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Uranium Traps in Phreatic Sandstone-type Prospect, Taunsa Area, Dera Ghazi Khan, Eastern Sulaiman Range, Pakistan: Evidences from Autoradiography and Optical Microscopy

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Abstract: Taunsa uranium occurrence like other uranium resources in Pakistan is hosted by the Late Miocene-Pliocene age Litra Formation of the Siwalik Group molasse sediments. Taunsa uranium prospect is a unique phreatic-type uranium resource in terms of its disturbed geological setting of the eastern limb of the Zindapir anticline in the eastern Sulaiman range. Autoradiography technique was used to locate the spots of anomalous uranium concentration in thin sections from ore of Taunsa prospect. Twenty polished thin sections from uranium ore ranging from 200 ppm-600 ppm were attached to detectors for a month which produced prominent alpha track which were used to find the traps of uranium. Subsequently, these spots were studied under SEM and EPMA for further investigations of uranium phases. Autoradiography revealed that Taunsa uranium ore is mostly associated with organic matter (probably petroleum), black shale clasts, biotite, fougerite (a green colour rusty mineral) and with micritic clasts. This study suggests that prospective facies of the host sandstone containing relatively abundant black shale clasts, organic matter and biotite may be targeted during exploratory drilling in Taunsa uranium deposit and its extensions in the eastern limb of Zindapir anticline.

Keywords: Uranium prospect, traps, autoradiography, Litra Formation, Sulaiman range.

Introduction

Sandstone-type uranium deposits are hosted by medium- to coarse-grained, arkosic sandstone facies, containing relatively abundant organic matter and/ or pyrite. Generally, these sandstones are fluvial in origin, bounded by mudstones (IAEA-TECDOC-1629, 2009). Primary uranium minerals are predominantly pitchblende, coffinite and to a lesser extent vanadate and phosphates (Finch, 1967 1985; Grutt, 1972). The common reducing agents which cause oxidized uranium to precipitate include carbonaceous material, sulphides, hydrocarbons and some of the ferromagnesian minerals (e.g., chlorite).

Generally, major sandstone-type deposits occur in Palaeozoic to Tertiary age but some deposits also occur in sandstone belonging to Precambrian, associated with carbonaceous matter of probable algal origin, and with inorganic reducing agents such as basic dikes and sills in Australia (Ryan, 1979) and in Canada (Alexandre et al., 2015). Sandstone-hosted uranium deposits can be divided into four main types; roll front- tabular-, tectono-lithologic-, and basal channel- types (OECD/NEA, 2003).

Pakistan possesses some of the favourable geological conditions required for the origin of uranium resources both in hard and soft rock settings (Dahlkamp, 2009). However, uranium in Pakistan is mined only from sandstone of the Litra (Dhok Pathan) Formation both in Bannu basin and in Sulaiman range (Fig. 1).

Sandstone-type deposits of Pakistan show characteristics of roll front-type deposits such as the occurrence of oxidized sandstone between the ore lenses or the two limbs of the roll. However, in some ore bodies, only one limb is present and the ore body can still be classified as roll-type (Finch, 1985). Roll-front deposits are normally characterized by disequilibrium due to downdip movement of the roll-front (Rosholt, 1959). In Taunsa area, the surface outcrop hosts four radioactive sandstone units interlayered with marker mudstones. Theses radioactive sandstone units are known as Sandstone-1 (S-1), Sandstone-2 (S-2), Sandstone-3 (S-3) and Sandstone-4 (S-4) in stratigraphic order (Fig. 2). Thickness of the radioactive parts of the host sandstone varies from 30 m-100 m while the strike-wise length varies from 4 km-10 km. Radioactivity on the outcrop ranges from 200 cps-500 cps with SPP2 NF scintillation counter against a background value of 70 cps. Radioactivity on surface is generally associated with hard cemented layers locally termed as hard bands. However, sporadic vertebrate bone fossils embedded in host sandstone generally show higher radioactivity. Taunsa uranium occurrence is a phreatic-type uranium prospect. It shows extremely positive disequilibrium due to its very young nature. Young uranium ore is weakly radioactive, and can be hardly detected with gamma ray spectrometers (Culbert and Leighton, 1988).

This paper discusses the traps of the Taunsa uranium ore established with autoradiography and optical microscopy, and their implications for uranium exploration in Litra Formation in the eastern limb of the Zindapir anticline in Sulaiman range.

Materials and Methods

In this study low grade samples of Taunsa uranium ore ranging from 200 ppm-500 ppm were treated with autoradiography, and the radioactive spots were then

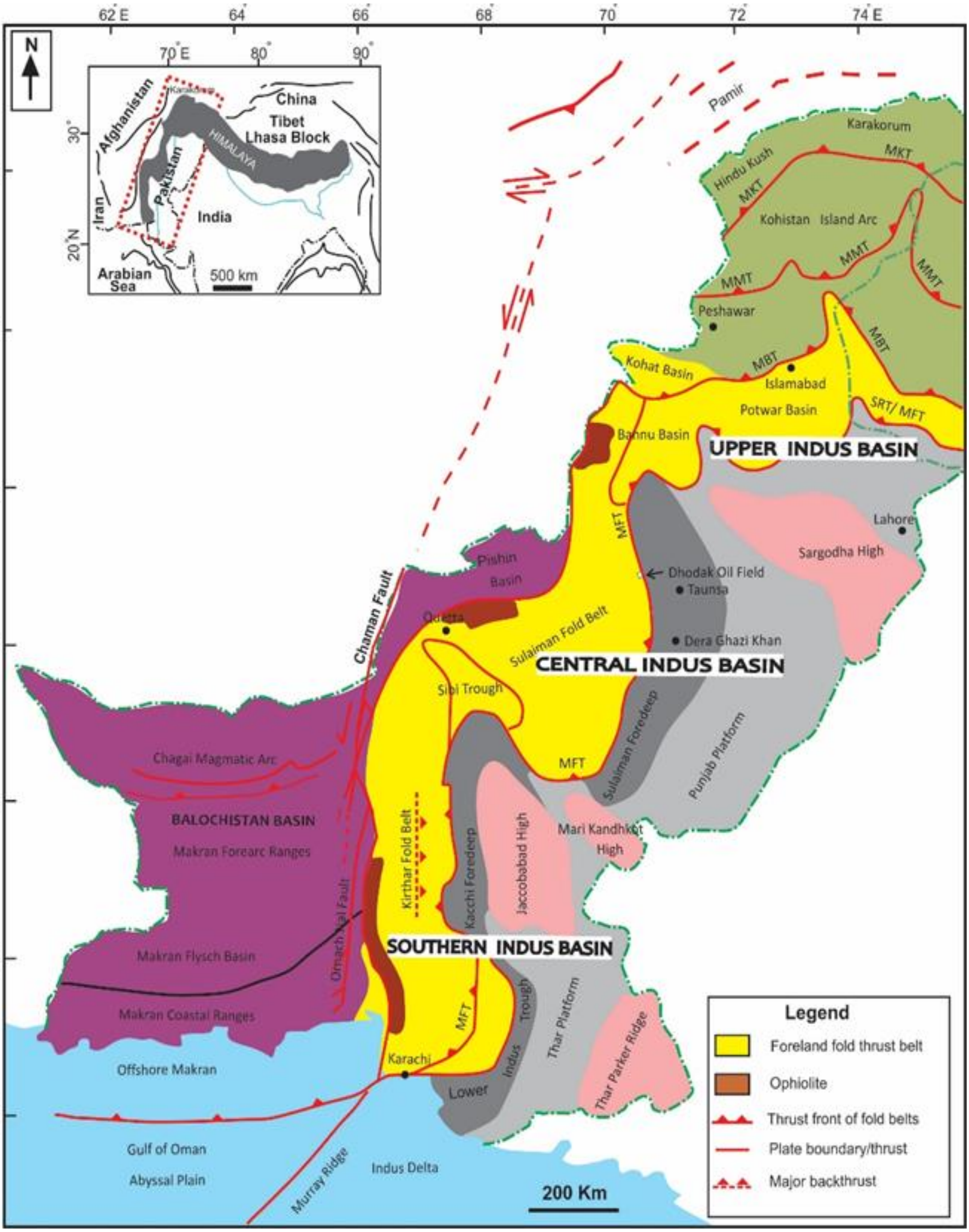

Fig. 1 Tectonic and sedimentary basin map of Pakistan (modified after Aziz and Khan, 2003). Inset shows the entire Himalayan Range (modified from Najman, 2006). The dotted rectangle in the inset map is the Pakistani terrain. MKT: Main Karakorum Thrust, MMT: Main Mantle Thrust, MCT: Main Central Thrust, MBT: Main Boundary Thrust, SRT; Salt Range Thrust, MFT: Main Frontal Thrust.

studied under transmitted and reflected microscopes. Subsequently, these uranium-bearing spots were further investigated by scanning electron microscope (SEM) and electron probe micro-analyzer (EPMA).

To get alpha tracks for delineating spots of anomalous uranium concentration, twenty polished thin sections were used. Pieces of photographic film (Fujifilm Pro 160, roll) were attached to these thin sections following the method of Xu et al., (2010). This procedure is discussed below:

1. To remove emulsion, the film was immersed in a strong alkaline water (15 g-20 g of NaOH in 100 ml water) for half an hour and the remaining transparent film (the base film) which is cellulose acetate film or nitrocellulose film, was rinsed with water and dried.

2. The film sheet was cut into pieces in slightly smaller sizes than the corresponding thin sections.

3. Then film pieces were washed with alcohol and fixed to thin sections with clamps for irradiation from the radioactive elements in thin section for some period depending on the concentration of uranium (Table 1).

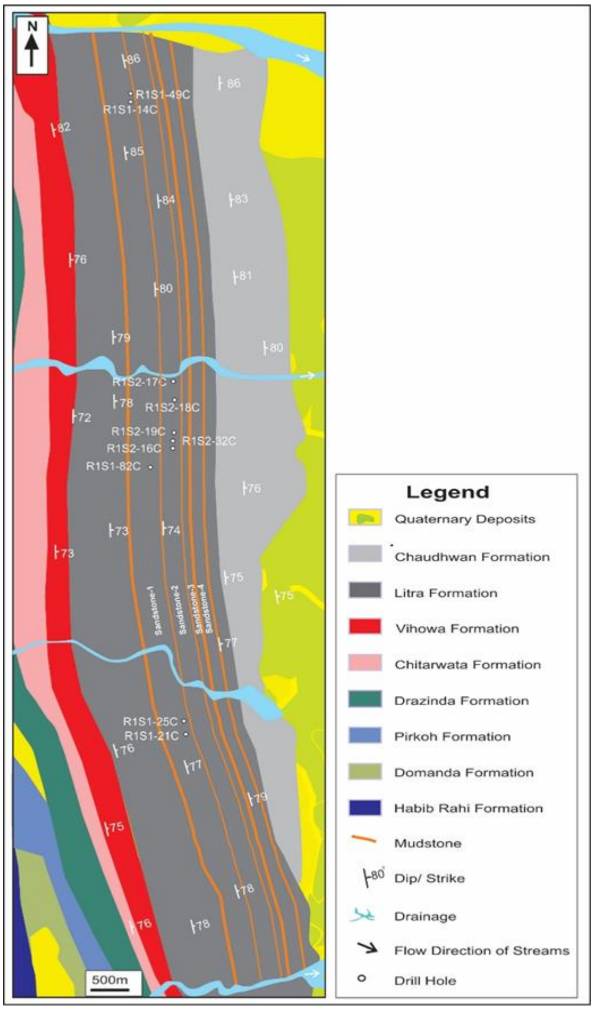

Fig. 2. Geological map of study area (modified from Hassan, 2002). Map shows locations of core samples (drill holes).

Table 1 Irradiation duration of detectors with uranium ore of different concentrations

S.No.	Uranium concentration (ppm)	Irradiation time (days)
1	<30	40
2	30-100	30
3	100-200	30-20
4	200-500	20-10
5	500-1000	10-5
6	>1000	2-5

4. When the required irradiation period expired, the films were removed and immersed in a strong alkaline solution at a constant temperature of 600^0 C to 650^0 C. Ten pieces of the film can be etched in 100 ml etching solution in 60 minutes for much better effects. Either of the following etching solutions can be used which have the same effect:

i. 40g KOH+5g $KMnO_4$+100 ml H_2O
ii 30g KOH+5g NaOH+5g $KMnO_4$+100 ml H_2O

5. After etching to about 60 minutes, the films were removed and rinsed with water. Some precipitates still adhering to the film were removed by immersing the films in 1:1 hydrochloric acid for more than 30 minutes.

After rinsing and drying, the films were observed under optical microscope for alpha tracks, and their corresponding spots on thin sections were studied for the association of uranium, i.e., uranium traps.

Results and Discussion

Autoradiography shows that alpha tracks are associated mainly with organic matter, black shale clasts, biotite and rarely with a green color secondary mineral phase (fougerite) and with micritic clasts (Fig.1). Organic matter is an important reductant in sandstone-type ores (Granger et al., 1961; Motica, 1968; Rackley et al., 1968; Squyres, 1972; Fischer, 1974). Different organic reductants include coalified vegetal matter, woody fragments, structureless organic matter (humate) and petroleum (dead oil) (IAEA-TECDOC-1629, 2009). The role of organic matter for uranium in sandstone-type uranium is four-fold including mobilization, transportation, concentration, reduction and preservation (Leventhal, 1979).

In mobilization, the organic matter is decomposed into CO_2 and organic acids which mobilize uranium from surrounding rocks. Moreover, uranium is transported as dicarbonate or as soluble organic complex in water.

In the concentration process, the organic matter either chelates or binds uranium with its respective functional groups (e.g., humic acids) and precipitates uranium at the interface of recharge and aquifer waters (Leventhal, 1979).

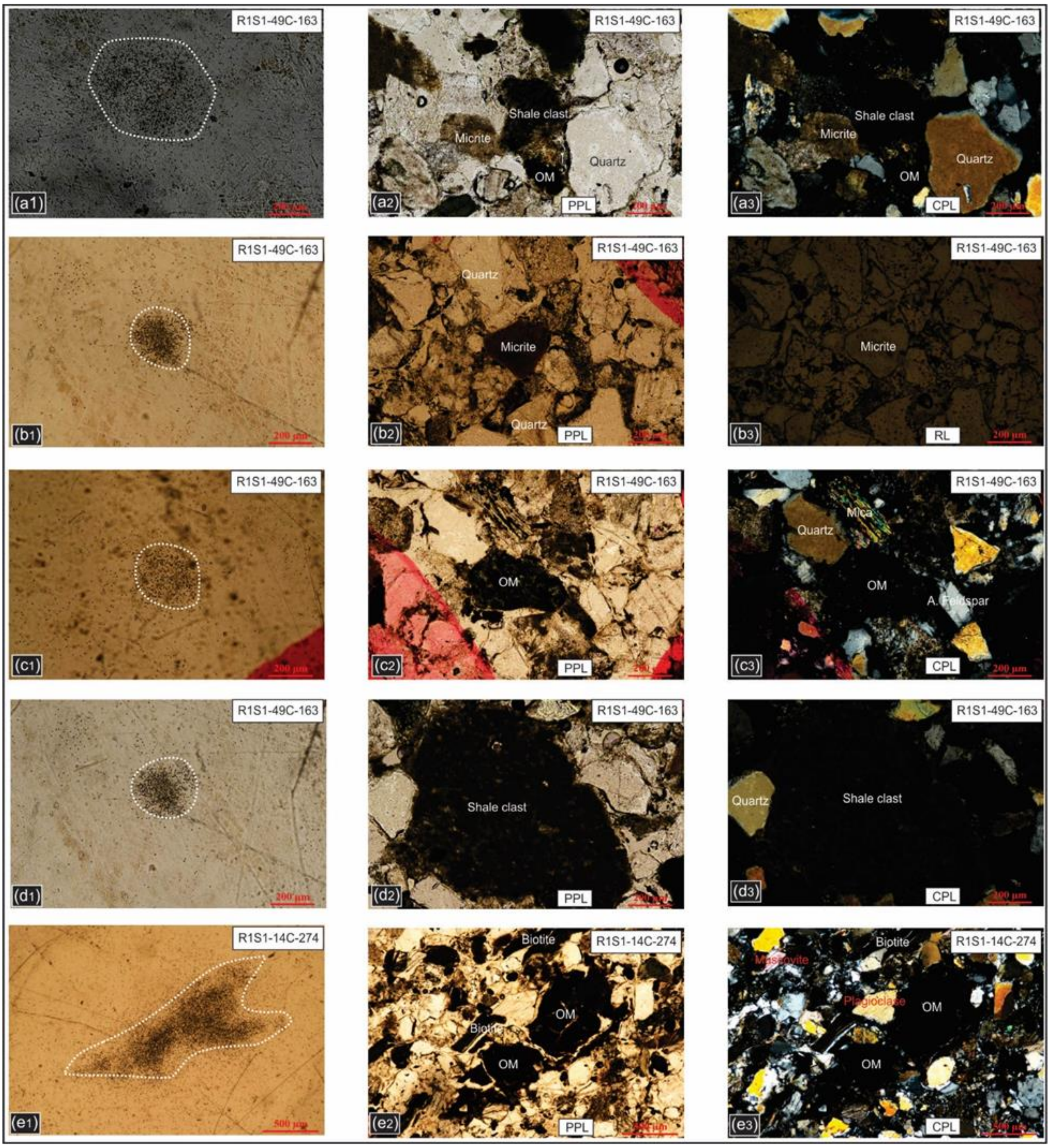

Fig. 3 (a-e). Alpha tracks and photomicrographs showing uranium traps. First picture (1) shows alpha tracks enclosed by the white dotted boundary; second (2) and third (3) pictures show the plane and cross polarized light views (PPL and CPL), respectively except b3 which shows reflected light (RL) view. Sample names have been indicated on the top right corners of the photomicrographs. Uranium is trapped by organic matter (OM), black shale clasts, biotite and a green diagenetic rusty mineral (fougerite). Alpha tracks are enclosed by a white dotted boundary.

In reduction, uranium attached to the organic matter is slowly reduced into its primary phases such as pitchblende or coffinite through abiogenic decomposition.

In preservation process, the reduced uranium phase intimately associated with organic matter is protected from oxidation and mobilisation.

Uranium and organic matter are intimately associated with most sandstone-type uranium deposits in the United States (Leventhal, 1979). However, some uranium deposits in the United Sates lack considerable organic matter. In some south Texas uranium deposits, organic carbon is less than 0.1 percent (Goldhaber and Reynolds, 1977). These deposits are generally low grade (<500 ppm) and the genesis of uranium deposits in this case can be explained by migration of H_2S seeping up along faults (Goldhaber et al., 1979). H_2S is also produced by bacteria which reduce sulphate phases using petroleum as an energy source (Leventhal, 1979).

The average organic matter content (i.e., organic C) in Taunsa uranium prospect is quite low (0.086% ± 0.093 C org.) like that of south Texas uranium deposits with < 0.1% C org. (Goldhaber and Reynolds, 1977).

Secondly, organic matter in Taunsa uranium ore does not show a linear positive correlation as found in most sandstone-type uranium deposits in the United States (Granger et al., 1961). It implies that uranium ore in Taunsa prospect has not always been trapped only by the organic matter, and some other reducing phases, biotite and green secondary phases (fougerite) have also trapped uranium (Fig. 3). Furthermore, the role of multiple reductants, e.g., ilmenite, biotite and pyrite in addition to organic matter, in trapping uranium has also been confirmed by SEM and EPMA studies which were carried out along with autoradiography and petrography.

The organic matter in Taunsa uranium ore is possibly related with dead oil (solidified HC). This observation is supported by the extensive bleaching visible in the host sandstone on the outcrop which is generally considered an effect of petroleum migration into oxidized rocks (Schumacher and Abrams., 1996; Petrovic et al., 2008; Rainoldi et al., 2015).

However, the organic matter and uranium phase is

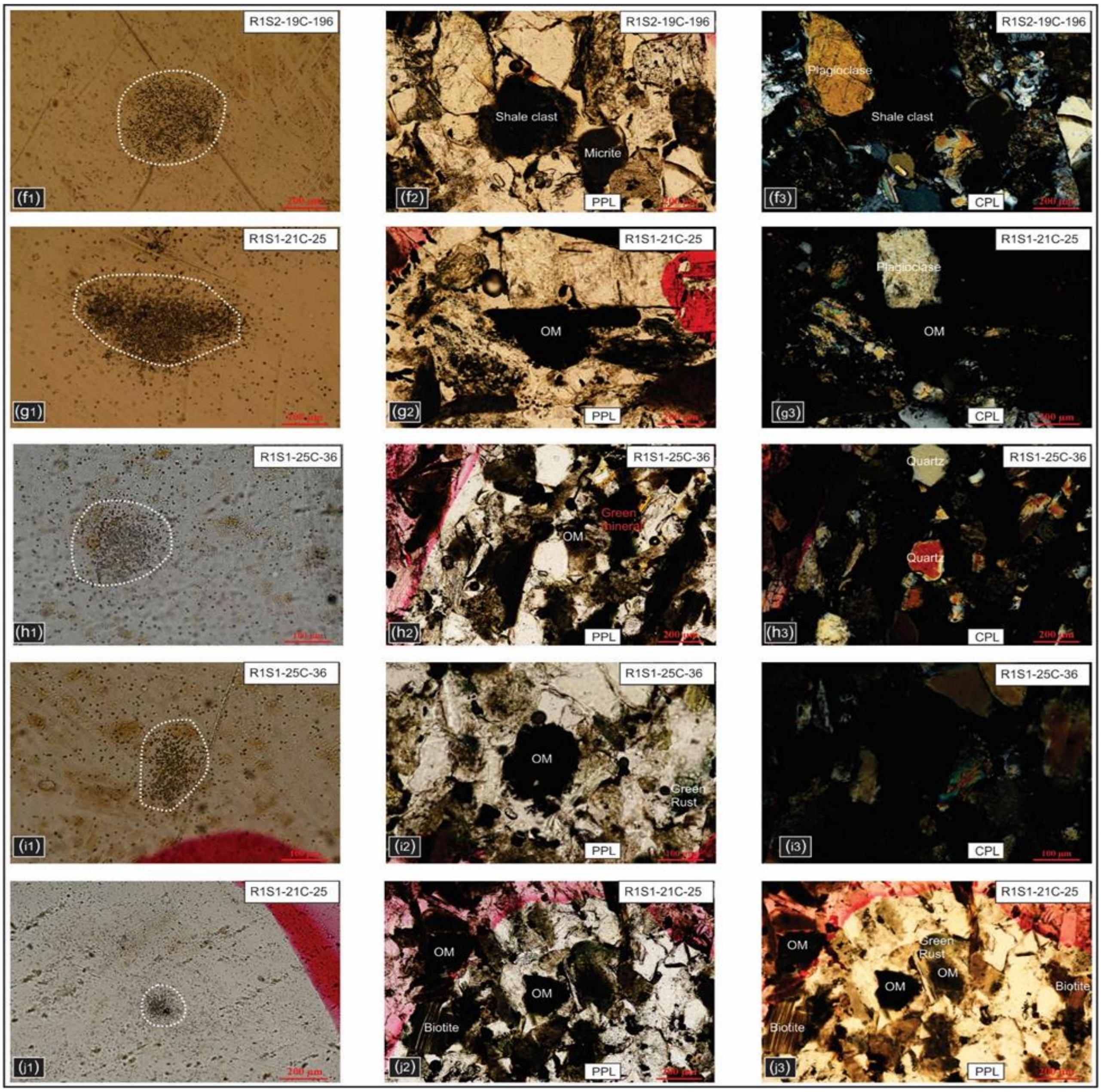

Fig. 3 (f-j). Alpha tracks and photomicrographs showing uranium traps. First picture (1) shows alpha tracks enclosed by the white dotted boundary; second (2) and third (3) pictures show plane and cross polarized light views (PPL and CPL), respectively. Sample names have been indicated on the top right corners of the photomicrographs. Uranium is trapped by organic matter (OM), black shale clasts, biotite and green diagenetic rusty mineral (fougerite).

not intimately associated because some organic matter does not show any alpha tracks, which may suggest the absence of uranium or any other radioactive elements in the organic matter.

It has been noticed in thin sections study that biotite has trapped uranium as indicated by SEM/ EPMA investigations. Biotite adsorbs and reduces oxidized uranium possibly by its ferrous iron (Idemitsu et al., 1995).

Uranium content of the black shale clasts in Taunsa uranium may be a syngenetically trapped uranium in marine environment during sedimentation and/ or it may be an epigenetically added uranium to black shale clasts within the host sandstone.

Black shales of the Sulaiman range near study area contain 30 ppm-70 ppm U_3O_8 which points to the fact that some uranium in the black shale may likely be syngenetic. However, the bulk samples analyses of the outcropping host sandstone have an average value of 2.904±2.29 ppm U_3O_8, while the reduced sandstone below the redox boundary contains an average value of 12.94 ±8.61 ppm U_3O_8. It may imply that anomalous uranium in the black shale clasts of the host sandstone may be related to both syngenetic (Swanson, 1961) and epigenetic processes.

A green color diagenetic mineral occurring along the grain boundaries and/ or within grains has been found to be associated with alpha tracks at some places. This green phase initially known as green rust (Feitknecht and Keller, 1950; Bernal et al., 1959) has been recently identified as a new mineral, fougerite (Trolard et al., 2007).

In addition, some greyish brown micritic (microcrystalline calcite) fragments show alpha tracks. Carbonate rocks are generally considered to lack uranium resources due to mobility of uranium in water containing carbonate and bicarbonate complexes (Goswami et al., 2017). However, some carbonate rocks are uranium resources, e.g., Todilto limestone in USA (Berglof and Mclemore, 2003) and dolostone of the Vempalle Formation in India (Goswami et al., 2017). Todilto limestone is associated with evaporites, and is considered to have deposited in lacustrine environment (Berglof and Mclemore, 2003). In Sulaiman range, a similar succession of micritic carbonate rocks, black shales and gypsum is found in Baska Formation. These carbonates and black shales may have contributed some uranium to Litra Formation. However, the possible potential of uranium in these units needs to be confirmed by systematic studies.

Conclusion

Autoradiography in low grade ore samples from Taunsa uranium prospect helped to delineate uranium traps which include organic matter, black shale clasts, biotite, fougerite and micrite. The organic matter seems to be humate (structureless organic matter) and is related possibly to hydrocarbons sourced from the older marine black shales of the Sulaiman range. Uranium trapped in black shale clasts seems to be mainly epigenetic. Biotite and a green rusty mineral (fougerite) are also uranium traps probably due to reductive character of the ferrous iron in their structure. The organic matter in micritic limestone fragments may have trapped uranium.

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