crystals are seen set in glassy matrix (Figure 2 *b*). Metals and metal sulphides in the form of rounded crystals and crystalline aggregates are also seen (Figure 2 *c*). No crystalline uranium mineral could be identified in the slag. Cellulose nitrate (CN) films exposed to thin sections of the slag shows scattered α-tracks (Figure 2 *d*) over translucent to opaque material (probably Fe-oxides or hydroxides) and along grain boundaries, suggesting the presence of adsorbed uranium. Positive chromograms of the samples and etching pattern on CN films point to the presence of dispersed leachable uranium in the slag. However, this does not discount the possibility of the presence of crystalline U-minerals in the ore. Textural development, i.e. the crystal habits of spinifex and skeletal nature along with numerous shapes, is typical of furnace slags[4]. More than 80% of the material is crystallized, while the rest occurs as devitrified and normal glass.

The extent of crystallization seen in the slag is intriguing because an over-cooled melt generally forms glass, which would later devitrify to form a microgranular aggregate with the formation of crystals restricted to deeper levels in the melt. The occurrence of well-developed crystals can be explained by the formation of a large number of crystal embryoes owing to high ΔT (difference between melt temperature and surface temperature), followed by high crystal growth rate[5]. High crystal growth rate requires high rate of diffusion (mobility) of the atoms/ions/particles in the melt. A high ΔT reduces the viscosity of the melt, thereby hindering growth rate. To counter this, the melt needs to have high volatile content, which will increase the mobility of the components. In the present case, large amounts of CO_2 and SO_2, released during the breakdown of limestone (added as flux during smelting) and sulphide minerals in the ore, may act as volatiles in the melt. The high content of limestone in the ore-feed is indicated by excessively high CaO concentration in the slag (Table 1), while the high FeO concentration suggests the domination of Fe-bearing sulphides, most probably pyrite and chalcopyrite.

Major, minor and trace-element chemistry of slag samples from Bansda and Dhavadiya (Table 1) points to fairly good similarity between slags of the two areas. A definite relationship between the uranium concentrations in the slag vis-à-vis the major element composition could not be established from the data. The most striking feature of the composition is the excessively high content of FeO and CaO in the samples. Although a wide range of possibilities exist to explain the chemical composition of these slags, keeping in mind the local geology dominated by the Archaean basement gneisses and intrusive granites vis-à-vis the probable nature and genesis of the proto-ore, the excess FeO can be interpreted to be originating from Fe-bearing sulphide minerals (pyrite, chalcopyrite, pyrrhotite, etc.), while the excess amount of CaO + MgO can be interpreted to be originating from the limestone added as fluxing material used for smelting the sulphide ore. Keeping in mind the geology and nature of possible host rocks in the vicinity (BGC/metasediments of Bhinder basin/granitic intrusives), the probable host rock seems to be quartz veins in the periphery of a granite intrusive into the TTG gneisses of BGC. This assumption is supported by the occurrence of vein quartz with malachite stains found scattered near the slags.

The high order of radioactivity recorded in the slags of Bansda and Dhavadiya points to the presence of ore-grade uranium concentrations, associated with sulphide mineralization in the vicinity of the basement BGC, intrusive granites and the cover sequence of the Bhinder basin. Though it is not possible to pinpoint the location of mineralization, it is postulated that it is in the immediate neighbourhood of the slag occurrence. Uranium mineralization is closely associated with sulphide minerals, and also with high Cu, Zn and Ni content. The degree as well as extent of radioactivity associated with base metals in the ancient slags of Bansda and Dhavadiya warrant concerted efforts to locate the primary uranium–base metal deposit in the area. This may be achieved by well-planned geophysical (electrical and/or electromagnetic) surveys in the first phase and drilling in the subsequent phase. Since the probable country rocks are basement gneisses, attempts need to be made to thoroughly investigate the BGC terrain for potential high-grade uranium mineralization.

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ULF emissions associated with seismic activity recorded at Kolhapur Station

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In this communication, we present some observations and preliminary results associated with seismogenic ultra low frequency (ULF) emissions. We have analysed ULF data for two moderate earthquakes that occurred on 17 April and 21 May 2006, with magnitudes greater than 4. For recording ULF data using ground-based method, three induction-coil magnetometers have been installed at Shivaji University, Kolhapur, India (16.40°N, 74.15°E). Data have shown maximum enhancement in intensities of magnetic field for 0.1 Hz, 3–5 days before the earthquakes. Polarization parameter and geomagnetic pulsation in terms of *Kp* index have been used to conclude that the observed enhancement in magnetic field (before the earthquake) is seismogenic.

Keywords: Earthquakes, electromagnetic emissions, polarization ratio, power spectrum analysis.

EARTHQUAKE prediction is one of the most challenging problems in earth sciences. Association between possible

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