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Geological relationship between hydrocarbon and uranium: Review on two different sources of energy and the Indian scenario

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A R T I C L E I N F O

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A B S T R A C T

Organic matter (OM) is often found to be associated in many cases with uranium (U) mineralization displaying spatial, statistical or molecular relationships. The hydrocarbon (HC) system and uranium metallogeny often show geographic, geologic and temporal similarity, which indicates the biophile tendency of uranium. Search for either energy sources can lead to the discovery of the other, and therefore knowledge of their co-occurrence is crucial. The known depositional relationship of uranium ore to oil- and gas-bearing structures indicated well define proved and probable reserves at different places in the world. Further, hydrocarbon (HC) and uranium association can be most promising in sandstone followed by black shale, peat-bog, lignite and phosphorite types of host. Twenty six Indian basins from a hydrocarbon potentiality view are examined along with major seven uranium provinces and other significant uranium occurrences to discuss the U-OM correlation.

1. Introduction

Presently, uranium is a significant source of energy. Crustal abundance of uranium is ~2.7 ppm (Heier and Rogers, 1963; Taylor, 1964) and global cumulative mine production is about 60,000 tonnes per year, out of which two-third production of uranium is from Kazakhstan, Canada and Australia (World Nuclear Association, 2020). However, unlike other sources uranium gives more than 10,000 times energy per kg (Table 1). Nuclear energy is a carbon emission free electricity source. Hydrocarbons (HCs) and uranium co-exist in certain deposits, and therefore their study in that context is of immense economic potential. This is because a single investment cost to locate either HC or uranium deposit can give discovery of both of them. The general aspects of uranium and HC are presented in Repository Section 1.

This article reviews the OM and uranium association globally with a focus on the Indian context. We principally discuss the organic-inorganic interaction mechanism, which is often observed as significant process in mineral resource development (Mao et al., 2014).

2. Overview of coal and hydrocarbon association

Extraction of significant amount of hydrocarbon from coal deposits are practised in China (Thomas, 2002). Notwithstanding, none of the world's giant oil fields come from coal-bearing rocks. Also, there are several coal reserves devoid of oil and gas. To judge potential for oil in coal, one needs to know about the organic matter and material in coal (Flores, 2014).

Depth-wise ranking of coal and petroleum are similar. There exists an optimum total carbon to be attained by coal so that crude oil starts generating. Relation between coal rank and the category of hydrocarbon can be checked in USA but not in Europe since in the later region, not too many coal fields exist (Francis, 1961). One of the first studies on finding out the link between coal and petroleum came from a coal mine where oil production started rather early in Pennsylvania (USA) (Chapman, 1973). A clear-cut relation between coal and petroleum generations do not exist (Chapman, 1973).

Diagenesis transforms the organic matter into kerogen and *"rest of the massive organic matter"*. The former transforms into oil, wet gas and condensate, and the later into coal and CH_4 (review in Biswas, 1987). Maximum similarity is noted between the Type III kerogen and coal. Type III kerogen/coal is associated with mainly gas, and sometimes oil depending on the content of liptinite (Tissot and Welte, 1984). Depositional environment and age of coal are important factors in generating oil from coal. In coal, *(i)* liptinite (exinite) materials that consist of

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Table 1
Comparison of different types of fuel and corresponding energy generation (source: https://www.world-nuclear.org/information-library/nuclear-fuel-cycle/introduction/energy-for-the-world-why-uranium.aspx).

Fuel type	Energy production
Dry firewood	16 MJ/kg
Lignite coal (Brown)	10 MJ/kg
Black coal (low quality)	13–23 MJ/kg
Hard Black coal (high quality)	24–30 MJ/kg
Natural Gas	38 MJ/m^3
Crude Oil	45–46 MJ/kg
Natural Uranium - in typical reactor	500,000 MJ/kg

lipid-enriched kerogen (Type II, 20–30%) and alginate/algal/sapropelic kerogen (Type I; 10–20%), *(ii)* certain vitrinites are oil-prone (Flores, 2014). Type III kerogen is gas prone (Biswas, 1987). Coal and carbonaceous shale, together called peat bog, fund in continent can act as a source rock for petroleum (Zimmerle, 1995).

Coal derived gas, even if of minor amount, has the potential to be explored. Hydrogen generating coal has potential for generating oil. Plan materials and their potential for preservation ultimately control the hydrogen index of the coal. High H_2-rich index of coal can be *(i)* Eocene coals deposited in transgressive rift environment, and *(ii)* Oligocene Miocene coals developed in regressive environment (Flores, 2014). Hydrocarbons derived from coal are profusely found from Cenozoic reservoir rocks (Flores, 2014). However, coals having same maturation and origin can show quite different hydrocarbon generation capability.

Around 60% of the world's oil provinces are related with coal deposits (Tiratsoo, 1951). Coal and hydrocarbon have been found to coexist in few places in the world including several Indian oil fields e.g. *(i)* the Kalol pay horizons IX and X in Cambay, Gujarat, within the Middle Eocene coal-shale-sandstone sequence (Biswas, 1987), *(ii)* Oligocene coal-shale sequence in Assam, and *(iii)* paleocene-Eocene clastic sequence in western offshore (Biswas, 1987).

The carbon ratio theory, referred in many petroleum geology text books, links coalification grade with the specific gravity of oil, and finally with the petroleum production (Biswas, 1987). Carbon ratio can be comparable to the degree of metamorphism of the country rock (Levorsen 1967). The theory led to decide that where the calcification process has gone beyond a certain point (61–63% of fixed carbon, 37–39% of volatile), attempt for hydrocarbon exploration should be avoided (Francis, 1961). No oil fields are found where the coal has fixed carbon ratio of 80%, where the magnitude is 70%, small gas fields are noted, and 55–65% is the ideal range of most of the major oil fields (Tiratsoo, 1951). High grade anthracite is usually not associated with hydrocarbons (Levorsen 1967).

3. Different relationship between OM and U

Common association of OM and U (Russell, 1958) has led many uranium geologists to comment on the importance of OM (e.g., Granger et al., 1961; Szalay, 1964; Fischer, 1974; Rackley et al., 1968; Motica, 1968; Squyres, 1972; Breger, 1974; Manskaya and Drozdova 1968; Sctimidt-Collerus, 1969). As far as the biophile tendency of uranium is concerned, three types of relationships between U and OM can be observed (Goswami et al., 2017a, 2018).

I. Spatial relationship: At the microscopic, hand specimen and regional map scales, it is seen that the respective distributions of U and OM match. For example, polished hand specimen of columnar stromatolite (layers produced microbially by sediment trapping and binding) may show alpha tracks oncellulose nitrate (CN) film (Fig. 1), which indicate a spatial association of OM and U.
II. Molecular relationship: Chemical bonds can be established between functional group of the OM and the uranium compounds. In this regard, the interactions of dissolved OM with inorganic colloids were reviewed extensively by Philippe and Schaumann (2014) and schematic depiction of various sorption mechanisms were summarized (Fig. 2).
III. Statistical relationship: A significant positive correlation coefficient is often found between total organic carbon (TOC) and U content (e.g., Huang et al., 2015) (Fig. 3).

However, it is not necessarily obvious to find all three types (I, II and III stated above) of relationships to co-exist in the same outcrop. Fundamental determinants of uranium mineralization are pH, Eh, oxidation states and the abundance of different ions (viz., OH^-, CO_3^{2-}, PO_4^{3-}, SiO_4^{4-}, SO_4^{2-}). Mainly four major factors (source, migration, precipitation and preservation) influence uranium and OM coexistence. The attributes like type and distributions of OM, chemistry as well as migration pathway of the uranium carrying solution system, redox chemical reactions, porosity and nature of fractures in the host rock, maturity of the OM and diagenesis of the host rocks control the following five steps: mobilization, transportation, concentration, reduction and preservation:

Primarily OM can be considered as an influencer on mobilization of uranium from igneous rocks, where decomposition of biological material raises the partial pressure of CO_2 and forms organic acids in open system and oxidizing condition. Both of these processes may mobilize uranium by leaching and U-OM complex development respectively (Spirakis, 1996).

The second possible role is a extension of point I when organic decomposition products act as transporting agents for oxidized uranium

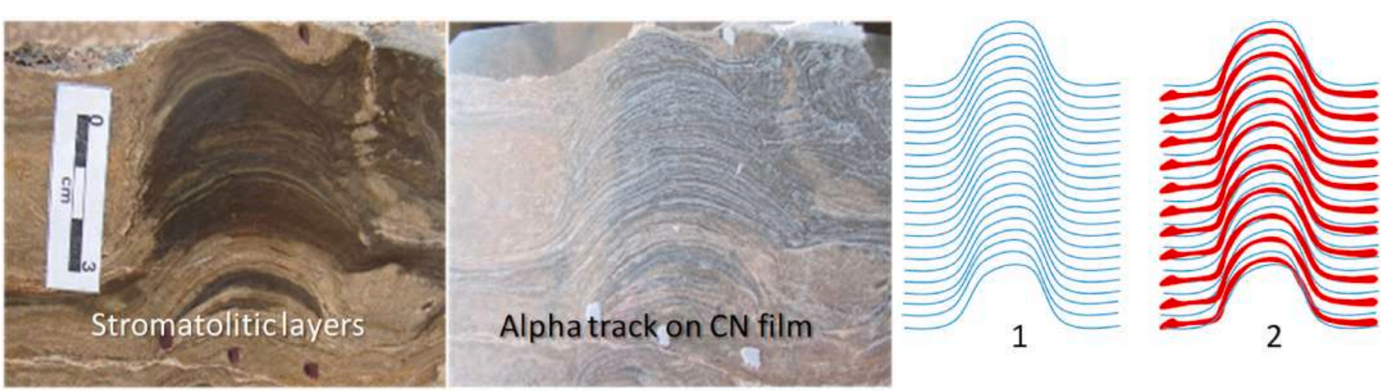


Fig. 1. Columnar stromatolites layers showing white alpha tracks on cellulose nitrate (CN) film placed over the polished sample. This spatial relationship can be visible directly on hand specimen scale. Sketch is shown for such physical adsorption of uranium phase (red) along organic layers (blue) **(this work).** (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

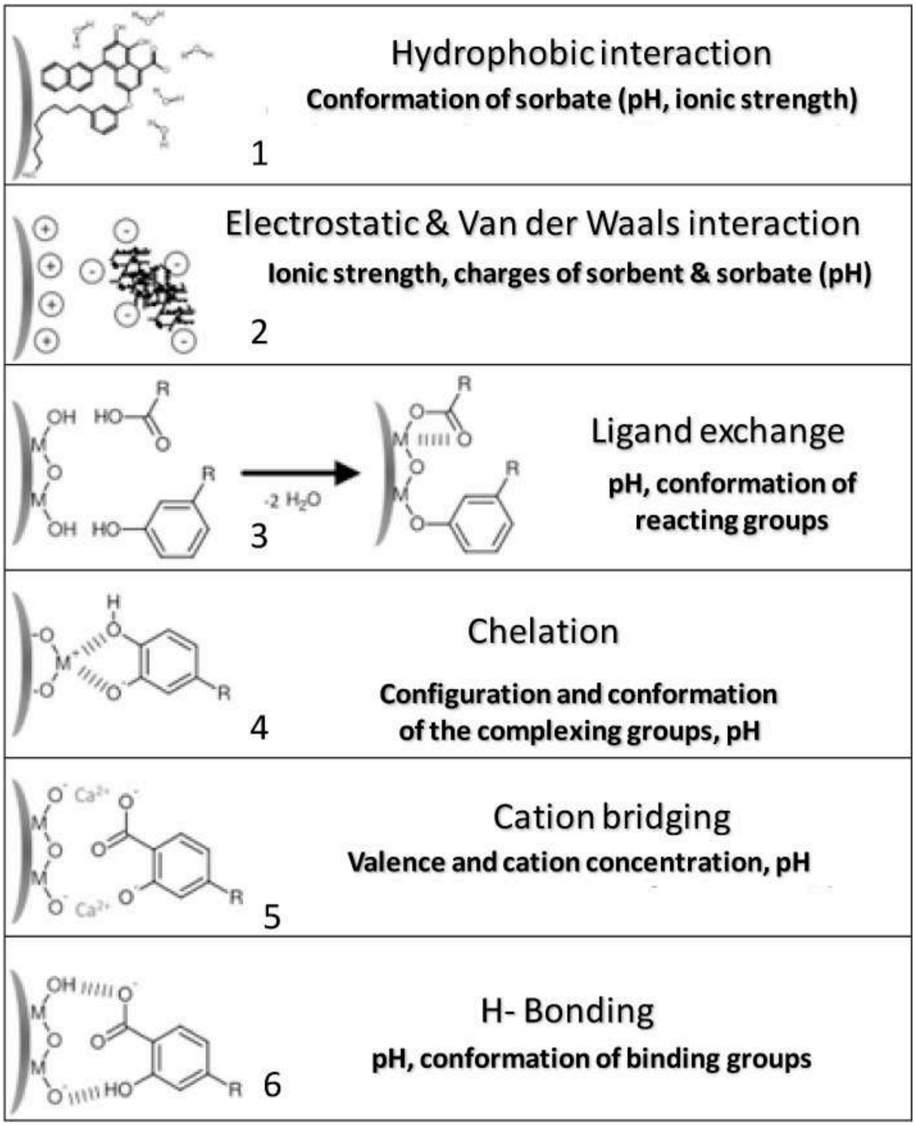


Fig. 2. Depiction of different molecular relationships, bonding, sorption mechanisms of OM (modified after **Philippe and Schaumann, 2014**).

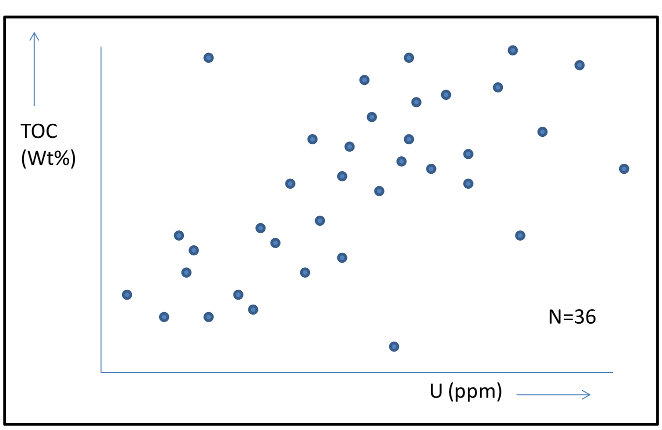


Fig. 3. Schematic depiction plot of positive correlation between TOC and uranium **(modified after Goswami et al., 2018)**.

species. There is certain OM such as fulvic and humic acids as well as smaller organic molecules, which are capable of complexing and transporting oxidized uranium species (Spirakis, 1996).

This step depicts possibly the most important role, in which OM is essential in forming high-grade uranium mineralization. The OM can help to concentrate uranium 10,000 times from water (Szalay, 1964) by acting as reductants. OM and U can also participate in typical redox reaction, in which OM can give electron to uranium. Thus, by gaining electron U become reduced and by losing electron OM become oxidized. On the basis of this ratio, water with 50 ppb U passing over OM may result in water with 500 ppm U (Leventhal, 1979). In fact, 1 to 1 ratio by weight between OM to U is found in the Grants District (Granger et al., 1961). Rocks with 1% OM can contain ~ 1% U. Concentration factors as high as 10 have also been shown for fulvic acid by Jennings (1976) and Jennings and Leventhal (1976).

Not all OM is capable of directly concentrating uranium. For example, the uranium content of petroleum is generally in the range of only a few parts per million (Erickson et al., 1954). Moreover, even the presence effective types of OM are also not the sufficient criteria unless uranium is present. The best evidence for the importance of OM in concentrating U is to look at those deposits in which OM is absent. In some south Texas deposits OM is negligible (<0.1%) (Eargle et al., 1975; Goldhaber and Reynolds, 1977; Goldhaber et al., 1979). These deposits are generally of low grade (<500 ppm) and the possible genesis was explained by H_2S seepage along faults. However, the H_2S is produced by

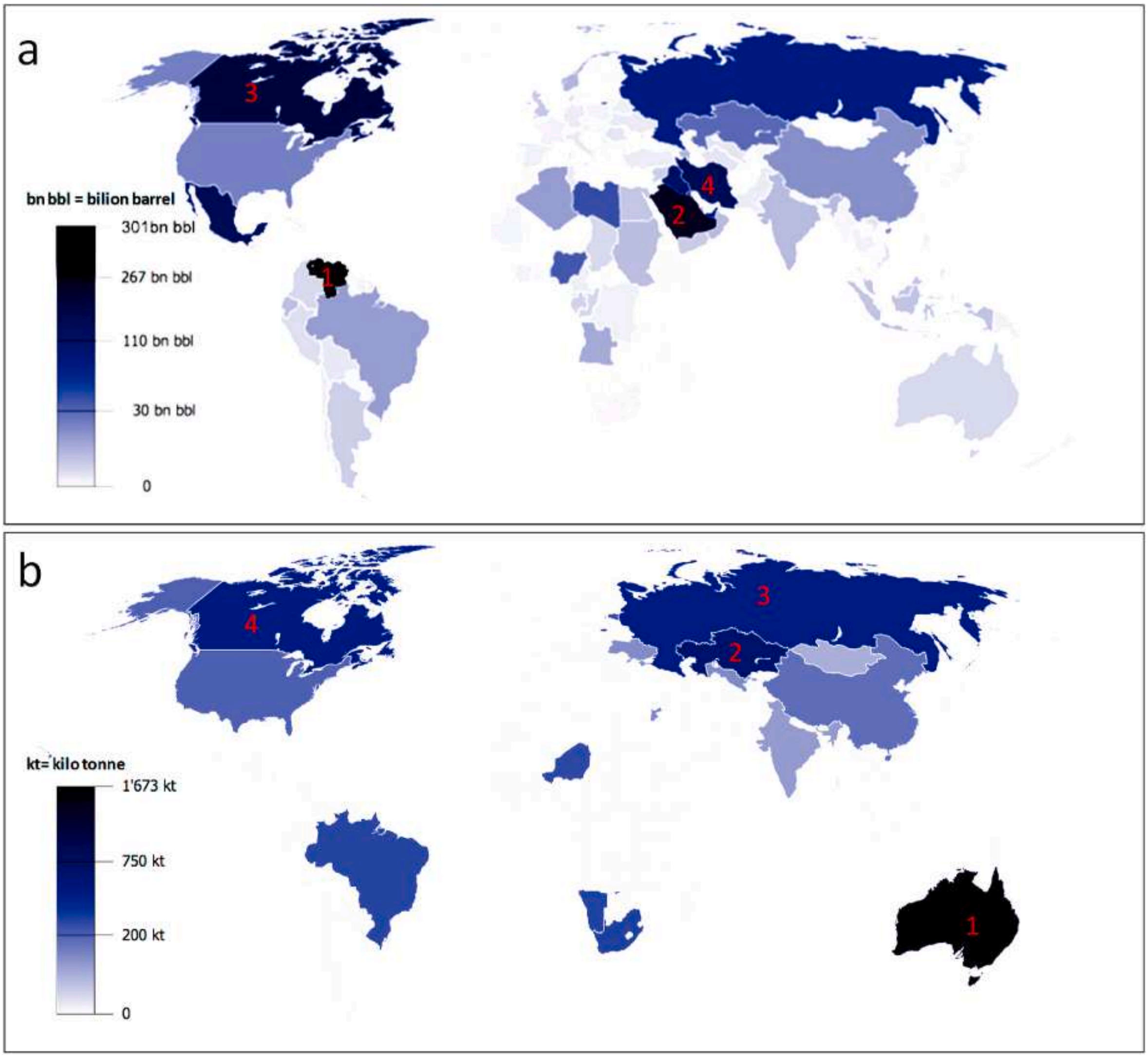


Fig. 4. a. Global oil reserve map (in billion barrel unit). **b.** Global uranium reserve map (in kilo tonne unit). Top four countries are shown with numbers in order of decreasing reserves. **(Available in public domain made by author, Emilfaro, who grants all the right to use this work for any purpose; https://en.wikipedia.org/wiki/List_of_countries_by_uranium_reserves#/media/File:Uranium_Reserves.png)**.

bacteria that reduce sulfate using petroleum as an energy source.

I. This step concerns the close association of U and OM in the interstices between sand grains. Presumably the OM can create a reducing environment by providing electron after the depletion of oxygen from the uranium bearing solution. OM can reduce the uranium concentrated by the OM itself in the earlier stage. Thus the OM play a dual role of concentrating uranium from solution and also of chemically reducing it to insoluble pitchblende and coffinite minerals.

II. This step points out the importance of the bulk OM. After the ore-forming process OM may help in preserving uranium mineralization by physically enclosing the uranium and chemically preventing oxidation. Undoubtedly the OM can be oxidized, but under natural ground water chemistry and flow conditions the time for oxidation may often be longer enough so that deposits as old as~ 130 Ma old can be preserved. When OM is dispersed in black shales, U can be distributed uniformly inside the organo-mineral matrix. In such cases U content is generally <1%. OM of continental origin in organic debris with coal and tree trunks can exhibit higher U enrichment up to 10%. High U concentrations (20%) have also been noticed in some bitumen derived from fluid hydrocarbons, but in these cases barren and mineralized bitumen can coexist in the same deposit. Uranium minerals often take part in competition with other epigenetic minerals like sulphides, carbonates, silicates for filling the cell lumens. Migrated OM is observed to be associated with U in different sedimentary environments (Landais, 1996). Association between coal and hydrocarbon is presented in the Repository Section 2. In fact, few fluvial sandstone bodies are good examples of reservoirs for oil, gas, coal deposits and uranium (and even gold) (de Vries, 1985).

The concept of biogenicity and syngeneity of OM is to be considered (Oehler and Cady, 2014; Goswami et al., 2018). The geochemical analysis of uranium and OM may lead to an understanding of the hydrocarbon (HC) and U association and the maturity of kerogen. Uranium is considered to be an important carbon-free fuel source that preferentially accumulates in the tetravalent state (+4) in anoxic condition. In a redox reaction OM can participate in reducing uranium by losing electron. Thus, uranium reduces and OM oxidizes simultaneously. Therefore, OM can be leached out after oxidation as organic acids are also formed often. The relationship between HC and U therefore may give insights into the past interactions between OM and U. It is important to understand the stage and type of interaction (Table 2) so that the simulation experiment of Mao et al. (2014) is interpreted accordingly.

Table 2
Role of organic matter at different stages of uranium mineralization (modified after Leventhal, 1979).

1. Mobilization Decomposition of OM raises partial pressure of CO_2 in the ground water and soil also adds organic CO_2 and organic acids, which leach and mobilize uranium.
2. Transportation U can be transported as bicarbonate anion or as soluble organic complex in ground water and surface water.
3. Concentration OM with specific functional groups (such as humic acids) perform ion exchange and/or chelate uranium. Concentration factors of >10,000 times have been observed. Humic acids can precipitate at interface of recharge as well as aquifer waters or where pH becomes more acidic or where increased salt content is encountered.
4. Reduction Slow reduction of U, which is held by organic matter as the organic matter decomposes by abiogenic processes
5. Preservation Reduced uranium may be mixed intimately with refractory OM, which is protected from oxidation.

All of the elements of HC system (i.e., source, reservoir and cap rocks and suitable trap formation) must develop over geological time for the system to be viable. With time source rocks in progressively subsiding basins are subjected to increasing pressure and temperature. The geothermal gradient is higher in the Earth's crust than the mantle region. The crustal gradient promotes transformation of OM into hydrocarbon. In this context, the difference between the host rock (reservoir rock) and the host structure (trap structure) must be understood. For HC (including kerogen/bitumen, crude oil, asphalt, natural gas and condensates) sedimentary rocks are the source as well as host rock. Igneous and metamorphic rocks do not contain HC except in some special circumstances when fractured hard rocks act as reservoirs. Therefore, the structures are more important in case of rocks other than sedimentaries. However, for uranium mineralization such a concept does not apply because uranium is a biophile as well as large ion lithophyle element (LILE). U is mostly incompatible in nature and derived from late-stage magmas. However, after coming to the crust they are enriched in certain rocks as per geochemical association and compatibility. Therefore, in case of uranium the term source implies the rock in which uranium content is relatively higher than the natural crustal abundance and from which uranium can be removed easily.

Like HCs, uranium can also be liberated and migrated through the proper pathway and then remobilized and concentrated/enriched at suitable areas. For uranium redox reactions are very much important because uranium has mainly two valence states (i.e., +4 and + 6). Naturally uranium precipitates in reduced form as primary uranium ore minerals (e.g., pitchblende, uraninite and coffinite).

HC generating simulation experiments (Mao et al., 2014) showed that U can play role in enhancing the yield of gaseous HC, in promoting the total gas output, and also in increasing the total HC production. One of the oldest (~2.0 Ga) sedimentary rock hosted uranium ore deposits is located in western Africa, Francevillian Series (Gabon). Here uranium in sandstone is associated with migrated OM, which occurs as secondary porosity infillings (Cortial et al., 1990; Lafaye and Weber, 1993). Cabon provides an example of uranium ore deposits in sandstone reservoirs with HC traps, capped by impermeable black shales.

Mao et al. (2014) referred a simulation of hydrocarbon generation to show how U promotes gas output. Low-mature hydrocarbon source rock containing kerogen type III was the starting material on which UO_2CO_3 solution was added. Uranium enhanced the gas yield and augmented the total gas output.

4. Range of geologic settings of uranium and HC deposits (Fig. 4)

Uranium mineralization with solid bitumen, the altered residues of crude oils, is observed in Precambrian, Cambrian, Permian, Triassic, and Tertiary sedimentary rocks. However, broadly ~70% oil deposits are formed in Mesozoic (252 - 66 Ma), ~20% in Cenozoic (65 Ma), and ~10% in Paleozoic (541 - 252 Ma) (Internet ref-1). As far as Precambrian Era is concerned, at Elliot Lake and Blind River, stratiform type uraniferous kerogen layers of the Matinenda Formation (i.e., black argillite) occur along with eukaryotic algae, which became significant kerogen and pyrobitumen precursors. The Early Proterozoic Pechenga Series, Kola Peninsula, Russia, and McArthur Group, Northern Territory, Australia are examples for dry gas accumulation. Rift-related tectonic settings for oil are observed in Franceville Basin, Gabon (2 Ga), western Africa, Pine Creek Geosyncline (1.8–2.2 Ga) and McArthur Basin (1.6–1.8 Ga), northern Australia (McKirdy and Imbus, 1992). World's oldest commercial oil and gas reserves occur in Siberian Platform (Craig et al., 2013) and Arabian Shield area of late Riphean and Vendian ages respectively. Similarly, ~1.4-Ga Roper Group, McArthur Basin and the ~1.1 Ga Oronto Group are amongst the oldest sediments being explored for HC resources.

Since 2009 the International Atomic Energy Agency (IAEA) has been reviewing the existing classification schemes for uranium deposits to standardize classification. IAEA classification of uranium deposits in 2013 proposed the definition of uranium deposits for the Organization for Economic Co-operation and Development (OECD) as "a mass of naturally occurring mineral assemblages from which uranium has been or could be exploited at present or in the future" (OECD, 2014, 2017). A total of 15 types of deposits have been recognized in this new IAEA classification scheme, which covers ~1807 deposits (UDEPO database), and >40 subtypes/classes (IAEA tecdoc-1842, 2018) (Repository Table 1).

The prime criteria of the classification scheme are based on five factors: *I.* host rock (types 1, 9, 10, 12, 13, 14 and 15). *II.* structure (types 3, 7 and 8). *III.* both host rock and structure (types 2, 4 and 6). *IV.* metasomatic alteration (type 5) and *V.* Surficial process (type 11).

However, for HC deposits the concept of trap, which mostly form in permeable portions of rocks, is most significant. A porous and permeable reservoir rock and impermeable cap rock association are required factors in forming structural or stratigraphic traps. Different types of traps (viz, fault traps, pinch-out traps, anticlinal traps, unconformity traps) form whenever a permeable layer is capped by an impermeable layer. The database from different sources viz.,Organization of the Petroleum Exporting Countries (OPEC), World Factbook of Central Intelligence Agency (CIA) give overall ideas on global HC reserves. There are different classification scheme of petroleum systems based on the complexity of the overlying rock, reservoir lithology, kerogen type, features of HC charging, migration and entrapment, single-sourced or multiple-sourced systems and reservoir qualities (Magoon, 1912; Demaison and Huizinga, 1991; Magoon and Dow, 1994; Zhao and Al-aasm, 2012; Zhao et al., 2019).

A visual evaluation is essential to compare the global reserve of oil and uranium (Fig. 5a and b). From the map it is clear that the top four oil reserves are in Venezuela (20%), Saudi Arabia (18%), Canada (13%) and Iran (9%). On the other hand, the top four uranium reserves are in Australia (31%), Kazakhstan (12%), Russia (9%) and Canada (~9%). It is important to note that Precambrian HC is less important than Phanerozoic. Similarly, metallogenic occurrences of uranium follows certain time bounds. At the beginning during cratonization only magmatic processes were operational in an anoxic atmosphere with surface-related exogenic conditions like physical weathering, hence *quartz pebble conglomerate* (QPC) type uranium deposits were only formed. Before ~2.2 Ga uranium deposits are less due to anoxic condition, which restricts remobilization process by redox reaction (Dahlkamp, 1993).

However, after onset of great oxidation event during the Early Proterozoic, besides physical/mechanical enrichment process, chemical processes also become operational. After a rapid spread of marine microorganisms around 2.2 Ga, generation of photosynthetic oxygen lead to the activation of chemical processes and convert +4 uranium into +6 state to dissolve and transport in solution. The transported uranium into

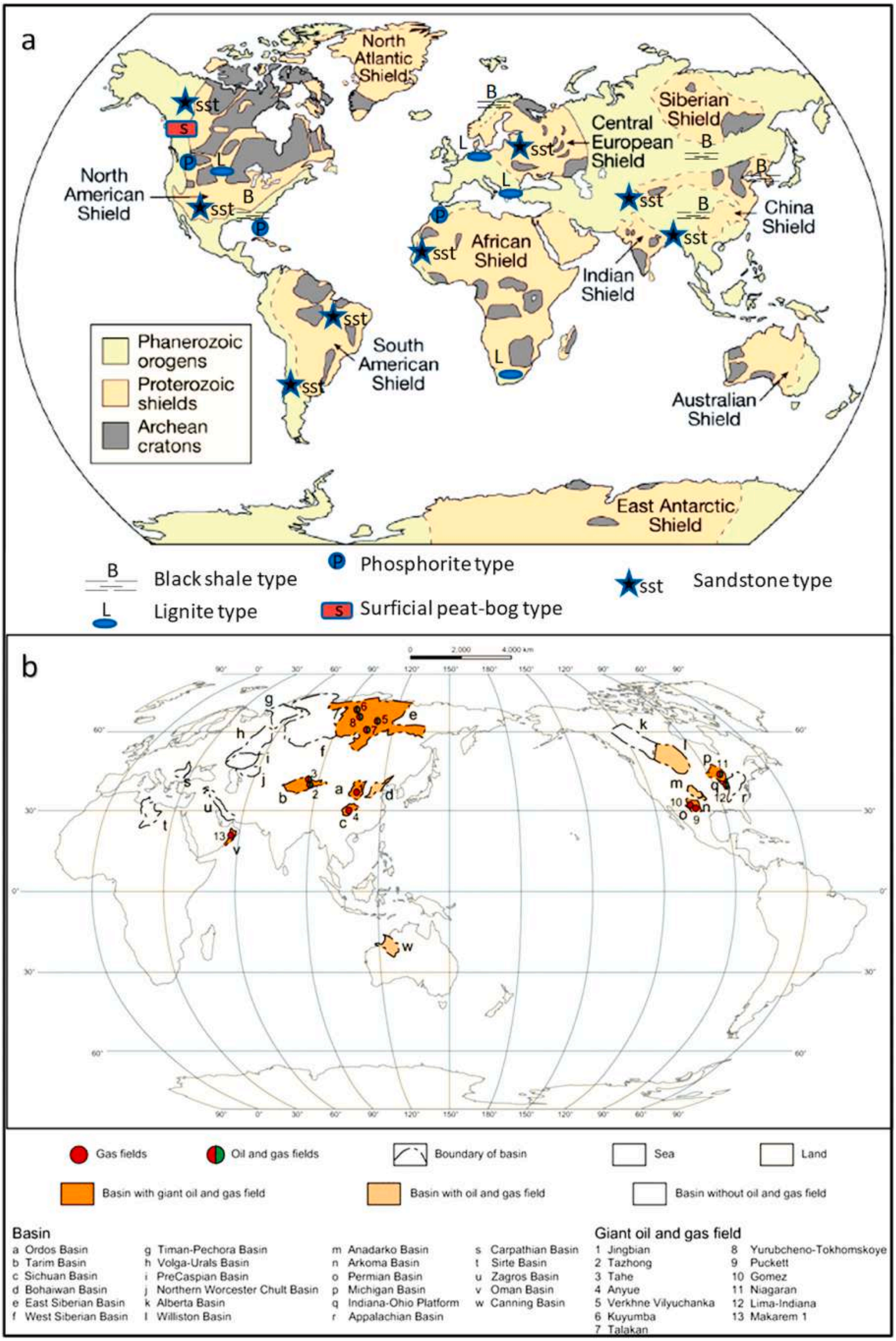


Fig. 5. Important areas of OM and U association around the world. a. few important U-OM association b. major oil and gas field **(after Liu et al., 2017).**

the shallow water basins and accumulated along with carbonaceous pelite, psammite, and carbonate sediments where marine microorganism (algae) generated reducing conditions. It is also noted that such uraniferous sediments form either Proterozoic unconformity type deposit or acted as source for subsequent enrichments in younger rocks. Further, Cambrian to Silurian (~500-400 Ma) time is characterized by uranium accumulation in euxenic basins along with debris of newly appeared land plants, which form low grade uraniferous biack shale (Dahlkamp, 1993. IAEA, 2012a,b, 2013a,b).

Therefore, age-specific occurrences are observed due to coincidence of geologic processes favourable for the proper combination of uranium sources, structures to serve as conduits for fluid flow, suitable redox reactions, preservation potential factors, long-term changes in element abundances, global heat flow patterns, tectonic history, compositions of the atmosphere and ocean and biologic activity.

The OM can either adsorb uranium by physical trapping or can act as electron donor to reduce and precipitate uranium. The chemical process is more significant and commonly observed phenomenon (Goswami et al., 2017a, 2018). Oil and gas structures play significant role in uranium enrichment and producing oil and gas field show about a million tons of uranium ore (Russell, 1958) at several places (e.g., Salt dome, Texas, Poison Basin and Gas Hills district, Wyoming, Brown's Park formation near Maybell and Morrison Colorado, Ambrosia Lake district near Grants, New Mexico, Inter- River area and Circle Cliffs, Utah). The H_2S content in the natural gas and in dissolved state in oil-water phases is considered as an important factor in formation of uranium deposits (Russell, 1958). In OM rich sandstone U can be reduced by direct catalytic effects of bacteria, which can act as electron donor. Biogenic H_2S production is also an important process in uranium metallogeny (Landais et al., 1987; Landais, 1996; Spirakis, 1996). Now, among different types of uranium mineralization, presence of OM can be directly related to 5 types, i.e., sandstone, surficial, phosphorite, lignite and black shale type (Repository Table 1). Apart from these situations, OM can play indirect role in other types as well. Fig. 6a and b exemplify association between OM and uranium mineralization globally.

5. Indian context

Role of HC in sandstone hosted U-mineralization is well manifested in different parts of the world (e.g., Kazakhstan, Canada, Australia, Russia, USA, China and Africa) and an analogy (Reynolds and Goldhaber, 1978; Aubakirov, 1998; Fyodorov, 1999; Huang et al., 2005; Jaireth et al., 2008) is possible for India, especially in those relatively less explored basins with the presence of potential HCs.

A total of 26 Indian sedimentary basins of different age ranges and geological settings are divided into 4 major categories (i.e., category I to IV) from HC potential viewpoint (Dwivedi, 2016; Shaw and Mukherjee, 2022). Category I basins (seven basins: Assam Shelf, Assam Arakan Fold Belt, Cambay, Cauvery, Krishna-Godavari (KG), Mumbai offshore and Rajasthan) have established commercial production. Category II basins (three basins:Kutch, Mahanadiand Andaman-Nicobar) have known HC accumulations but commercial production has not started yet. The basins of Category III (seven basins including Himalayan Foreland, Ganga, Vindhyan, Saurashtra, Kerala-Konkan, Lakshadweep and Bengal) have indicated HCs and are geologically prospective. However, category IV basins (nine examples:Karewa, Spiti-Zanskar, Satpura-South Rewa, Damodar, Narmada, Decan Synecline, Cuddapah,Bhima-Kaladgi, Pranhita-Godavari, Bastar, Chhattisgarh) have uncertain potential. But, they may be prospective in the future by analogy with similar basins. Deep-water basins beyond the Indian east and west costs may also be of interest in future HC exploration. However, according to PRMS (Petroleum Resources Management System, DGH, 2017–18 report), basins are divided simply into three categories (1: Reserves to be produced, 2: Contingent resources to be monetized, and 3: Prospective resources to be explored).

Around 75% of Indian uranium resources are found in Proterozoic host rocks and the remainder occurs in Phanerozoic rocks. Indian uranium deposits are mostly of low-grade and altogether account for ~3% of the world resources (Chaki et al., 2011). Indian uranium deposits are mainly distributed in the following six major provinces:

1. Singhbhum Shear Zone, Jharkhand; 2. in parts of Chhattisgarh; 3. Southern parts of Meghalaya; 4. Cuddapah Basin, Andhra Pradesh; 5. in parts of Karnataka and 6. Aravalli- and Delhi Supergroups, Rajasthan and Haryana. A map (Fig. 6) is shown to provide a better understanding of mainland as well as the sea within the Indian territory. Disposition of HC and U provinces together along with all basins of different categories and agesare shown.

Several scientific officers from the Atomic Minerals Directorate for

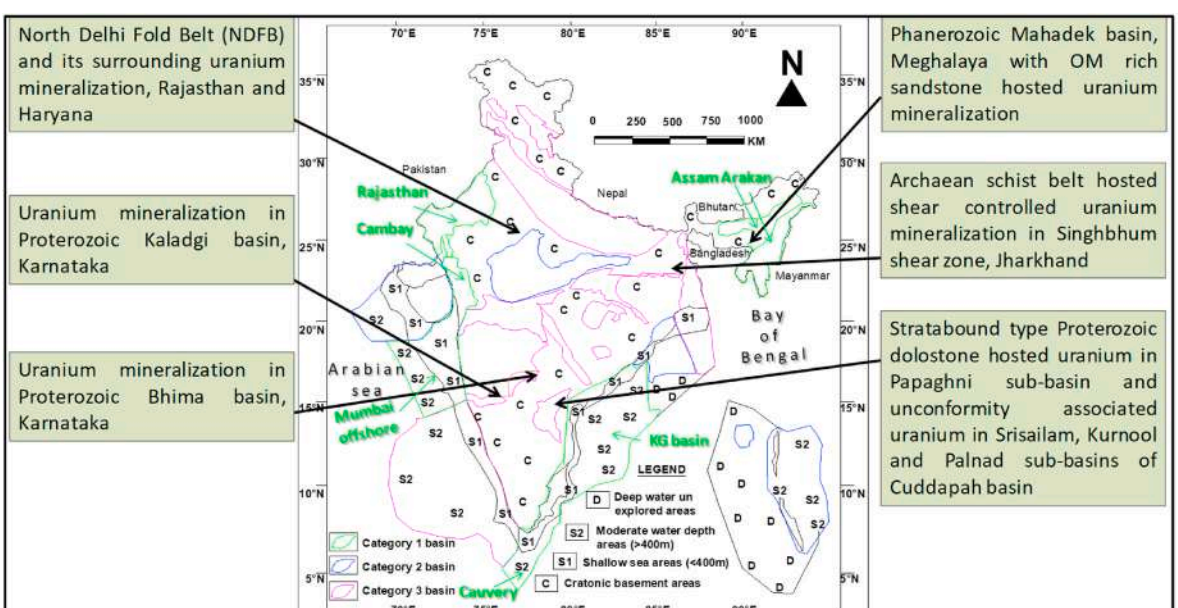


Fig. 6. Indian sedimentary basins (both in mainland India and modern ocean basin) of different category as per HC potential and main uranium provinces (marked by arrows). **(Source: Published AMD reports from Exploration and Research for Atomic Minerals (EARFAM) journal and Directorate General of Hydrocarbons, 2017–18 reports).**

Exploration and Research (AMD), India reported as the significance of organic/carbonaceous matter in uranium enrichment from sandstone of Mahadek, Shillong basin (e.g., Sen et al., 2002; Mahendrakumar et al., 2008; SinhaPadhi et al., 2010; Chopra et al., 2015; Bhattacharjee et al., 2017), where the Upper Cretaceous sandstones of the Lower Mahadek Formation comprises of a uranium deposit. Further occurrences are also reported fromthe Middle and the Upper Siwalik sandstones (Swarnkar et al., 2002; Kumar et al., 2010; Kothari et al., 2011) of Miocene to Pleistocene age, in which the concentration of uranium is controlled by the redox interface, porosity-permeability barriers and abundance of reductants such as organic carbon, pyrite, anaerobic bacteria and also even vertebrate fossils from Middle Siwalik in Hoshiarpur district, Punjab. In fact, uranium is concentrated in Haversian lamellae part of vertebrata, while the Haversian canal do not show any uranium. This is because calcium phosphate, acts as the key carrier of uranium (Kumar et al., 2010).

Uranium has been reported from the petroliferous Cambay basin (Gujarat, India) withinthe Andimedan Formation (408 ppm), Sattapadi shale and the Bhubangiri Formation (8–10 ppm) (Nabmier and Giridhar, 2008).

Further, occurrence of uranium was also reported in Gondwana rocks (Gupta and Sharma, 2013). Conglomerate near Allapakonta and Vembakam, Chittoor District, Andhra Pradesh showed uranium occurrence at the base of Satyavedu Formation of the Upper Gondwana sediments of Palar basin (Sharma et al., 2016). Another interesting study revealed about tectonolithologic control of uranium mineralization in Triassic Denwa Formation of Upper Gondwana sequence in the Satpura Gondwana Basin (Ranjan et al., 2010).

However, the role of OM in uranium mineralization is evidenced in the Papaghni sub-basin, Cuddapah basin, where microbial mat and stromatolites play a significant role (Sharma and Shukla, 1998, Goswami, 2015; 2016, 2017a,b, 2018). Apart from the Proterozoic Kurnool (Koppunuru), Srisailam, Bhima and Kaladgi also showed evidences of past organic activities (Latha et al., 2011; Patnaik et al., 2016, AMD report, 2020) along with Aravalli-Delhi, Chattisgarh, Vindhyan, and Krol-Tal (Banerjee et al., 1992). Moreover, the observed uranium anomalies in the Abujhmar basin (Chaturvedi et al., 2006), suspected OM in black shale in Bijawar basin (Bandyopadhyay et al., 2016), Kushalgarh Formation, Delhi Supergroup (Mandal et al., 1984; Singh et al., 2019) and the chemical sediments of Gwalior Group (Absar et al., 2010) require special attention along with other Precambrian basins. In fact the Mesoproterozoic intracratonic Abujhmar basin at the NW end of the Bastar Craton shows uranium occurrences in the Gundul Formation of Abujhmar Group. The spread of uranium anomalies all over the basin is significant, but the controlling factors have remained indeterminate (Chaturvedi et al., 2006). On the other hand, the Paleoproterozoic Gwalior Group of the Bundelkhand Craton showed Pb–Pb age of 1866 ± 250 Ma for the BIFs, which suggest the terminal stage of global Palaeoproterozoic BIF development (Absar et al., 2010) and the Hudsonian Orogeny (Goswami et al., 2019). The Paleoproterozoic Bijawar Group, sandwiched between Archean-Paleoproterozoic Bundelkhand Granite Gneiss Complex (BGC) and Mesoproterozoic Vindhyan Supergroup, showed evidences of carbonaceous interbands in the Bajna dolomite (Bandyopadhyay et al., 2016) with high concentrations of Cu (up to 1366 ppm) and the *total organic carbon* (TOC) from 47% to 91%. Overall a comprehensive summary can be found in a palaeobiological review article on Proterozoic and Cambrian successions of India after Sharma et al. (2016). The well defined seven categories of biological evidence (viz. MISS: Microbially Induced Sedimentary Structure) and stromatolites, acritarchs, OWM (Organic Walled Microfossils), carbonaceous remains, trace-fossils and Ediacaran fossil evidences, stable isotopic evidences and organic geochemical evidences} are discussed along with the present status on unsolved problems and future research scopes by Sharma et al. (2016).

6. Tectonics vis-a-vis U and HC

As far as the structural deformation is concerned, fault and anticlinal fold hinges act as most common and suitable trap for HC (Chapman, 1973). However, for uranium mineralization folds are less common host structure than the fracture and fault zones, which are significant. Therefore, the importance of folding, fracturing/faulting and associated tectonics and orogeny are needed to conceptualize.

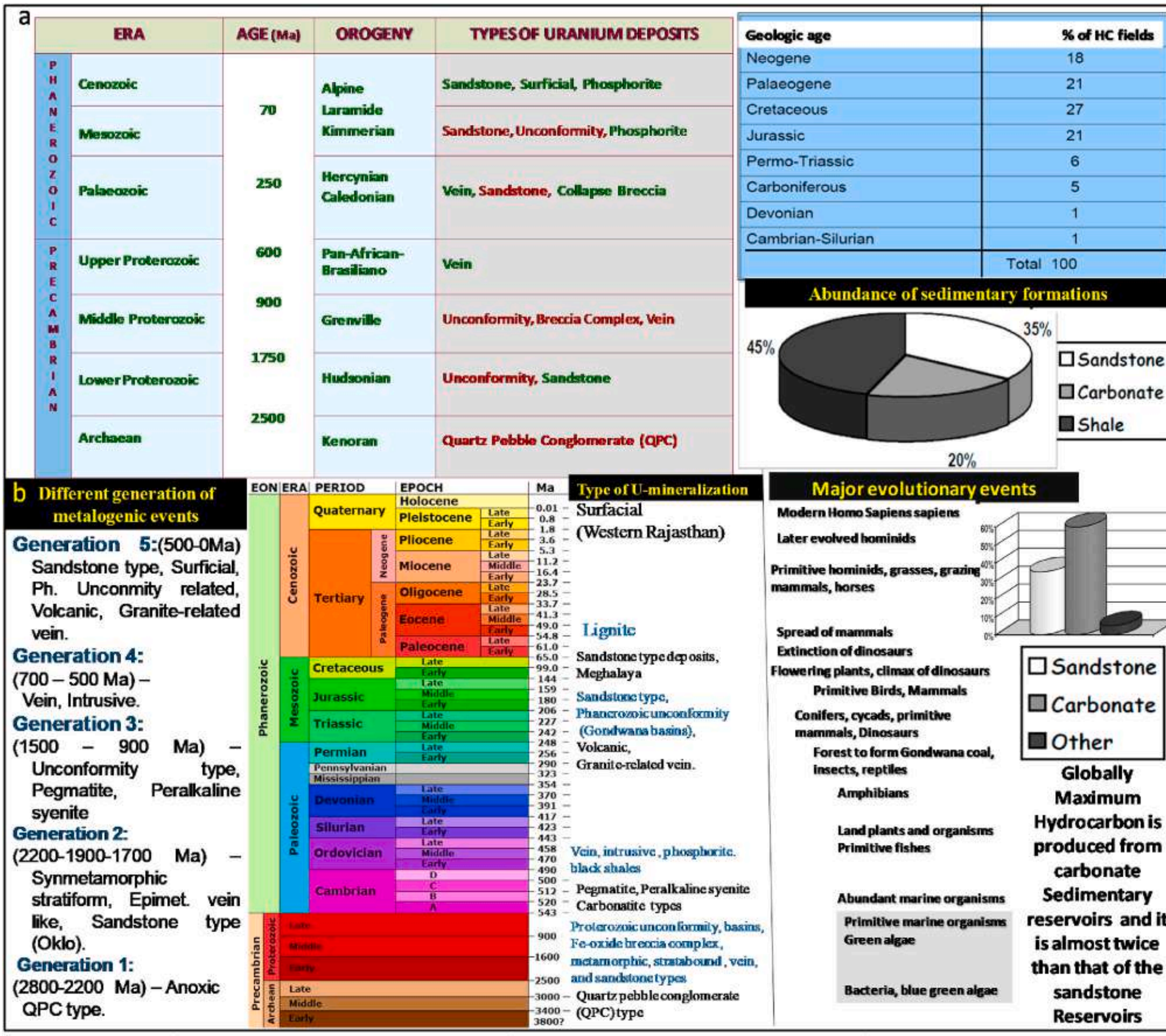
There are several orogenic events in Earth history, which accelerated the enrichment mechanism of uranium as well as HCs (Fig. 7a–c) by forming suitable traps. Although the Earth is ~4.6 Ga old, commercial quantities of HCs are usually found in rocks not older than half a billion years (Gluyas and Swarbrick, 2004). The oldest live oil recovered to date is sourced from Mesoproterozoic rocks within the Velkerri Formation (Roper Group) of the McArthur Basin of northern Australia, where the initial oil generation and migration happened before 1280 Ma (Craig et al., 2013). Thus, Precambrian HC fields are mostly migration-related deposits. The geologic age of reservoir rocks must be known because rocks of different ages exhibit different petroleum characteristics and productivity.

Before 2.2 Ga protocrust development, cratonization, granitization and absence of free atmospheric oxygen characterized the Earth as a chemically inactive stage for uranium enrichment. Only few mechanical/physical processes (develop quartz pebble conglomerate, QPC type uranium mineralization) were operational in presence of prokaryotic cyanobacteria/blue-green algae (no HC possibility). After the great oxidation event ~2.2 Ga, chemical process was so active that huge remobilization of uranium (after conversion from +4 to + 6 valence state) took place. Shallow water Proterozoic basin development and marine microorganism flourished also supporting uranium deposition. The algae generated a reducing environment that supported uranium precipitation after enrichment at suitable places e.g., unconformity, reduced sandstone, geosynclinal phosphorites. Subsequently these enriched precipitates also acted as source for further enrichment in youngerrocks of suitable geotectonic settings with magmatic-anatectic and metamorphic processes. After 0.5 Ga, the appearance of land plants and euxenic basins were significant events for both U and HC. Therefore, restrictions in the distribution of U and HC to specific epochs in the Earth's history are related to the tectonic evolution.

Uranium is found in five principle generation controlled by time stratigraphic parameters and orogenic events. However the majority of the Phanerozoic orogeny (e.g., Pan African, Hercynian-Caledonian, Alpine-Laramide-Kimmerian) were supportive of exogenic processes to generate the U and HC association especially after Permo-Triassic mass extinction event.

The OM-rich black shale is the best source rock for HC and is more abundant than sandstone and limestone. Generally speaking, limestone is more common than sandstone as a reservoir rock. Therefore, role of fracturing is essential in creating permeability and forming suitable host. Permeability act as pathway for HC and uraniferous fluids. The concepts of biogenicity (origin of an organic remnant in a host rock from a life form) and syngeneity (relative age of the organic residue compared to the age of the host rock) are much more sensitive under such conditions where tectonics can play role in mobilizing organic matter and thus allochthonous OM and U association and deformation can often create ambiguity. Therefore, even in deformed fractured igneous rocks (e.g., granite) OM-U association, HC reservoir can be expected under certain conditions.

Plate tectonics often plays a role in the subsidence needed to form a basin in which sediments may accumulate to form stratigraphic successions and in creating hydraulic pathways for enrichment of migrating uranium from source to host rock/structures. In this context the '4 P factors' (Goswami et al., 2017a, 2018) are of immense significance. Provenance, porosity-permeability, precipitation and preservation are the main factors controlling uranium mineralization in sandstone aquifers, with impermeable cap/seal rocks above and below along with



C Indian HC basins	Source rock	Reservoir rock	Cap rock
Assam-Arakan	Paleocene – Lower Eocene, Cabonaceous Shale of Barails (Oligocene)	Oligocene Barail and Miocene Tipam Siltstone	Kopili shale (upper Eocene)
Mumbai Offshore	Paleocene to early Eocene Panna Formation	Eocene to Oligocene Bassein Limestone.	Late Middle Miocene Shales
Cambay	Oligocene Cambay shale, Kalol & Anklesvar Formations (Palaeocene to early Eocene)	Middle Eocene Kalol Siltstone, Paleocene Olpad siltstone	Upper Eocene –Oligocene Tarapur Shales, Telwa & Kanwa Shales (Middle Eocene Oligocene)
Cauvery	Cretaceous Sattapadi Shale, Kudavasal Shale of Paleocene Kamalapuram Formation.	Precambrian basement, Oligocene multi-stacked Siltstone	Post unconformity Shales, Kamlapuram
Krishna-Godavari	Golapalli Formation and Gajulapadu Shales (Permo Triassic to Pliocene)	Permain to Pliocene Clastics with few local Carbonates and volcanic reservoir	Red clays (lower Gondwana), Raghavpuram Shale and basaltic trap (late Jurassic to Upper Cretaceous)
Rajasthan	Cambay & Tharad Shale of Barmer- Sanchor sub basin (Permian to Cretaceous) and Palaeozoic limestone	Barmer & Tharad Siltstone, Carbonate reservoirs in Cambrian to Palaeocene sequences	Intraformational Shale and Tight Lime stones

Fig. 7. a. Time-bound geochronologic-stratigraphic and orogeny-related concentration of uranium and HC fields with abundances of HC fields and natural abundance percentages of 3 major types of geological sedimentary formations. **b.** geological time scale and evolution of global and Indian uranium metallogeny vis-a-vis organic evolution with a chart on relative abundance of HC reservoir rocks**. c.** Indian HC basins and source, reservoir and cap rock properties. (Based on this review, compilation from generalized information in several text books, and reports from Directorate General of Hydrocarbons and Dahlkamp, 1993).

the facility of migration of HC fluids into the uraniferous aquifer through reactivation structures (Fig. 8a).

The role of HCs in the uranium mineralization process is not the sole factor acting to reducing basinal brine or as a reductant in precipitating U, but also as a chemically active HC-bearing fluid during diagenesis of host sediments. HCs often have a direct role in the origin of the U ore phase. Therefore, suitable host structures (Fig. 8b–e) can give pathway of fluid migration to suitable localities but the valid question raised is what kind of fluids are involved in uranium mineralization and how can the origin be constrained (Cao et al., 2016). To address another important point regarding the direct role of HCs in the uranium mineralization based on fluid inclusions, sulfur isotope data along with established HC events during the diagenesis of host sandstones. The review after Cao et al. (2016) revealed coupled bacterial uranium mineralization and HC oxidation that may be followed by later recrystallization of ore phases. This can happen in association with epithermal-mesothermal hydrothermal solutions under HC induced reducing conditions. Thereby HC migration (allochthonous) along with fluids, episodic faulting, diagenetic alterations (e.g., pyritization, chloritization, calcification, silicification, kaolinitization etc) and role of

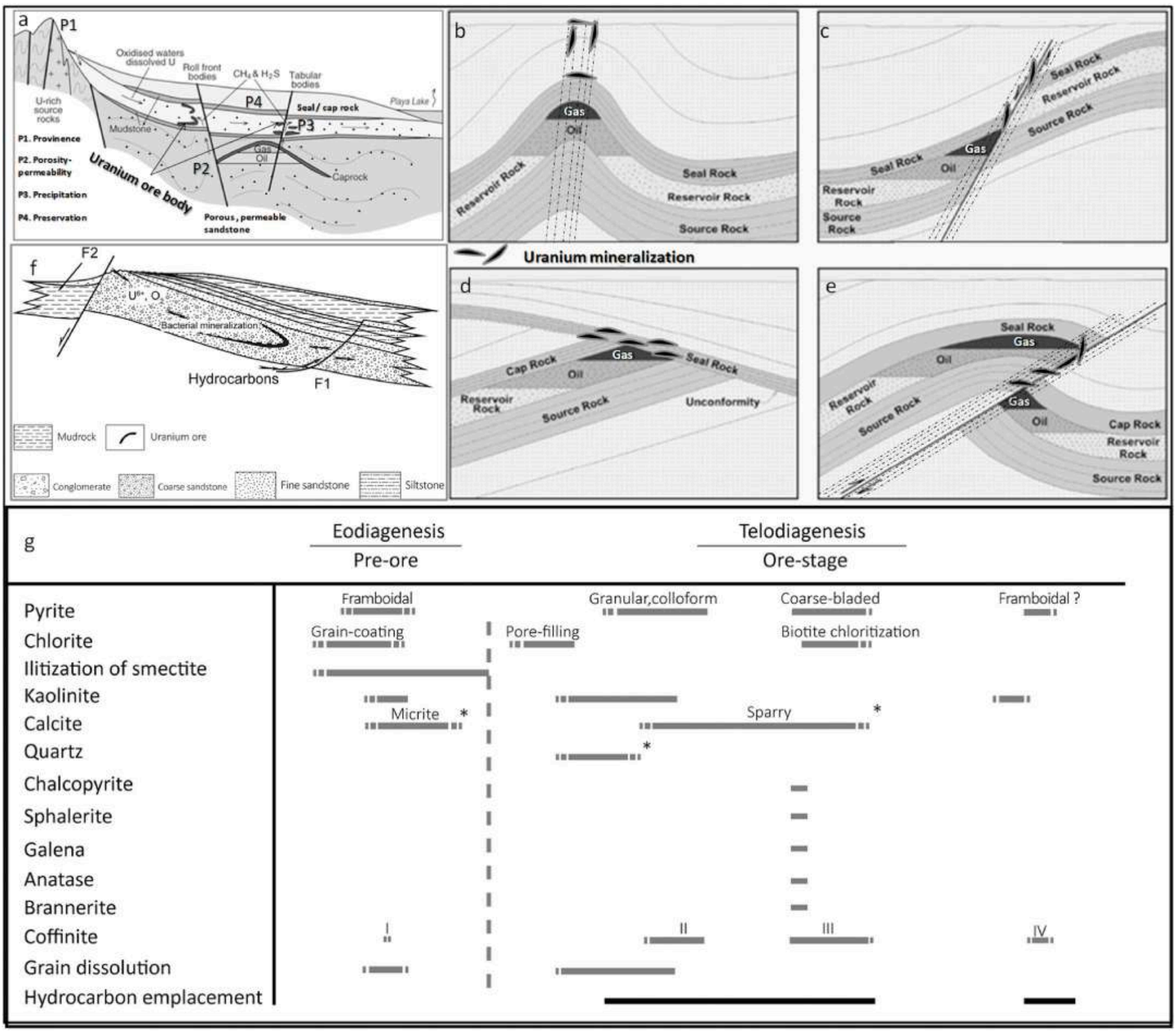


Fig. 8. a. Simplified diagram showing different components of HC-U system (modified after Jaireth et al., 2008). The uranium anomalous granitoid provinance (subjected to chemical and physical weathering), uraniferous sediment and solution transportation and deposition into the basin, post-depositional diagenesis, autochthonous OM, basinal bacteria, major HC generation and migration event along with reactivation of deep faults. **b-e.** different structures and settings of trap associated with tectonothermal events. Initiation and propagation of multiple fractures along fold hinges, older normal faults, reverse faults, unconformity, resulting in HC upwelling from deeper depths along nearby structures due to tectonic pumping. **f.** geological formation sequence of conglomerate, sandstone, siltstone, shale and faults of different generation suggest downward percolation of oxygenated U-bearing groundwater into the host, gravity driven vertical movement of scavenged uranium from upper horizons, migration of deep seated HCs to the upper unit and bleached zone with red beds near local nose structures (typical of roll front deposit). Continuous seepage of HCs from nearby faults into the highly porous and permeable host sandstone cause reduction zone development to facilitate U precipitation (after Cao et al., 2016). **g.** Diagenetic sequence of major authigenic minerals in case of sandstone hosted U-HC association. The asterisk marks (*) suggest the minerals with available fluid inclusions (after **Cao et al., 2016**). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

HCs in reduction and precipitation of U are well described (Fig. 8f and g).

Uranium-rich marine black shale with a wide geographic distribution from around Norway to Estonia (Alum Shale Formation, Cambrian and Early Ordovician) exhibits a typical signature of thermal maturation that restricted fluid migration and remobilization of uranium in southern Sweden. However effects of the Caledonian orogeny is prominent in northern Sweden, where U, P, and Ti were mobile phases and precipitated as phospho-silicates U–Si–Ca–P (±Ti ± Zr ± Y) and minor amounts of uraninite (Lecomte et al., 2017).

7. Discussions

Uranium is a biophile element, which often tend to be associated with HCs and only physico-chemical condition is important in secondary surficial condition. Secondary geochemical dispersion mechanism is significant without any involvements of endogenic processes. The source of radon in crude oil is considered to be the disintegration of uranium (Levorson, 1967). The presence of helium in a hydrocarbon trap cannot be interpreted in a straightforward way for several reasons. *(i)* The helium content of the basin depends on the age and the geologic history of the basin (Nabmier and Giridhar, 2008). Since helium invariably leaks from traps, > 2% of helium in trap is unlikely (Nabmier and Giridhar, 2008). *(ii)* Helium can be produced by radioactive decay of U and Th, and therefore inorganic in origin. The inorganic gases also contain few inert gases, N_2, H_2S and CO_2 (Kinghorn, 1983). *(iii)*Deep seated basement rocks, such as granites, can be the source of helium. Expulsion of helium can be augmented by deformation or thermal activity of the basement. *(iv)* The relationship between radioactivity and the helium released from the basement is obscure because helium is found always in deep wells and the basement rock's property is not always well known (Selley, 1985).

Coal, oil, gas and uranium are the main energy supplying materials. Among these, the first three resources exhibit dominant occurrence in sedimentary basins. Uranium shows a more diverse occurrence. But, out of ~1880 known uranium deposits of the world, ~900 are sediment, especially sandstone accounting for ~50% of the total uranium deposits. This sandstone-hosted uranium mineralization occurrences are spread over ~110 sedimentary basins worldwide (IAEA, 2018). According to Feifei et al. (2017), coexistence of sandstone type uranium are identified with either oil and gas fields or coal fields in about 85 basins. Thus statistically ~75% of all uranium producing basins exhibit oil, gas or coal accumulation. Therefore, the identification of such multi-energy producing sedimentary basins must be of utmost target for the future. Note that25°Ñ 50°N in the northern hemisphere shows major distribution of such multi-energy basins in the east-central Asia and the western USA (Feifei et al., 2017).

In a broader sense, radioactive mineral's presence on the Earth's crust can have a "direct influence" on generating HCs (Levorsen, 1967). Since pure quartz arenite and limestone is practically devoid of radioactivity, such rocks naturally cannot be related to oil generation by radioactive disintegration. In contrast, (black) shales usually are characterized by a high radioactivity, for example bituminous shale has $1*10^{-3}$ to $7*10^{-3}$ ppm of uranium (Levorsen, 1967). Because in sandstones the radioactivity is quite variable spatially, no generalized comment of this sort can be made for this rock type. In fact, radioactivity is not a universally agreed mechanism for oil generation. This is because of the two striking observation worldwide: *(i)* an oil field can be devoid of radioactivity, and *(ii)* a radioactive terrain can be devoid of oil (Landes, 1951).

8. Conclusions

Organic matter (OM) can be associated with uranium (U) in several ways since the latter shows a biophile tendency. Therefore co-occurrence of OM and U needs focused study from the proved and probable reserves worldwide. Sedimentary basins consisting of sandstones, black shale, peat-bog, lignite etc. are important from this perspective.

Author contribution

SM did overall correction, literature enrichment and article finalization. SG did overall MS conceptualization, writing, proof correction, literature survey on U and HC association and article shaping. SZ worked mainly on the uranium part.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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