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Uranium Mill Tailings Generation and Management Challenges

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Abstract: Uranium mining has been ongoing for the past 200 years, yet managing uranium waste remains a significant challenge. This difficulty arises due to strict regulations imposed on uranium-related industries, driven by the assumed health risks associated with radiation. As a result, much of the waste generated by nuclear industries is currently stored. In India, uranium mining and milling waste is stored that require large tracts of land and continuous monitoring. Given the high value of land in India and the potential expansion of uranium mining to meet net-zero emission targets, an effective solution for managing uranium mill tailings is urgently needed. Meanwhile, India's construction sector is rapidly growing that require a huge quantity of materials. This paper reviewed the history of uranium mining and mill tailings generation and propose that the construction industry could effectively utilize these tailings. This approach not only addresses resource scarcity but also promotes environmental sustainability by conserving natural resources, reducing ecological imbalances, and advancing a circular economy.

Keywords: Uranium, Mining, tailings, Circular Economy, Construction Industry, NORMs.

1. Introduction: The mining industry plays a crucial role in the global economy, holding a key position in the resource supply chain. There is substantial evidence indicating that enhancing mining activities can significantly improve a country's socio-economic conditions. In India, the Atma-Nirbhar Bharat initiative emphasizes innovative and cost-effective strategies to boost domestic mineral production. While the mining and milling processes contribute greatly to development, they also generate large amounts of waste which is dependent on the mineral being extracted, location, ore grade, and composition. The rock to mineral ratio [1] is an important metric that defines the amount of waste generated by a mining industry and is a sum of all the waste (waste rock, tailings and slag) to the final metal produced. The RMR is different for different metals for ex. for gold it is in the range of 10^5-10^6 while for Fe it is around 10. For Indian Uranium industry also RMR is 10^5-10^6.

Presently, energy generation is prime requirement for a self sustainable nation. Earlier, this requirement was fulfilled by the coal but with the known effect of the environmental pollution and green house gas emission by the coal burning, a lot of focus has been shifted towards the use of low carbon emission energy sources such as solar, wind, hydrothermal etc. Among the available resources, solar, wind and nuclear are found to be the best in terms of Indian scenario. Singhal et. al. [2,3] has carried out a detailed analysis on available energy sources in terms of their availability, scalability, raw material availability, waste generation, green house gas emission and cost. Based on these estimates and known facts it was concluded that out of all the energy sources available, nuclear energy is one of the great options because it is a low carbon emission, high density energy source that has capacity factor >90%. This suggests that nuclear energy is as reliable as coal energy in terms of energy generation and has very low carbon emissions like other green technologies.The fuel for nuclear energy is uranium which is present naturally in low concentrations in soil, rock and water. Figure 1 depicts the global distribution of uranium in various rocks, industrial waste and sea water [4].

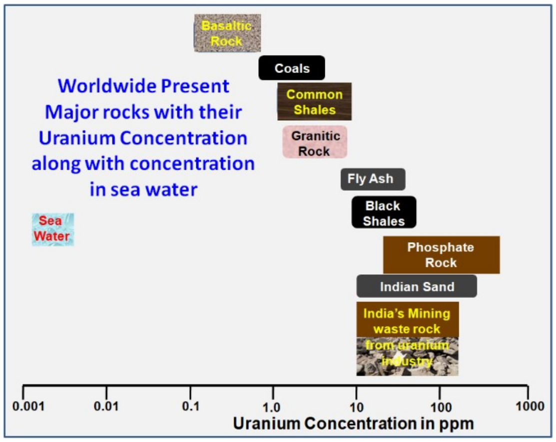

Figure 1. Uranium concentration in various matrices such as different rocks, industrial waste and sea water.

Uranium is present naturally both in the earth and in sea water. The terrestrial reserves are estimated to be ~5 million tons while sea water reserves account for ~4.5 billion tons. In spite of almost ~1000 times more amount of uranium in sea water; most of the uranium presently is extracted from terrestrial reserves. The reason being; extremely low concentrations of uranium in sea water ~3.3 ppb, presence of large concentration of other competing ions and biofouling [5]. These processes make the uranium extraction from sea water uneconomical. In the past decade the technologies have been progressed significantly and the cost to extract uranium from sea water has come down drastically but still it has not reached to its commercial viability yet. Once the uranium extraction from sea water becomes commercialized; the nuclear energy will be a renewable source of energy [6]. Sea water is almost an infinite source of uranium. The uranium mill tailings which are the byproduct from uranium mining can be largely ignored in case of uranium mining from sea water [5].

2. The History of Uranium Mining: The history of uranium mining date back >200 years ago wherein the uranium is extracted from its Pitchblende ore for different purposes. Uranium mill tailings are the waste generated from the processing of uranium ore [6]. Here a brief historical overview of uranium mining is given.

The Central European uranium deposits were the first industrially mined deposits in the world [7] (Milos, 2017). Initially uranium minerals were noticed in some silver ore deposits in 1565. Later uranium was discovered by Klaproth in 1789 [8]. After the discovery of uranium Klaproth used yellow uranium compound; U_3O_8 in making colored glasses [9] and these colored glasses became very popular [10]. It is to be noted that the original production of uranium color glasses, also known as Vaseline glasses in 1853 was 84.6 kg which rose to 12,776 kg in 1886 [11]. This suggests the vast usage of these colored glasses [12]. These glasses were primarily used for decorative purpose but in some cases they were also used in some ultra-violet protective eye glasses. Other uses of uranium glass include use in photographic dark room window glasses and electric light bulbs. With time, the applications of uranium in other areas were also discovered. In Germany, uranium salts were used in dying textiles, papers and leather. Dentist in United States and England used uranium yellow for coloring artificial teeth. Uranium was also used to improve the elasticity and hardness of the steel. After ~100 years of use the radioactive properties of uranium were discovered by Henri Becquerel in 1896. Follow up research by Marie Curie had led to the invention of two other radioactive elements in uranium series namely, radium and polonium. Soon the interest on radium developed [13] and Vaseline glass industry saw a dip. Uranium mining is then being carried out for the production of radium which became a material of high demand. In 1911, Kelly first demonstrated therapeutic use of radium and was among the first to treat cervical cancer using radium [13]. Radium was used in luminescent paints, to decorate the dials of clocks and watches and in medical industry. It was also being used in water, food, facial creams and toothpaste until its toxicity was reported and the famous case of radium girls came to the light [14]. By 1930, the radium use declined drastically.

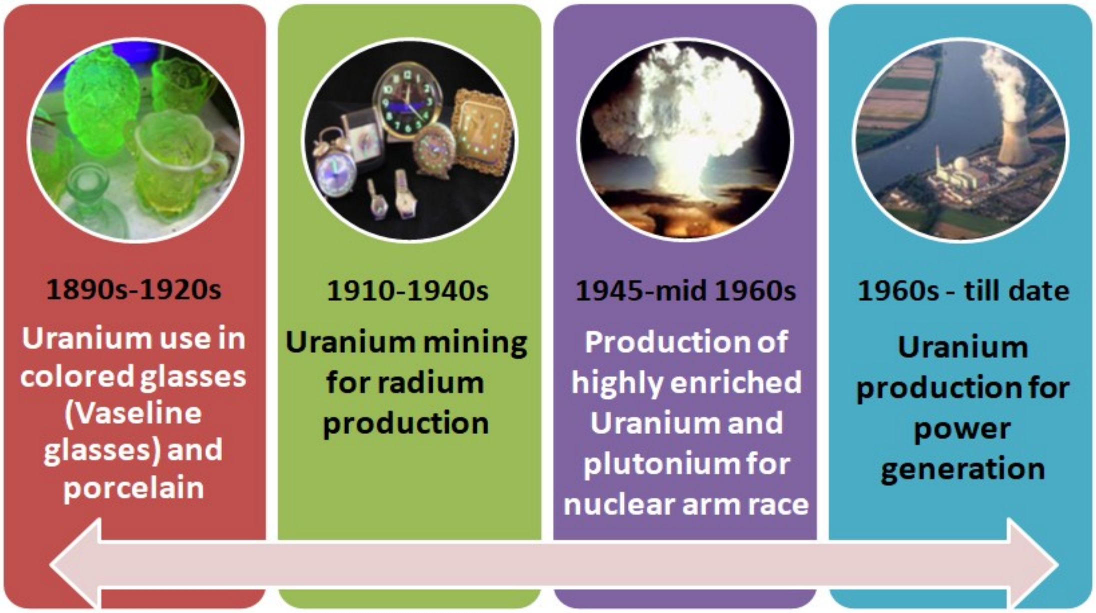

Figure 2. Use of uranium in industries at different time periods.

Otto Hahn and Fritz Strassmannin 1934 found that uranium can undergo fission and release a large amount of binding energy. This energy can be used to fulfill the power requirement and replace coal because it is an extremely dense and clean source of energy.It was also discovered that uranium can be converted to ^{239}Pu which like ^{235}U isotope is also fissile by thermal neutrons and a weapon grade material. On 2 December 1942, Enrico Fermi has made the world's first artificial nuclear reactor named Chicago Pile-1 (CP-1) which has shown the world that a controlled self sustained chain reaction is possible.With the start of World War - II, the efforts has been shifted from generating power from this extremely clear energy source to the production of nuclear weapons to become the world power.The Cold War between the Soviet Union and the United States resulted in massive uranium stockpiles and the creation of tens of thousands of nuclear weapons using enriched uranium and plutonium derived from uranium ores. After 1949, uranium ore became a highly strategic mineral resource.In United states, during the mid 1950s almost 750 uranium mines were in operation and 7 million tons of uranium ore has been identified till 1958. The US government was the sole purchaser of this produced uranium till 1971 to meet the requirement in the country [7].

After the World War - II, the construction of nuclear power plants for electricity generation started and United States in December 1951 made the first small scale nuclear reactor, EBR- 1 to produce electricity. The technology spread all over the world and uranium is now being used in the construction of nuclear power reactors. Figure 2depict the importance of uranium mining in different industries and their dominance in a particular era.

3. Uranium mining at different time periods: Different countries have mined uranium based on the resources available and the requirement [15]. Figure 3 shows the uranium production from 1945 till 2010. It is clearly seen from Figure 3 that uranium production sees a fluctuating pattern over time and can be correlated well with the events and regulations came during that time. It can be seen that uranium production increases from 1945 till 1960 and then decreases. This is because two nuclear accidents occur in the

United States of America in 1961 and 1966. Then the uranium production again increases and peak around 1980 because of oil crises. The world has understood the need to explore other clean and independent sources of energy. Following this, the production suddenly decreased because of two major nuclear accident; three miles accident in United States on March 28, 1979 and Chernobyl, Soviet Union on April 26, 1986. However, the demand continued to climb. After this uranium production again increases from 2000 onwards. This shows that the activities related to uranium is strongly affected by the past effects and the policies formed so forth.

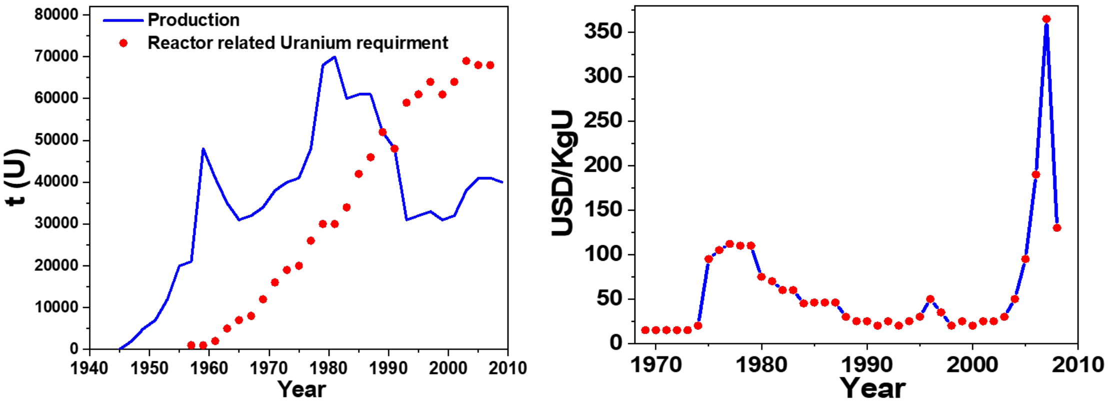

Figure 3. Production, demand of reactor related uranium along with the cost over different time periods.

Apart from these nuclear accidents, uranium industry is badly suffered by the stringent regulations imposed on uranium related activities. This was largely because of the radiation induced effect such as cancer [16] and hereditary effects which were found at extremely high doses but still unproven at low doses. It is noteworthy that in 1928, the radiation exposure limit for the public was set at 1 Sv per year, which was reduced to 500 mSv per year in 1934. By 1966, the limits were further lowered to 50 mSv per year for workers and 1 mSv per year for the public. Currently, the accepted radiation dose limit is 20 mSv per year for workers and 1 mSv per year for the public [17].The ratcheting down of dose limit can be correlated with several events and the studies published related to radiation risk. Some of the prominent events were discovery of mutation in fruit fly by Muller [18] in 1927 and linear no threshold model (LNT), Hiroshima Nagasaki bombing in 1945, US NAS recommendation to adapt LNT in 1956 and nuclear accident as mentioned above. This demonstrates the increasingly stringent regulations imposed on uranium-related industries.

Uranium mining results in generation of large amount of uranium tailings. The next section of this article will focus on the generation of these tailings.

4. Uranium Mill Tailings generation:

Terrestrial mining of uranium results in large amount of unutilized material generation called as uranium mill tailings. Tailings are the by-product of uranium mining and primarily the sandy process waste material that contains the radioactive decay products from the uranium chains (mainly the U-238 chain) along with the un-extracted uranium, heavy metals and all the associated liquids. After finding the uranium ore; mining is being carried out to extract this ore and depending upon the ore grade, its mineralogical properties and depth from surface; uranium extraction activities are being planned [19]. The processes involved in uranium mining and milling till the formation of yellow cake and the associated tailings are shown in Figure 3.

Worldwide, uranium mining and milling is based primarily on hydrometallurgical operations such as leaching, solvent extraction, and precipitation. The first step in uranium extraction is uranium mining. Uranium ore can be present in underground depository or on the surface. A particular particle size is suitable for the maximum uranium leaching and therefore the ore is crushed and grind to achieve a desired particle size by suitable techniques. Uranium is leached from the ore either by acid leaching or alkaline leaching process depending on the ore mineralogy. If the ore is present on the surface in-situ leaching process is adopted for uranium extraction. Based on the exploration of different uranium reserves and with the advancement of technologies; the leaching method changes. In 1990, 55% of the uranium comes from underground mines (acid or alkaline leaching) but presently around 55% of the uranium is recovered from *in-situ* leaching process [15]. Here, a brief description about each process with their advantages will be discussed.

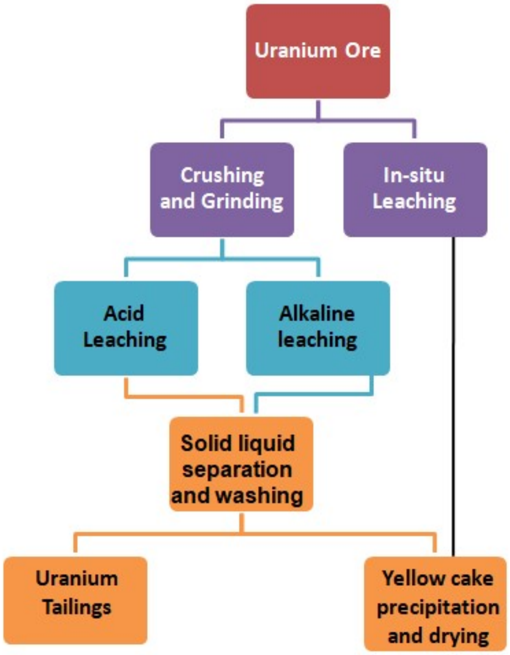

Figure 4. Processes involved in uranium extraction from uranium ore.

Acid leaching is one of the most common underground methods for uranium extraction from its ore [15]. Sulphuric acid is used for leaching. The amount of the acid consumption is controlled by the ore chemical properties and it may vary from 10 - 100 kg of H_2SO_4 per tonne of ore. In certain cases, heating is used to reduce the leaching time and increase the efficiency of the process. Sometimes an oxidant such as manganese dioxide or sodium chlorate is also added to achieve satisfactory uranium extraction and convert all uranium into oxidized form that is readily soluble. By this process ~85-95% uranium recovery is reported[5]. The resulting leach solution is then separated from solid and after repeated washing solidgoes to the tailing ponds while the leach solution goes for the further processing. The concentration of uranium in leach solution is normally 1- 2 g/litre along with the presence of other ions that is present in the ore. The selective uranium extraction is carried out by solvent extraction or ion exchange process wherein uranium is extracted selectivelyin organic phase by a suitable solvent or with an ion exchanger and then strip back into the water by contacting it withan inorganic salt solution, such as sodium chloride, ammonium sulphate or changing the pH conditions in ion exchange. Both the organic solvent and ion exchangers are repeatedly used in the process and after reaching the designated leaching efficiency the waste goes into the tailing ponds. The yellow-cake is precipitatedfrom the strip solution, and the resulting solid is dried. The chemical reaction involved in the process are shown below [20]:

$$UO_3 + 2H^+ \rightarrow UO_2^{2+} + H_2O \quad (1)\ \text{dissolution}$$

$$UO_2 + 0.5O_2 + 2H^+ \rightarrow UO_2^{2+} + H_2O \quad (2)\ \text{oxidation}$$

$$3UO_2 + ClO_3^- + 6H^+ \rightarrow 3UO_2^{2+} + Cl^- + 3H_2O \quad (3)\ \text{oxidation}$$

$$UO_3 + H_2SO_4 \rightarrow UO_2SO_4 + H_2O \quad (4)\ \text{leaching}$$

$$UO_2SO_4 + SO_4^{2-} \rightarrow [UO_2(SO_4)_2]^{2-} \quad (5)\ \text{leaching}$$

$$[UO_2(SO_4)_2]^{2-} + SO_4^{2-} \rightarrow [UO_2(SO_4)_3]^{4-} \quad (6)\text{leaching}$$

The concentration of uranium sulphate complex formed by reaction 4, 5, 6 depends on the concentration of sulphuric acid used and uranium. The formed compound can be selectively extracted using an anion exchanger followed by elusion and precipitation. All the other processed material goes to the tailing ponds and need proper management.

Alkaline leaching process for uranium production is adopted where the ore is rich in limestone and henceforth acid leaching is not favourable. Since uranium is known to form different complexes under different conditions, in this process soluble carbonate complexes of uranium are being formed to leach out the uranium. The most common alkaline leaching solutions are mixtures of sodium carbonate and sodium bicarbonate [19]. The chemical reactions involved in the process are:

$$2UO_2 + O_2 \rightarrow 2UO_3 \quad (7)$$

$$UO_3 + 3Na_2CO_3 + H_2O \rightarrow Na_4UO_2(CO_3)_3 + 2NaOH \quad (8)$$

$$2Na_4UO_2(CO_3)_3 + 6NaOH \rightarrow Na_2U_2O_7 + 6Na_2CO_3 + 3H_2O \quad (9)$$

Side reactions:

$$Al_2O_3.3H_2O + 2Na_2CO_3 \rightarrow 2NaAlO_2 + 2NaHCO_3 + 2H_2O$$

It can be seen that in alkaline leaching process, Na_2SO_4 formation occur if pyrite is present. Because of this acid drainage may occur from the tailings and efforts are being taken to remove this sulphate. In *insitu* leaching process, the uranium is extracted from the host rock by chemical solutions followed by the recovery of uranium at the surface [21]. In this process a suitable leach solution is injected into the ore zone below the water table. The uranium is leached into the solution which is pumped to the surface for further processing. The technology was first used in 1962 at Ukraine for uranium extraction and has evolved a lot since then. In 1990s, *insitu* leaching process contributes 13-15% of world uranium production and now it contributes ~55%. The leaching can be carried out either with acid (H_2SO_4) or with the base (Na_2CO_3) depending upon the ore composition. The chemical reaction involving these processes are same as mentioned above in acid and alkaline leaching process. Since the process does not involve any ore extraction, the generation of uranium mill tailings is least in this process.

5. Challenges associated with Uranium Mill tailings management: Uranium mill tailings, although primarily a material that is present naturally in the earth but its management involve huge cost and challenges [22, 23]. One of the primary challenges is radioactivity present in tailings [24]. Uranium mill tailings contain radioactive isotopes, primarily uranium and its decay products, such as thorium and radium. Although most of the uranium is extracted from the ore, still some uranium is present in these tailings along with the decay product. Many of the radioactive isotopes present in uranium tailings have long half-lives and they remain radioactive for extended periods. For example, uranium-238 has a half-life of about 4.5 billion years, while radium-226 has a half-life of approximately 1,600 years. Because of these long lived isotopes and stringent regulations kept; managing these long lived radioisotopes requires careful planning and monitoring.It is also postulated thaturanium tailings if not adequately contained can contaminate soil, water, and air. As per many reports radioactive and heavy elements present in the tailings can leach into the surrounding environment, posing risks to ecosystem and human health [22]. The other challenges that lie in tailings management is erosion. Uranium tailings are often stored in surface impoundments or piles. These structures are susceptible to erosion, which can lead to the dispersion of contaminated particles into the surrounding environment through wind and water erosion. Proper containment and erosion control measures are essential to mitigate this risk.Therefore, it is essential to find a suitable solution for tailings management that can mitigate all these challenges and environmental monitoring for a long period of time.

Because of the above challenges, tailings impoundments or ponds are constructed to ensure the containment and isolation of these tailings. These are engineered containment structures where tailings are deposited and stored. Some of the countries also explore the option of placing uranium mill tailings in underground repositories, where geological formations provide natural barriers against the release of radioactive materials. In certain situations, mined-out areas are backfilled with processed tailings, effectively returning the tailings to the underground workings of the mine. This approach aims to minimize surface storage and reduce the long-term environmental impact. Geotechnical engineering measures, such as compacting and stabilizing tailings, are employed to enhance the physical properties of the stored material. This helps reduce the potential for erosion, settling, and the generation of dust.

Addressing uranium mill tailings management requires a multi-faceted approach involving rigorous environmental monitoring, engineering controls, risk assessments, and stakeholder engagement. Effective management of uranium tailings is essential to minimize environmental impacts and protect public health in both active and legacy mining areas.

6. Worldwide scenario on management of tailings containing natural radioactivity: As mentioned above uranium can be extracted from ore via acid leaching, alkaline leaching or in-situ leaching process and depending on the process, the characteristics of uranium tailing changes. The utilization of these tailings in any form requires its thorough characterization including mineralogical, chemical and radiological constituents, their transport behavior and impact on the environment. Since the tailings have materials of potential interest, these baseline studies are necessary to see their potential in different industries [25]. In today's world efforts are being put to follow the principal of circular economy which states "sharing, leasing, reusing, repairing, refurbishing and recycling existing materials and products as long as possible". Many industries have successfully fulfilled this approach by utilizing their waste in different industries for example ceramic, glass, building materials, agriculture and cement industries etc [26]. One of the major concerns with uranium mill tailings management is the low radioactivity content. Few industries such as thermal power plant which produce fly ash and fertilizer industry that produce phosphogypsum; both of these products is known to have radioactivity content but they are being utilized successfully in other industries [27]. Herein, we will mention some of these studies and show that similar strategies should be adopted for uranium mill tailings management also.

Coal remains one of the major primary source for power production not only in India but throughout the World. The burning of coal produces fly ash as a by product. The major constituent of fly ash are silica, alumina, calcium oxide and iron oxide; all the major elemnts used in various industries. Coal is known to contain radioactivity and the content may varry depending upon the geological origin and age of formation [27]. A significant portion of this radioactivity is passed to the fly ash also. A large amount of work has been carried out in India on utilization of fly ash in bricks, cement, fertilizer and polymer industry. Government of India has also issued various notification on fly ash utilization time to time and emphasized on incresed utilization of fly ash and creating wealth from waste at the expense of natural reserves. It is important to mention thatin 1996-97 there was a huge gap in fly ash generation and its utilization but with time and recently in past 7-8 years its utilization has incresed exponentially suggesting the growth and demand in the sector. These efforts suggest the technological advancement, public acceptance in fly ash utilization. Some of the major industries for fly ash utilization are the cement, road and highway constrction, tiles and bricks and reclamation of low lying areas.

Since India is the most populous country in the world and land is a very precious community, Government of India has adopted various technologies to use industrial waste in construction materials [26]. Various standards have also been formulated in this direction [28-31]. Here it is important to mention that Indian uranium ore reserves are in low to very low category. Therefore the uranium concentration present in tailings is very low. The uranium mill tailings come under the category of Naturally Occurring Radioactive Materials (NORMs) [27]. Other industries such as oil, gas, coal, and phosphate also produce NORMs [27] and these residues have been successfully repurposed in construction sector. For instance, fly ash has been optimized for use as construction materials while maintaining radiological safety [32-34]. Kovler has demonstrated radiological concern in using industrial byproducts in construction [35]. He has also stressed that a paradigm shift is required in management of radioactive waste with justified dose limits [27]. In India, the Atomic Energy Regulatory Board (AERB) permits the use of NORM-containing waste in construction if radiation doses remain below 1 mSv/year, in line with natural background levels. In India, millions of tons of uranium mill tailings is present that needs proper management. Therefore, technologies should be developed to manage this waste.

7. Conclusions: Uranium being fuel for the nuclear energy, its production is predicted to increase as more and more reactors are being planned in the coming years which in turn will increase the tailings. Therefore, it is essential to develop technologies where these tailings can be used. So far the construction industry has been the major choice for the waste utilization; in line with the principle of circular economy it is postulated that uranium mill tailings can also be used as raw materials in construction industry since the basic composition of tailings and various construction materials matches.

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