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BIOLEACHING - AN ALTERNATE URANIUM ORE PROCESSING TECHNOLOGY FOR INDIA

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Abstract

Meeting the feed supply of uranium fuel in the present and planned nuclear reactors calls for huge demand of uranium, which at the current rate of production, shows a mismatch. The processing methods at UCIL (DAE) needs to be modified/ changed or re-looked into because of its very suitability in near future for low-index raw materials which are either unmined or stacked around if mined. There is practically no way to process tailings with still some values. Efforts were made to utilize such resources (low-index ore of Turamdih mines, containing 0.03% U_3O_8) by NML in association with UCIL as a national endeavor. In this area, the R&D work showed the successful development of a bioleaching process from bench scale to lab scale columns and then finally to the India's first ever large scale column, from the view point of harnessing such a processing technology as an alternative for the uranium industry and nuclear sector in the country. The efforts culminated into the successful operation of large scale trials at the 2ton level column uranium bioleaching that was carried out at the site of UCIL, Jaduguda yielding a maximum recovery of 69% in 60 days. This achievement is expected to pave the way for scaling up the activity to a 100T or even more heap bioleaching trials for realization of this technology, which needs to be carried out with the support of the nuclear sector in the country keeping in mind the national interest.

Introduction

The continued depletion of high grade ores and growing awareness of environmental problems associated with the traditional methods have provided impetus to explore simple, efficient and less polluting biological methods in uranium mining, processing and waste water treatments [1-2]. Hydrometallurgical methods have some disadvantages such as low recovery, involvement of high process and energy cost and increase in pollution load of water resources [3-5]. Uranium could also be recovered by microorganisms that can catalyze the oxidation of uranium and also associated metals, and hence can influence its mobility in the environment. Industrial-scale bioleaching of uranium is carried out by spraying stope walls with acid mine drainage and the *in-situ* irrigation of fractured underground ore deposits [6]. The recent upsurge of interest in this area is motivated by the fact that it is a simple, effective, potential, and relatively less expensive tool involving low energy consumption and is environmentally benign, primarily due to the uranium solubilising and accumulating properties of certain microorganisms. Besides, its industrial application to raw material supply, microbial leaching has some potential for remediation of mining sites, treatment of waste and detoxification of dump/sludge. Commercial application of bioleaching of uranium from low-grade ores has been practised since the 1960s. The seven leading uranium producing countries in descending order are Canada, Australia, Niger, the Russian Federation, Kazakhstan, Namibia and Uzbekistan. The two largest producers viz. Canada and Australia alone account for over 50% of global production. The presence of microorganisms in leaching operations has been found to be beneficial in catalyzing the uranium dissolution process [5,6]. Currently this technology is applied on a commercial scale not only in the recovery of uranium but also for extraction of copper, nickel, gold etc. through heap, dump and *in-situ* leach techniques [2]

Bioleaching

Depletion of high-grade mineral resources results in tendency for mining of ores/minerals deeper underground. Minerals such as betafite (samiresite), brannerite, and parsonite have lower bio-dissolution characteristics (16-35%) whereas most other minerals displayed moderate (40-55%) to higher level of uranium dissolution (60-88%). The role of microorganisms in mining, ore processing and waste-water treatment is likely to become increasingly important [6]. The modest nutritional requirements of microorganisms are provided by the aeration of an iron- and/or sulfur-containing mineral suspension in water or by the irrigation of a heap [7]. Small quantities of inorganic fertilizer can be added to ensure that nitrogen, phosphate, potassium and trace element limitation does not occur. For e.g., *Acidithiobacillus* group of bacteria utilizes the energy from Fe(III) in acid medium in presence of oxygen at an optimum pH of 1.5-2.5 at ambient temperature to leach 70-98% metal content of the substrate. The optimum factors of nutrients, pH and temperature are very essential to maintain the intended growth and activity of these microbes [7,8].

Mechanism of uranium bioleaching

There has been a long-standing debate as to whether the microbially assisted leaching of uranium ore is by direct or indirect mechanism. In the leaching of uranium, the bacteria do not directly attack the uranium mineral. But they generate Fe(III) from pyrite and soluble Fe(II). Fe(III) readily attacks minerals incorporating U(IV) to converting it to U(VI) which is soluble in dilute sulfuric acid [9]. The bio-oxidation is about 10^5- 10^6 times faster than the chemical oxidation. The uranium solubilisation by indirect mechanism can be described as:

$$FeS_2 + H_2SO_4 \rightarrow 2\,FeSO_4 + H_2O + 2\,S^o \quad \textbf{(1)}$$

$$FeSO_4 + H_2SO_4 + O_2 \rightarrow Fe_2(SO_4)_3 + H_2O \quad \textbf{(2)}$$

The Fe(II) can be re-oxidised by microbes to Fe(III) which takes part in the oxidation process again. Sulfur formed is simultaneously oxidized depending on the species to H_2SO_4 which aids (oxidizing agent) the dissolution of uranium as follows [10]:

$$2S^o + 3O_2 + 2H_2O \rightarrow 2\,H_2SO_4 \quad \textbf{(3)}$$

The insoluble uranium(IV) is oxidised to the water soluble uranium(VI) sulphate as:

$$UO_2 + Fe_2(SO_4)_3 \rightarrow UO_2SO_4 + 2\,FeSO_4 \quad \textbf{(4)}$$

The Fe(III) can be generated by the oxidation of pyrite (FeS_2) which is often associated with the uranium ore [10]. In an acidic solution without the bacteria Fe(II) is stable, and leaching mediated by Fe(III) would be slow. *A. ferrooxidans* can accelerate such an oxidation reaction by a factor of more than a million [8].

Bioleaching of uranium in Indian scenario

With the detection of uranium first ever at Jaduguda (quartz-chlorite-biotite-schist type) in 1951, the first uranium processing plant was commissioned in 1967 at the site. Different mining methods and leaching techniques were studied to find the most suitable alternative keeping the cost and the environmental impact as low as possible [11]. The uraninite in Jaduguda, Bhatin and Narwapahar ores is entirely UO_2 (IV) and UO_3 (VI) types and hence UO_2(IV) remains undissolved or slowly dissolved in absence of iron(III) in the traditional method [11]. Use of oxidative leaching technique (H_2SO_4/MnO_2, $NaClO_3$, $Fe_2(SO_4)_3$ or pyrolusite leaching) is the most important process by which uranium ores are being processed in India. It was reported that at 1 kg/ton ferric sulphate consumption with 1kg/ton acid, the same amount of uranium can be leached as with 20 kg/ton of acid and 4 kg/ton pyrolusite [12]. The sulphate leach liquor (pH 1.0- 2.0) typically carries 0.5-0.6 g/L uranium and other impurities like Fe, Al, V, Cu, Mn etc. in varying concentrations depending on the nature of the mineral and leaching conditions employed.

For a country with limited energy resources and for long-term energy security exploiting microorganisms for bioleaching is an alternative, highly selective, eco-friendly and economically attractive option [2]. The procedures are not complicated and are easy to control and extensive technical knowledge is not required [3-5]. Moreover, the microorganisms used in this processes are able to grow in acidic environment with high metal content like U, Th, Cu, Ni etc. *A.ferrooxidans* was dominant species in the Jaduguda and Turamdih mine waters [12,13]. Indian researchers have reported 79% recovery in 12 h using $9gH_2SO_4$/kg of Jaduguda ore adopting BACFOX (bacterial film oxidation) method [14] containing *A.ferrooxidans*. Recently, 98% and 70% uranium bio-recovery from Turamdih ore was reported from the author's laboratory [15-17] in shake flask and column experiments respectively at 1.7pH as detailed in Table-1.

Table-1: Bio-extraction of uranium from a low grade ore [15-17]

S. No.	Scale of leaching	Conditions	Uranium recovery
1.	Shake flask	1.7pH, 10% PD, 30d, E_{SCE}: 680mV	98%
2.	Small columns-6kg	1.7pH, 60d, E_{SCE}: 635mV	~60%
3.	Columns-80-100kg	1.7pH, 60d, E_{SCE}: 638mV	69%

Column bioleaching for ore containing 0.0308% U_3O_8 was carried out by NML in collaboration with UCIL, in Jaduguda-CRD premises in two columns (1-bioleaching, 1-control leaching) as detailed in the set-up (**Fig.1a**) with ore input of 2.0 T per column {size distribution as in **Fig.1b**} for a period of 60 days. Leaching was carried out at a flow rate of 120 L/h for ~9 h/day at pH 1.7 [17]. The leach samples for U_3O_8 were analyzed at UCIL. An appreciable uranium bio-recovery of 68% was observed against a lower control recovery (53.5%) in 60 days. This technology, being the first ever on this scale in India, must be of great interest for consideration on 10-100T heaps at large scale.

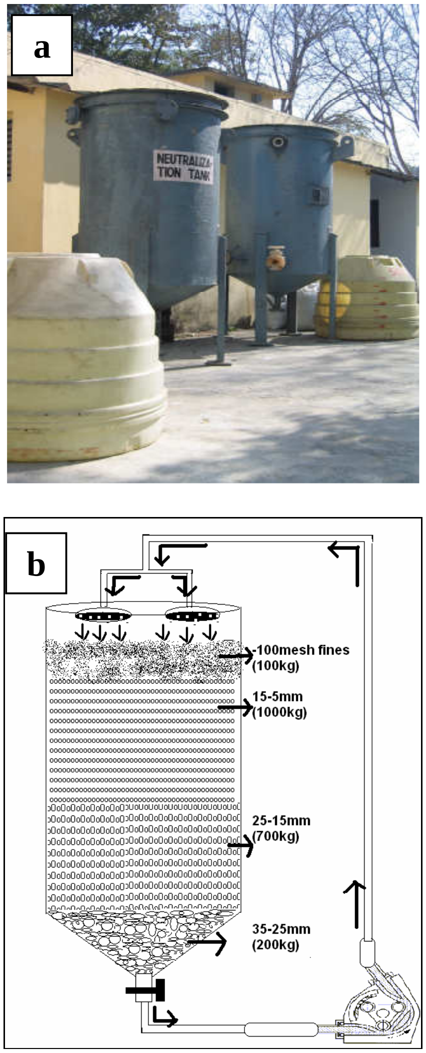

Fig.1: Column set-up (2.0T scale) at UCIL, Jaduguda

Conclusions and Future Applications

At present, bioleaching is being used commercially only for the recovery of copper, uranium and gold. In future, however, these processes will become important for several other metals such as zinc, nickel, cobalt and molybdenum not only for extraction but also for environmental clean-up. Investment and operating costs are much lower than that for conventional hydrometallurgical processes. The application of microorganisms for ore processing and waste remediation is likely to become increasingly important in Indian context in the coming years. This will further be driven by the need to process ores containing small concentration of uranium, the potential for reprocessing waste spoils and tailings, economic constraints, and possible legislative changes on the environmental impact of more traditional approaches such as hydrometallurgy. The use of acidophilic, chemolithotrophic iron- and sulfur-oxidizing microbes mainly mesophiles in processes to recover metals like copper, uranium and gold is now well established. In the immediate future, heap bioleaching is likely to be a major area of expansion while using thermophilic bacteria. Microorganisms that allow the recovery thereby reuse of uranium that are currently being either dumped in oxidized sludge wastes or retained in constructed wetlands, are also likely to be further developed and utilized by sustainable and integrated approach to uranium extraction, resource conservation and safeguarding the global environment. Bioleaching using acidophilic autotrophs has made significant strides in its development as a commercially viable technology for exploiting the low grade uranium ores. Industrially applicable technology for heterotrophic leaching of uranium carbonates, and silicates on an industrial scale, awaits development. In view of its potential as an eco-friendly process than traditional processes, bioleaching promises to replace many traditional extraction processes in not too distant future.

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