



# **A Review of In Situ Leaching (ISL) for Uranium Mining**

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Abstract: Uranium, a cornerstone for nuclear energy, facilitates a clean and efficient energy conversion. In the era of global clean energy initiatives, uranium resources have emerged as a vital component for achieving sustainability and clean power. To fulfill the escalating demand for clean energy, continual advancements in uranium mining technologies are imperative. Currently, established uranium mining methods encompass open-pit mining, underground mining, and in situ leaching (ISL). Notably, in situ leaching stands out due to its environmental friendliness, efficient extraction, and cost-effectiveness. Moreover, it unlocks the potential of extracting uranium from previously challenging low-grade sandstone-hosted deposits, presenting novel opportunities for uranium mining. This comprehensive review systematically classifies and analyzes various in situ leaching techniques, exploring their core principles, suitability, technological advancements, and practical implementations. Building on this foundation, it identifies the challenges faced by in situ leaching and proposes future improvement strategies. This study offers valuable insights into the sustainable advancement of in situ leaching technologies in uranium mining, propelling scientific research and practical applications in the field.

**Keywords:** uranium mining; in situ leaching (ISL); acid leaching; alkaline leaching; neutral leaching; bioleaching; blasting-enhanced permeability (BEP); reactive transport model (RTM)

# 1. Introduction

While fossil fuels, represented by coal, oil, and natural gas, continue to dominate the global energy landscape and meet the majority of energy demands, the escalating concern regarding climate change and environmental issues [1,2] has brought attention to the negative impacts associated with their production and utilization [3–10]. Specifically, the combustion of fossil fuels releases substantial amounts of greenhouse gases, intensifying global warming [3,5]. In response to the challenges, there is a worldwide momentum toward accelerating the advancement of clean energy, with nuclear energy, which is rooted in uranium mining, emerging as a noteworthy and environmentally friendly energy source [11,12]. Nuclear energy undergoes the process of fission, converting it into thermal energy, which is subsequently transformed into electricity through successive steps. Nuclear energy stands out as a clean and efficient energy suitable for diverse applications in power generation and beyond [13–15]. Its high energy density enables the provision of sustained and stable electricity supply [12,16]. In comparison to traditional fossil fuels, nuclear energy serves as a low-carbon energy source, avoiding the generation and emission of carbon dioxide ( $CO_2$ ) during its utilization [17]. It also minimizes air pollutants, such as nitrogen oxides, during operation, underscoring its significant environmental benefits [12].

Uranium, as the foundational material for nuclear energy, is commonly distributed throughout the Earth's crust at relatively low concentrations [11]. Globally, economically viable uranium deposits are unevenly distributed, with sandstone-hosted uranium deposits being the most prevalent [18–22]. Based on the latest data updated by the World Nuclear Association (WNA) in 2023, the total recoverable identified resources have reached



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**Copyright:** © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). 7.918 million tonnes U (tU) [23]. The top five countries with the largest uranium reserves are Australia (28%), Kazakhstan (13%), Canada (10%), Russia (8%), and Namibia (8%) [23]. The Organisation for Economic Co-operation and Development (OECD) reports that as of 2020, uranium production was carried out in 17 countries worldwide, with a total output of 47,342 tU. The top five uranium-producing countries are Kazakhstan (41.1%), Australia (13.1%), Namibia (11.4%), Canada (8.2%), and Uzbekistan (7.4%) [24]. Given the imperative transition toward sustainable energy, the OECD predicts a continual rise in global uranium demand [24]. Therefore, it is crucial to improve uranium mining technologies to meet this escalating need. This endeavor supports the sustainable development of the nuclear energy industry while creating a more stable global energy foundation, promoting the widespread adoption of clean energy.

The traditional uranium mining methods include open-pit mining [25] and underground mining [26]. In recent years, heap leaching [27] and in situ leaching (ISL, also known as in situ recovery, ISR) [28,29] have emerged as two breakthrough technologies utilized in uranium mining. In comparison to open-pit and underground mining, these two mining technologies offer distinct advantages such as environmental sustainability, efficient extraction, and cost-effectiveness [28,29]. Furthermore, it is crucial to emphasize that low-grade sandstone-hosted uranium deposits, which were previously economically unviable for extraction, can now be economically and efficiently mined using these two mining technologies. Regarding the heap leaching technology, some research has been conducted [27,30–34]. Ghorbani et al. have conducted a thorough and detailed review of its current development status, technological innovations, and future directions [32]. Peterson has provided a comprehensive introduction to this technology as a key method for extracting minerals from low-grade ores [27], highlighting the significance of heap leaching. Regarding the in situ leaching technology, its value is reflected in its increasingly widespread practical application in recent years. In 2021, approximately 63% of uranium was produced via in situ leaching [24,35].

The annual publication statistics offer valuable insights into the evolving research landscape concerning in situ leaching and its associated fields. As depicted in Figure 1, the volume of pertinent academic literature demonstrates a pronounced upward trajectory, signifying an escalating interest in the domain of in situ leaching. These studies associated with in situ leaching for uranium mining may have different emphases. Seredkin et al. comprehensively compare in situ leaching with traditional mining technology from the perspectives of exploration, environmental impact, and economics, emphasizing its advantages as an alternative to conventional methods of mining [36]. Bhargava et al. elucidate the fundamental principles of in situ leaching for uranium extraction and analyze factors that may influence its effectiveness [37]. Mudd provides the detailed application measures and resulting impacts of in situ leaching [38,39]. Furthermore, certain studies focus on the practical application effects of in situ leaching in specific uranium deposits [40–42]. While research on in situ leaching for uranium mining continues to expand, there is currently a lack of comprehensive studies providing systematic summaries of its sub-techniques for different conditions, the associated principles, and its recent advancements. Therefore, this study aims to thoroughly review the most recent progress of in situ leaching for uranium mining, addressing a gap in the existing research.

This review study was conducted through a systematic process of information collection, classification, analysis, and summarization. The information was sourced extensively, primarily from peer-reviewed journal papers and conference proceedings evaluated through databases such as Web of Science, Scopus, and China National Knowledge Infrastructure (CNKI). It was complemented by relevant information from international organizations, government reports, and company websites. The timeline of these publications spans over a century, from the early 20th century to the year 2023. Subsequently, the collected information was categorized based on different sub-techniques and their underlying principles, technological advancements, and practical application status. A meticulous analysis was conducted on this basis, summarizing the applicable conditions, advantages, and disadvantages of these in situ leaching sub-techniques, including acid leaching, alkaline leaching, neutral leaching, and bioleaching. Additionally, the recent technological advancements and the real-world field application of in situ leaching are introduced. Furthermore, the current challenges faced by in situ leaching are clarified, and future improvements are proposed. This comprehensive review offers researchers and practitioners a nuanced understanding of the latest developments in in situ leaching for uranium mining, precisely delineating the practical application scope of the technology. Through an in-depth analysis of the current technological landscape, this study aims to steer future research directions and facilitate technological enhancements. The continued development of in situ leaching techniques for uranium mining will play a pivotal role in maintaining the long-term sustainability of nuclear energy as a primary clean energy source going forward on a global scale.



Figure 1. Dynamics of scientific publications on in situ leaching from 1980 to 2023.

## 2. Overview of Uranium Mining Methods

The formation of uranium deposits is influenced by various factors such as geological structures, sedimentary processes, rock types, and fluid interactions [20,22,43–49]. The combined effect of these factors may lead to significant variations in the burial depth of uranium deposits, ranging from near-surface locations to several hundred meters or more underground. Open-pit mining and underground mining, which are traditional uranium mining methods, are classified based on the suitability for the burial depth of uranium deposits. Open-pit mining is primarily employed for uranium deposits with shallow burial depths [50–52]. Figure 2 illustrates the open-pit mining process for uranium mines. Its core procedures involve excavating and removing the topsoil and overburden covering the uranium directly to expose the ore body [53]. The exposed ore body is then ripped and transported to the stockpile and mill sit for subsequent processing to obtain uranium products. On the other hand, underground mining is applied to deeper-buried uranium deposits or those where open-pit mining is not feasible [54–56]. The method involves sinking a shaft or driving an adit near the ore body to extend levels at various depths, allowing miners to access and remove the ore to the surface for subsequent processing. The schematic diagram illustrating the utilization of underground mining for uranium extraction is depicted in Figure 3.



Figure 2. Schematic diagram of open-pit mining.



Figure 3. Schematic diagram of underground mining.

At present, in situ leaching stands as another extensively employed mining technique [2,35,48,57], alongside open-pit and underground mining. Unlike open-pit and underground mining, in situ leaching does not rely on burial depth as a criterion but is based on the properties of the uranium deposit. As a mining technique conducted under the natural conditions of ore deposits, in situ leaching is well-suited for uranium deposits situated within aquifers and characterized by favorable permeability [58–60]. This method involves strategically arranging and drilling wells, including injection wells and production wells (or recovery wells), within the uranium deposit. The leaching solution (or lixiviant) is injected to react with the ore, dissolving uranium. The uranium-bearing solution, known as the pregnant solution, is then brought to the surface for further treatment. Table 1 presents the applicable conditions for these three uranium mining methods, along with their respective advantages and disadvantages.

Method	Applicable Condition	Advantage	Disadvantage	Reference	
Open-pit Mining	Shallow burial depths of main ore bodies	Short construction period; Large mining space and high labor productivity; Safe working conditions	Subject to climate conditions; High infrastructure and equipment investment; Large land footprint and environmental damage	[50,61]	
Underground Mining	Significant burial depths or surface conditions unsuitable for open-pit mining	Limited climate disruption; Minimal impact on surface ecosystems; High mining efficiency	High extraction costs; Complex and challenging construction and maintenance of underground mining facilities; Potential impact on underground geological environment; Elevated safety risks	[26,53,54]	
In Situ Leaching	Situated in aquifers with favorable permeabilities	Safe and simple mining process; Short construction period and minimal infrastructure investment; Low labor intensity and high automation level; Less environmental pollution due to avoidance of waste rocks; Capability to process low-grade ore deposits	Requirements for geological and hydrogeological conditions; Slow extraction rate; Underground water management challenges	[58–60,62]	

Table 1. Comparison of uranium mining methods.

Compared to open-pit and underground mining, in situ leaching offers distinctive advantages. By operating directly within natural ore deposits, this mining method avoids extensive surface disruption and excavation. Consequently, it lowers the input costs, shortens the mining process, and entails a more minimal environmental impact. Moreover, in situ leaching involves the direct injection and cyclic utilization of leaching solution into uranium deposits, effectively eliminating the generation of unwanted by-products such as waste rock, radioactive dust, and emissions. This prevents health hazards for personnel in addition to reducing the expenses associated with waste disposal. Furthermore, while leaching uranium elements, the leaching solution also facilitates the extraction of uranium-associated elements such as selenium (Se), scandium (Sc), and molybdenum (Mo), thereby enhancing the overall recovery rate of valuable minerals [63].

In addition, in situ leaching significantly enhances the industrial value of low-grade sandstone-hosted uranium deposits. These deposits are widely distributed globally, encompassing countries like Australia, Canada, and Kazakhstan [11], constituting a vital component of the world's uranium resources. Considering the costs and environmental impacts, open-pit and underground mining are essentially impractical for these deposits characterized by low uranium content. Fortunately, in situ leaching can overcome this significant drawback. By operating within the ore body, in situ leaching reduces the excessive costs associated with waste processing and uranium milling while minimizing adverse environmental impacts. Consequently, these deposits can now be economically and efficiently mined, thereby expanding the scope of recoverable uranium resources and providing more robust support for the sustainable growth of the global nuclear energy industry.

## 3. In Situ Leaching Techniques

The schematic diagram of the in situ leaching process for uranium mining is shown in Figure 4. Injection and production wells are strategically positioned in the uranium field. The pre-configured leaching solution is injected into the target ore layer through injection wells to dissolve uranium. The uranium-bearing pregnant solution is then extracted to the surface through production wells. The connecting pipelines of the injection and production wells are equipped with header systems, which are linked to the processing plant via trunk lines. Following the extraction of uranium from the production wells, the pregnant solution undergoes specific processing [48,64,65], including uranium recovered by ion exchange, concentration, filtration, and drying. Ultimately, the qualified uranium concentrate, predominantly composed of triuranium octoxide ( $U_3O_8$ ), known as yellowcake [66], is obtained. Simultaneously, the treated solution is reintroduced into the ore body for cyclic utilization as the leaching solution [48].



Figure 4. Schematic diagram of the in situ leaching process.

Uranium, classified as a redox-sensitive lithophile element [67], exhibits a strong affinity for oxygen [68]. Theoretically, uranium can exist in four oxidation states [69], including +3, +4, +5, and +6. However, due to the instability of uranium in the +3 and +5 states, uranium minerals typically manifest in nature in the +4 (U(IV)) and +6 (U(VI))oxidation states [70–73]. Specifically, U(IV) typically appears in the solid forms of uraninite  $(UO_2)$  and coffinite  $(U(SiO_4)_{1-x}(OH)_{4x})$  [37,74], representing common and stable uranium configurations. These compounds have low solubility and are extensively present in uranium ores under reducing conditions. U(VI) primarily exists as the solid form of uranium trioxide (UO<sub>3</sub>) in the ore body and the soluble uranyl ion (UO<sub>2</sub><sup>2+</sup>) in the aqueous systems of uranium deposits [75,76]. These configurations are prevalent under oxidizing or oxygen-rich conditions. The coexistence of U(IV) and U(VI) is also observed in some uranium deposits [77]. Typically, U(VI) demonstrates greater chemical reactivity in the high oxidation state than U(IV) in the lower oxidation state [78]. Additionally, these two valence states of uranium exhibit significant differences in crystallographic and geochemical properties [79,80]. Consequently, the thoughtful selection of a leaching solution tailored to the unique conditions of uranium deposits is crucial, as it significantly impacts key aspects of uranium mining [37,48,57,81], including uranium dissolution efficiency, costeffectiveness, and environmental considerations. The choice of in situ leaching techniques aligns with the diverse geological conditions and mineral compositions of different uranium deposits. These techniques are systematically categorized based on the primary components of the leaching solution, encompassing acid leaching, alkaline leaching, neutral leaching, and bioleaching.

## 3.1. Acid Leaching

In situ acid leaching primarily employs strong acids as the main components of the leaching solution [37,39]. Upon contact with the ore, solid-state uranium is converted to a soluble form under conditions with pH < 2 [48,82], facilitating its extraction. In situations where uranium exists solely in the U(VI) form, characterized by its heightened chemical reactivity, it can directly react with the acid solution [76]. However, when uranium in the ore body predominantly exists in the U(IV) form or in a mixed form of both U(IV) and U(VI), the addition of an oxidizing agent to the acid solution becomes necessary [83]. This process oxidizes U(IV) to the hexavalent form, enhancing its reactivity and enabling subsequent reactions with the acid leaching solution to form soluble complexes. At present, sulfuric acid (H<sub>2</sub>SO<sub>4</sub>) is the most frequently utilized component in the acid leaching solution [64], while nitric acid (HNO<sub>3</sub>) and hydrochloric acid (HCl) are occasionally employed [84]. Common oxidizing agents used in conjunction with acid solutions include oxygen (O<sub>2</sub>) [82] and hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) [85,86].

For solid U(VI) in the ore body, such as UO<sub>3</sub>, the fundamental reaction in the in situ acid leaching process is expressed by Reaction (1) [75,76]. Under the influence of H<sup>+</sup>, U(VI) transforms from a solid state to the most soluble uranyl ion  $(UO_2^{2+})$  form. Depending on the composition of the acid solution, uranyl ion combines with sulfate, nitrate, and chloride ions, ultimately existing in the productive solution as complexes like  $UO_2SO_4$ ,  $UO_2(NO_3)_2$ , and  $UO_2Cl_2$ .

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

In the case of U(IV), such as UO<sub>2</sub>, it undergoes reactions with commonly used oxidizing agents [75,83,87]. The fundamental principle of the oxidation process relies on its ions acting as electron transfer mediators to facilitate the uranium transformation from the tetravalent form into the hexavalent form [88]. Reaction (2) describes the oxidation process in the presence of oxygen as the oxidizing agent [82], while Reaction (3) depicts the oxidation process when hydrogen peroxide is utilized as the oxidizing agent [85,86].

$$2UO_2 + O_2 \rightarrow 2UO_3 \tag{2}$$

$$UO_2 + H_2O_2 \rightarrow UO_2^{2+} + 2OH^-$$
 (3)

After undergoing Reaction (2) or Reaction (3), hexavalent uranium converted from  $UO_2$  subsequently reacts with the acid solution. The oxidation of  $UO_2$  and subsequent reaction with an acid can be combined and represented as Reaction (4) [87].

$$UO_2 + O + 2H^+ \rightarrow UO_2^{2+} + H_2O$$
 (4)

However, it is crucial to note that the in situ acid leaching technique has certain application limitations. This technique is not suitable for uranium deposits with high carbonate content, as it can lead to inefficiency and economic impracticality caused by the excessive consumption of the acid leaching solution. This is attributed to neutralization reactions [89] described in Reaction (5), where carbonate reacts with the acid leaching solution. Specifically, for optimal leaching results, the technique is applicable when the carbonate content in the ore body is less than 8% [37,90] and less than 2% in the aquifer [39,91].

$$2\mathrm{H}^{+} + \mathrm{CO}_{3}^{2-} \to \mathrm{H}_{2}\mathrm{O} + \mathrm{CO}_{2} \tag{5}$$

## 3.2. Alkaline Leaching

In situ alkaline leaching involves the use of an alkaline leaching solution to react with the solid uranium in the ore. Uranium becomes soluble at pH > 9 [90], facilitating subsequent extraction. While alkaline leaching exhibits lower extraction efficiency due to low kinetics and high energy consumption compared to acid leaching, its less corrosive nature and lower impurity content partially compensate for these drawbacks [28,90]. In

addition, alkaline leaching solutions can be employed for uranium deposits rich in alkaline accessory minerals, such as carbonates and some silicates [90,92]. These deposits, which are unsuitable for acid leaching due to their high acid consumption [75], find compatibility with the alkaline leaching technique.

An alkaline leaching solution is primarily composed of carbonate or bicarbonate [93,94], complemented by an oxidizing agent. The most utilized predominant constituents are sodium carbonate and sodium bicarbonate [95]. For alkaline leaching of U(VI), the reactions are delineated by Reaction (6) for carbonate ions and Reaction (7) for bicarbonate ions, respectively. In cases where the alkaline leaching solution reacts with U(IV), it is expressed through Reaction (8) for carbonate ions and Reaction (9) for bicarbonate ions, respectively. Both uranyl decarbonate ions (UO<sub>2</sub>(CO<sub>3</sub>)<sub>2</sub><sup>2-</sup>) and uranyl tricarbonate ions (UO<sub>2</sub>(CO<sub>3</sub>)<sub>3</sub><sup>4-</sup>) are soluble complexes that are eventually pumped to the surface for further processing.

$$UO_3 + 3CO_3^{2-} + H_2O \to UO_2(CO_3)_3^{4-} + 2OH^-$$
(6)

$$UO_3 + 2HCO_3^- \rightarrow UO_2(CO_3)_2^{2-} + H_2O$$
 (7)

$$UO_2 + 2CO_3^{2-} + 0.5O_2 + H_2O \to UO_2(CO_3)_2^{2-} + 2OH^-$$
(8)

$$UO_2 + 3HCO_3^- + 0.5O_2 \rightarrow UO_2(CO_3)_3^{4-} + H_2O + H^+$$
 (9)

Alkaline leaching is generally not recommended for uranium deposits with significant concentrations of pyrite (FeS<sub>2</sub>) and other sulfides [75,96]. This is due to the ability of carbonate or bicarbonate ions in an alkaline environment to accelerate the oxidation reaction of pyrite, leading to the formation of various soluble iron–carbonate complex compounds. This complex situations includes coexistence of FeHCO<sub>3</sub><sup>-</sup>, FeCO<sub>3</sub><sup>0</sup>, Fe(CO<sub>3</sub>)(OH)<sup>-</sup>, and FeCO<sub>3</sub><sup>2–</sup> [97]. Because of this, a considerable amount of alkali is consumed, making the extraction process ineffective and unfeasible from an economic standpoint.

## 3.3. Neutral Leaching

In situ neutral leaching, a recently developed technology, is termed as such due to its ability to maintain a pH value within the range of 6.8–8.2 during application [98]. Compared to acid leaching and alkaline leaching, this approach employs a gentler leaching solution, resulting in reduced impact on comprehensive groundwater pollution and potentially a smaller environmental footprint [98]. Consequently, it is regarded as a promising green technology. The fundamental principle of this method involves utilizing bicarbonate ions  $(HCO_3^-)$  in the weakly acidic form in water to complex with uranium, forming uranyl ions. Depending on the source of bicarbonate ions, current neutral leaching methods are categorized into  $CO_2$ - $O_2$  leaching and weak acid leaching.

# 3.3.1. $CO_2$ - $O_2$ Leaching

In neutral CO<sub>2</sub>-O<sub>2</sub> leaching, the leaching solution consists of native groundwater mixed with O<sub>2</sub> as an oxidizing agent and CO<sub>2</sub> as a complexing agent [74,98,99]. In this process, injected carbon dioxide dissolves to form carbonic acid (H<sub>2</sub>CO<sub>3</sub>) [100], as depicted in Reaction (10). Carbonic acid then dissociates into bicarbonate ions (HCO<sub>3</sub><sup>-</sup>) in the solution, as illustrated in Reaction (11). Simultaneously, injected oxygen oxidizes a significant portion of U(IV) to the hexavalent form, following Reaction (2). Subsequently, U(VI) can form soluble uranyl decarbonate ions (UO<sub>2</sub>(CO<sub>3</sub>)<sub>2</sub><sup>2-</sup>) through Reaction (7). Alternatively, some UO<sub>2</sub> may directly react with O<sub>2</sub> and HCO<sub>3</sub><sup>-</sup> to generate uranyl tricarbonate ions (UO<sub>2</sub>(CO<sub>3</sub>)<sub>3</sub><sup>4-</sup>) following Reaction (9). The chemical reactions involved in the fundamental principles of neutral CO<sub>2</sub>-O<sub>2</sub> leaching are listed below.

$$CO_2 + H_2O \leftrightarrow H_2CO_3$$
 (10)

$$H_2CO_3 \leftrightarrow H^+ + HCO_3^-$$
 (11)

$$2UO_2 + O_2 \rightarrow 2UO_3 \tag{2}$$

$$UO_3 + 2HCO_3^- \to UO_2(CO_3)_2^{2-} + H_2O$$
 (7)

$$UO_2 + 3HCO_3^- + 0.5O_2 \rightarrow UO_2(CO_3)_3^{4-} + H_2O + H^+$$
 (9)

In addition to the fundamental reactions involving the dissolution of U(IV) and U(VI) mentioned above, the  $CO_2$ - $O_2$  leaching solution also induces other water-rock interactions [98,101]. When carbon dioxide dissolves in water, it forms  $H_2CO_3$ ,  $HCO_3^-$ , and  $H^+$ , leading to an increase in  $H^+$  concentration and subsequently elevating groundwater acidity. This change facilitates the dissolution of carbonate minerals in uranium deposits, forming bicarbonate species that are readily complex with uranium minerals. Moreover, these reactions with carbonate minerals may enhance the porosity and permeability of the deposit pore media [102], favoring the flow of leaching solution and uranium-bearing pregnant solution.

Furthermore, in addition to the formation of uranyl–carbonate complexes of U(VI), the  $CO_2-O_2$  leaching solution can also induce intricate reactions that lead to the generation of uranyl–carbonate ternary complexes of U(IV) [98]. This phenomenon arises from the propensity of U(IV) to engage in chemical reactions with ligands, including oxygen, nitrogen, and sulfur, under some specific conditions, thereby forming coordination bonds. In the presence of carbon dioxide, this process further evolves, giving rise to the generation of uranyl–carbonate ternary complexes, represented by entities such as MUO<sub>2</sub>(CO<sub>3</sub>)<sub>3</sub><sup>2–</sup> and M<sub>2</sub>UO<sub>2</sub>(CO<sub>3</sub>)<sub>3</sub><sup>0</sup>, wherein M<sup>2+</sup> denotes divalent cations like Mg<sup>2+</sup>, Ca<sup>2+</sup>, Sr<sup>2+</sup>, and Ba<sup>2+</sup> [103]. The resultant uranyl–carbonate ternary complexes effectively mitigate the tendency of U(VI) complexes to adsorb onto the charged surfaces of iron (hydro)oxides and/or clay minerals in the presence of carbonate [98], thereby promoting the facilitated migration of uranium-bearing pregnant solution. In essence, the generation of U(IV) complexes is influenced by the presence of carbon dioxide and oxygen, and these complexes, in turn, intricately impact the migratory behavior of U(VI) complexes, ultimately leading to a favorable increase in uranium recovery rates.

In summary, neutral  $CO_2$ - $O_2$  leaching involves a more intricate mechanism in comparison to acid leaching and alkaline leaching. These complex mechanisms collectively contribute to an enhancement in uranium recovery rates. Consequently, the application of  $CO_2$ - $O_2$  in situ leaching is increasingly attracting attention and recognition. However, it is important to note that the physical conditions of the aquifer in uranium deposits can influence the status of carbon dioxide. Moreover, the transport of gaseous carbon dioxide within the porous structure of uranium deposits is subject to capillary action mechanisms [104–106]. Additionally, the dissolution of carbon dioxide may lead to a pH decrease, potentially causing the dissolution of surrounding rock minerals and triggering the secondary precipitation of carbonate minerals [107,108], posing a risk of blockages. Therefore, when implementing  $CO_2$ - $O_2$  in situ leaching, comprehensive consideration must be given to the properties of gases, the characteristics of the aquifer, and the interactions with other constituents within the ore.

# 3.3.2. Weak Acid Leaching

Weak acid leaching [109] employs a low concentration of sulfuric acid, in conjunction with oxidizing agents such as oxygen or hydrogen peroxide, as the principal components of the leaching solution. The development of this technique primarily addresses the application restrictions associated with acid leaching in uranium deposits characterized by high carbonate content. Distinctively deviating from the conventional acid leaching process, which involves the direct dissolution of U(IV) and U(VI) by H<sup>+</sup>, this approach allows the acid to engage in initial neutralization reactions with carbonates present in the ore, such as calcite and dolomite, as elucidated in Reaction (5). Despite the consumption of H<sup>+</sup>, the process gives rise to the formation of  $HCO_3^-$ , involving reactions detailed in Reactions (10) and (11). Subsequently,  $HCO_3^-$ , acting as a weak acidic ion, facilitates the dissolution of U(VI) in accordance with Reaction (7) and the dissolution of U(IV) following Reaction (9) with O<sub>2</sub> as the oxidizing agent or Reaction (12) with H<sub>2</sub>O<sub>2</sub> as the oxidizing agent. The chemical reactions involved in the fundamental principles of weak acid leaching are delineated below.

$$2H^+ + CO_3^{2-} \to H_2O + CO_2$$
 (5)

$$CO_2 + H_2O \leftrightarrow H_2CO_3$$
 (10)

$$H_2CO_3 \leftrightarrow H^+ + HCO_3^- \tag{11}$$

$$UO_3 + 2HCO_3^- \to UO_2(CO_3)_2^{2-} + H_2O$$
 (7)

$$UO_2 + 3HCO_3^- + 0.5O_2 \rightarrow UO_2(CO_3)_3^{4-} + H_2O + H^+$$
 (9)

$$UO_2 + 3HCO_3^- + H_2O_2 \to UO_2(CO_3)_3^{4-} + 2H_2O + H^+$$
(12)

However, careful consideration must be given to the quantity and concentration of  $H_2O_2$  when it is chosen as the oxidizing agent during the weak acid leaching process. While oxidizing U(IV), excessive concentrations of high-concentration  $H_2O_2$  may also participate in reactions described in Reaction (13), potentially causing the re-precipitation of soluble uranium from the solution [109]. Therefore, the judicious selection of the appropriate oxidizing agent and its dosage proves particularly crucial for the ultimate efficacy of uranium leaching when using this approach.

$$UO_2^{2+} + H_2O_2 + xH_2O \rightarrow UO_4 \cdot xH_2O \downarrow + 2H^+$$
(13)

## 3.4. Bioleaching

Bioleaching is regarded as a more environmentally sustainable and cost-effective method for uranium mining, positioning it as a superior alternative to other chemical in situ leaching techniques [75,110,111]. This approach is particularly well-suited for uranium deposits with elevated pyrite and sulfide contents [75,96]. Given the impact of microbial activities on the pH and redox conditions within the system, subsequently influencing uranium solubility, this method involves the introduction of leaching solutions containing specific types of microorganisms into the ore body. These microorganisms, typically dominated by acidophilic, autotrophic iron-, or autotrophic sulfur-oxidizing prokaryotes [75], can alter the environmental conditions in both the uranium deposit and aquifer system, thereby enhancing uranium dissolution and facilitating the recovery process.

The acidophilic Fe- and S-oxidizing microorganism consortia, comprising the principal constituents of the bioleaching solution, fulfill a dual purpose by not only establishing an acidic environment but also facilitating oxidation. Within the acidic milieu created by the microbial consortia, U(VI) transforms into soluble uranyl ions, aligning with the underlying principle of acid leaching for UO<sub>3</sub>, as delineated in Reaction (1) [75]. For U(IV), the trivalent iron ions within the microbial consortia act as oxidizing agents, driving Reaction (14) to convert solid UO<sub>2</sub> into soluble uranyl ions [35,75,112], facilitating subsequent extraction. These chemical reactions involved in the fundamental principles are delineated below.

$$UO_2 + 2Fe^{3+} \rightarrow UO_2^{2+} + 2Fe^{2+}$$
 (14)

Moreover, in the application of bioleaching, a consistent provision of dissolved  $O_2$  and  $CO_2$  is required to sustain acidophilic Fe- and S-oxidizing microorganisms [75]. This highlights the synergistic impact of  $CO_2$ - $O_2$  in situ leaching mechanisms in bioleaching, contributing to increased production yields. Meanwhile, the presence of oxygen initiates the production of soluble ferric iron from Fe<sup>2+</sup>, as depicted in Reaction (15), and the generation of sulfuric acid from reduced sulfur compounds, including elemental S, as illustrated in Reaction (16) [75].

$$4Fe^{2+} + O_2 + 4H^+ \to 4Fe^{3+} + 2H_2O$$
(15)

$$2S^0 + 3O_2 + 2H_2O \to 2SO_4^{2-} + 4H^+$$
(16)

Reaction (15) signifies the biological oxidation of  $Fe^{2+}$  and the regeneration of  $Fe^{3+}$  in the sulfuric acid solution. This process consistently provides trivalent iron ions as oxidants for the oxidation reaction of  $UO_2$ , facilitating its dissolution. Meanwhile, Reaction (16) underscores the inherent advantage of the bioleaching method in extracting uranium from deposits rich in sulfur compounds. The dissolution of sulfides releases additional H<sup>+</sup>, thereby sustaining the acidic environment conducive to the uranium dissolution.

This approach displays an additional benefit in the effective mining of uranium deposits rich in pyrite (FeS<sub>2</sub>). Notably, the presence of pyrite, an inherent auxiliary mineral in this type of ore, undergoes Reaction (17) to release Fe<sup>2+</sup>, SO<sub>4</sub><sup>2-</sup>, and H<sup>+</sup> under the influence of microorganisms and acidic conditions [75,113]. Subsequently, the dissolved species repeatedly transform into Fe<sup>3+</sup> following Reaction (15), thereby expediting the bioleaching process by providing additional iron ions for the oxidation of U(IV) [114].

$$FeS_2 + 14Fe^{3+} + 8H_2O \rightarrow 15Fe^{2+} + 2SO_4^{2-} + 16H^+$$
 (17)

In addition to achieving a more environmentally friendly and efficient in situ leaching, microorganisms have the potential to immobilize uranium from aqueous solutions and seawater, contributing to environmental remediation [115–118]. However, it is essential to highlight the impact of gangue minerals on the bioleaching process. For instance, the generation of H<sup>+</sup> and SO<sub>4</sub><sup>2–</sup> during bioleaching may induce the dissolution of carbonate minerals, such as calcite and dolomite, leading to the subsequent precipitation of secondary minerals like anhydride and gypsum, potentially causing plugging issues.

In summary, the four in situ leaching techniques for uranium mining have been elaborately discussed, and their fundamental principles are illustrated in Figure 5. Their applicable conditions and respective merits and drawbacks are summarized in Table 2. It is imperative to thoroughly consider the distinctive and variable characteristics of each uranium deposit when selecting an in situ leaching technique for uranium mining.

Table 2. Typic characteristics of in situ leaching techniques.

Technique	Applicable Condition	Advantage	Disadvantage	Reference
Acid Leaching	Applicable to uranium deposits with low carbonate content	Low risk of groundwater contamination outside the wellfield; High leaching efficiency and short leaching cycles	Obligatory use of corrosion-resistant instruments and pipelines; Significant impact on groundwater within the wellfield; Possible formation of sulfate precipitates, causing blockage and permeability deterioration in the ore body	[37,48,64,78,87,119]

Technique	Applicable Condition	Advantage	Disadvantage	Reference
Alkaline Leaching	Widely applicable to uranium deposits with high carbonate content; Not suitable for uranium deposits with high pyrite content	Utilization of common equipment and pipelines	Low leaching efficiency and long leaching cycles; High risk of groundwater contamination outside the wellfield; Formation of carbonate or sulfate precipitates, potentially causing deposit clogging	[90]
Neutral Leaching	Wide applicability with no apparent restrictions	Leaching solution with gentler components for enhanced environmental friendliness; Simultaneous uranium mining with CO <sub>2</sub> utilization and storage for CO <sub>2</sub> -O <sub>2</sub> leaching	Possibility of gangue mineral dissolution and carbonate precipitation leading to deposit clogging due to pH drop	[2,99,109]
Bioleaching	Wide applicability, especially suitable for uranium deposits rich in pyrite and sulfides	High leaching rate and high overall leaching efficiency; Sustainable and environmentally friendly	Initial acid consumption must be considered until microbial oxidation of reducible sulfur compounds initiates acid production; Potential clogging due to gangue mineral dissolution and secondary precipitation resulting from the generation of $H^+$ and $SO_4^{2-}$	[75,96,111,120]
	Acid Leaching Di in	ssolution of U(VI) in the acid solution: ssolution of U(IV) in the acid solution the presence of an oxidizing agent:	$UO_{3} + 2H^{+} \rightarrow UO_{2}^{2+} + H_{2}O$ $UO_{2} + O + 2H^{+} \rightarrow UO_{2}^{2+} + H_{2}O$ $UO_{2} + 3CO_{2}^{2-} + H_{2}O \rightarrow UO_{2}(CO_{2})^{4-} + 2OH^{-}$	

#### Table 2. Cont.

Alkaline

Leaching

Neutral

Leaching

In-situ Leaching

of Uranium



Figure 5. Classification and fundamental principles of in situ leaching techniques.

Dissolution of U(IV) in the alkaline solution  $UO_2 + 2CO_3^{2-} + 0.5O_2 + H_2O \rightarrow UO_2(CO_3)_2^{2-} + 2OH^{-1}$ 

HCO<sub>3</sub>-

sources

CO<sub>2</sub>-O<sub>2</sub> leaching provides

CO<sub>2</sub> in water:

the ore:

HCO3- through dissolution of

Weak acid leaching provides  $2H^+ + CO_3^{2-} \rightarrow H_2O + CO_2$ HCO<sub>3</sub><sup>-</sup> through reacting initially injected sulfuric acid  $CO_2 + H_2O \leftrightarrow H_2CO_3$ 

with carbonate minerals in  $H_2CO_3 \leftrightarrow H^+ + HCO_3^-$ 

 $CO_2 + H_2O \leftrightarrow H_2CO_3$ 

 $H_2CO_3 \leftrightarrow H^+ + HCO_3^-$ 

# 4. Technological Innovations in In Situ Leaching

in the presence of an oxidizing agent:

Dissolution of U(VI) in the HCO3- solution:

Dissolution of U(IV) in the HCO3- solution in

 $UO_2 + 3HCO_3^- + 0.5O_2 \rightarrow UO_2(CO_3)_3^{4-} + H_2O + H^+$ 

 $UO_3+2HCO_3^-\rightarrow UO_2(CO_3)_2^{2-}+H_2O$ 

the presence of an oxidizing agent:

Considering the distinct variations among different uranium deposits, it proves challenging to devise a universally applicable in situ leaching strategy for all scenarios. While experiences from similar deposits offer insights, personalized technological procedures and parameter adjustments tailored to the unique characteristics of each deposit are necessary

to achieve maximum leaching efficiency. Therefore, recent technological innovations in in situ leaching have predominantly focused on customized approaches designed based on the specific features of individual uranium deposits.

# 4.1. Permeability Modification Technique for In Situ Leaching

In situ leaching relies on seepage of leaching solution in the porous media of uranium deposits [121]. As a result, a key element determining the usability of this mining method and a major barrier to its widespread adoption is the permeability of the uranium deposit [122]. In situ leaching techniques are typically considered unsuitable for uranium deposits with low inherent permeability (<0.5 m/d) [123]. To address this challenge, many scholars have concentrated their studies on permeability modification techniques for low-permeability uranium deposits, thereby broadening the application of in situ leaching [123–126]. While hydraulic fracturing, a widely used approach for enhancing permeability in oil and gas reservoirs [127,128], has been proven ineffective for uranium deposits [124], blasting-enhanced permeability (BEP) has emerged as a promising and effective technique for this specific context [125]. The schematic diagram in Figure 6 illustrates the underlying principle of the BEP technique for enhancing the permeability of uranium deposits. Experimental and simulation methods have substantiated that blasting can initiate well-connected fracture networks. The creation of sustainable and large-scale seepage channels within the networks is a crucial factor in improving the permeability of low-permeability uranium deposits. This enhancement enables subsequent applications of in situ leaching and facilitates the flow of leaching solution [125]. This application of BEP in in situ leaching for uranium mining is also referred to as in situ blasting leaching [126] or in situ fragmentation leaching [129].



Figure 6. Principle of blasting-enhanced permeability (BEP) technique for uranium deposit.

When applying the BEP technique to low-permeability uranium deposits, a thorough assessment of the natural burial conditions of the uranium deposit is crucial. The appropriate blasting method should be selected based on the burial depth of the deposit [129]. For shallow deposits, either drilling-blasting [130] or chamber-blasting [131] methods can be employed. Usually, drilling blasting is preferable for shallow-buried thick deposits due to its more pronounced cost-effectiveness [132]. Conversely, deeper deposits often require chamber blasting to ensure the generation of a larger space for relieving underground pressure post-blasting. Subsequently, specific parameters for the blasting process are further determined based on the morphology, occurrence, and thickness of the ore body. Finally, the leaching solution is selected from the previously mentioned acid, alkaline, neutral, or bioleaching solutions, considering the actual mineral composition and properties of the ore rocks. The technique improves the permeability of uranium ore by pre-crushing the rock, allowing previously unsuitable deposits for in situ leaching to effectively utilize this method [123].

Significantly, the BEP technique enhances the permeability of the deposit and reduces the size of mineral particles [133], contributing to a notable improvement in subsequent in situ leaching effectiveness. Enhanced permeability in the deposit facilitates increased flow rates of the leaching solution through the pore spaces, enhancing uranium dissolution efficiency and leaching solution transfer efficiency [104,134–138]. Meanwhile, smaller mineral particles with larger surface areas favor the contact and reaction between the leaching solution and uranium minerals.

In addition to the intrinsic characteristics of the uranium deposit, the effectiveness of permeability modification by the BEP technique is influenced by blasting-related parameters such as shock wave [139,140], blasting stress [141,142], and water-decoupling coefficient [143,144]. Therefore, it is necessary to customize the blasting-related parameters according to the specific conditions of each uranium deposit. This entails conducting pre-simulation assessments of the in situ leaching effects of blasting using established numerical models [123,125] and making necessary adjustments to related parameters so as to achieve the optimal leaching effect.

# 4.2. Prediction Technique for Fluid Flow and Geochemical Reaction for In Situ Leaching

The in situ leaching process of uranium deposits involves intricate fluid flow and geochemical reactions. The fluid flow within the porous media of the ore body, as well as the reactions between the leaching solution and uranium minerals within the ore body, undergo dynamic changes that significantly impact in situ leaching efficiency. To comprehend these dynamic changes more effectively, some scholars have proposed utilizing reactive transport models (RTMs) tailored to the specific characteristics of different uranium deposits to predict these dynamic variations accurately [145,146].

The reactive transport model plays a crucial role in studying the behavior of solutes in the subsurface environment [147–149]. Its wide application extends to predicting fluid behavior in porous media during petroleum and natural gas production [146,150], as well as in the sequestration of carbon dioxide in saline aquifers [151]. In recent years, this technique has progressively been applied to the in situ leaching of uranium deposits. When studying fluid flow and geochemical reactions in uranium deposits using reactive transport models, it is convenient to choose mature commercial and open-source software such as PHREEQC (version 3) [152,153], MT3DMS (version 5) [154], TOUGHREACT (version 4) [155], and Geochemist's Workbench (version 6) [156] to directly establish and simulate research models. In cases where customized or advanced simulations are required, it is necessary to utilize programming languages such as Python (version 3), MATLAB (version 2010), and R (version 4) to write code for more flexible control over model implementation and simulation.

In laboratory research focused on in situ leaching for uranium mining, column experiments are commonly employed [157,158]. These experiments utilize tall column-like containers to hole uranium ore samples, simulating the actual in situ leaching process by injecting leaching solutions into the column. Such experiments help researchers assess the flow of leaching solutions and the migration of uranium-bearing pregnant solutions, providing valuable insights into the effectiveness of in situ leaching. In 2019, Laurent et al. proposed a one-dimensional reactive transport model for column experiments [111]. This approach thoroughly integrates the hydraulic properties of the leaching solution. Grounded in chemical reaction kinetics, this reactive transport model also considers the influence of grain size, providing nuanced insights into the dynamics of the leaching process. Although limited to 1D laboratory experiments, this methodology offers a deeper understanding of the hydrological and chemical processes occurring during in situ leaching. Afterward, Lagneau et al. exemplified the application of the reactive transport model in practical in situ leaching of uranium deposits [159]. By employing a reactive transport model, they precisely fitted historical data for 61 wells in one uranium block and subsequently assessed and predicted results for another block. This demonstration underscores the robustness of the model in real-world production scenarios. In 2022, Collet et al. developed a threedimensional reactive transport model to comprehensively simulate coupled hydrodynamic and geochemical processes during in situ leaching [35], aiming to predict production outcomes. This 3D reactive transport model is based on the HYTEC program code [160,161]. It employs actual data from uranium deposits, incorporating deposit hydrodynamic parameters and mineral descriptions as a 3D geological model for hydrological processes. The simulation integrates geochemical processes and relevant mineralogical databases (including kinetic and mineral databases, along with underlying chemical processes) as a geochemical model. Finally, specific parameters of operational conditions (such as well placement, leaching solution composition, injection and extraction rates, etc.) serve as input parameters for coupled simulations of hydrodynamic and geochemical processes. This comprehensive reactive transport model facilitates fitting historical data and predicting future production in actual uranium mining scenarios. It accurately considers the details of practical in situ leaching, including realistic mineral balances, dissolution rates, and recovery rates. It has been successfully applied in large-scale, real-world, in situ uranium mining production, demonstrating precise predictive capabilities. Furthermore, reactive transport models are beneficial in the context of  $CO_2$ - $O_2$  leaching, which entails more complex mechanisms. They can serve to quantitatively elucidate site-specific geochemical processes during leaching and also aid in comprehending the storage of  $CO_2$  as a gas phase due to capillary mechanisms in the permeable pores of uranium deposits [98]. This dual function enhances insights into the effectiveness of CO<sub>2</sub>-O<sub>2</sub> leaching and its long-term environmental impact in the context of CO<sub>2</sub> utilization and storage.

Reactive transport models, in addition to their capability to predict production based on uranium deposit characteristics, are also employed for evaluating downgradient transport at in situ leaching sites. This aids in optimizing management decisions and facilitating groundwater remediation post-in situ leaching [162,163], contributing to maximizing returns and ensuring the sustainable development of uranium mining through in situ leaching.

# 4.3. Information Technology for In Situ Leaching

In order to enhance the efficiency and sustainability of the entire in situ leaching process for uranium mining, a data-driven and intelligent information system with comprehensive analytical capabilities has become a notable innovation. The system offers thorough information on geological, technical, and economic elements to optimize the development and operation of mining sites. It is specifically developed to facilitate intelligent management and decision-making during the in situ leaching of uranium deposits.

The Seversk Technological Institute of the National Research Nuclear University (MEPhI) developed an informational support software package specifically designed to manage the in situ leaching process for uranium mining [164]. Operating on client–server technology, the software facilitates interaction between client programs and data storage through SQL queries, making it applicable at any stage of in situ leaching operations. There are seven interconnected information systems within the software package: (1) the mininggeological information system (MGIS) collects and processes raw geological data, generates 2D/3D mathematical models, calculates uranium reserves, and visually presents the production layer's information status; (2) the technological information system constructs a model for the geological-technical mining complex, coordinates technical data processing, evaluates relationships, and generates operational reports to ensure data integrity; (3) the geo-technological modeling system simulates in situ leaching and pollutant migration using geological mathematical models of the uranium deposit and numerical models of the mining complex; (4) the geo-information expert analytical system (GEAS) visualizes all information in the entire mining operation process, analyzes hydrodynamic flow in the production layer, optimizes solutions, and reduces reagent usage in the in situ leaching process; (5) the techno-economic system employs economic mathematical models to calculate the economic performance of uranium mining units, including fundamental costs and other economic indicators related to unit development; (6) the computer-aided design system

designs and optimizes mining development patterns based on initial data derived from geological and mathematical models of the deposit; and (7) the mining planning system predicts and formulates mining plans for operational units, ensuring planned uranium production levels based on multifactor statistical models of productivity. The software package offers advantages such as a modular architecture, scalability, and expandability. Its optimal database structure ensures both data integrity and consistency, complemented by seamless integration mechanisms with existing enterprise information systems. The collaborative utilization of the seven information systems within the package enhances the intelligence of geotechnical enterprise management through comprehensive analysis of geological and geotechnical data, multifaceted modeling of geotechnical processes, and intelligent decision support.

The collaboration between the Seversk Institute of Technology and ARMZ Uranium Holding Company has led to further enhancements of the software package, giving rise to the Smart ISL site digital mining system [24]. This system is currently capable of managing uranium production through informationization, utilizing automated data collection and remote control of wellfields. It comprehensively analyzes geological and operational data, as well as hydrogeological and technical simulations. In practical production scenarios, the system optimizes processes, enhances extraction efficiency, and reduces risks. This progression toward digitalization and intelligence in in situ leaching technology provides a sustainable and efficient intelligent solution for future uranium mining.

# 5. Application Status of In Situ Leaching

In the early 1960s, the in situ leaching technique for uranium mining was developed by the former USSR and the USA [39]. Notably, in situ acid leaching technology found widespread application in the former USSR, while in situ alkaline leaching technology achieved commercial success in the USA [165]. Over several decades of continuous development and refinement, this technique has progressively seen practical on-site implementation in numerous countries worldwide [24]. Figure 7 illustrates the recoverable uranium resources under various mining methods within a mining cost of less than \$40/kgU, based on statistical data from the OECD report for 2021 [24]. It is evident from the figure that the quantity of uranium produced through in situ leaching has significantly surpassed the total quantity produced through open-pit mining and underground mining. This establishes in situ leaching as the primary method for low-cost uranium production.



Figure 7. Recoverable uranium resources by different mining methods.

Based on data from OECD and other relevant sources, the basic details of representative uranium mines utilizing in situ leaching are summarized in Table 3. The additional information associated with the uranium resources of these countries employing in situ leaching for uranium mining, including identified recoverable uranium resources, current production capacity, and projected future production capacity, is provided in Figure 8.

Country	Uranium Mine	Production Capacity (tU/Year)	Start Date	Technique	Reference
Australia	Beverley and Beverley North Uranium Deposit (Four Mile Uranium Mine)	Approximately 1200	2001/2014	Acid leaching/weak acid leaching	[24,38,78]
	Honeymoon Uranium Mine	Approximately 312 (average production for three years)	2019 (resumed production)	Acid leaching	
Kazakhstan	Katco Mine (Tortkuduk and Muyunkum Deposits)	3000–4000	2009	Acid leaching assisted by RTM simulation	[24,35]
	Zarechnoye Deposit	Approximately 1000	2020	Acid leaching with valuable by-product production	
Canada	Phoenix Uranium Deposit	Approximately 2300 (expected average production for ten years)	2023	Acid leaching	[166,167]
Russia	Dular Mine (Dobrovolnoye Deposit)	700	2020	Acid leaching assisted by Smart ISL site digital mining system	[24,168]
	Khiagda ISL Operation Plant	1000	2020	Acid leaching assisted by Smart ISL site digital mining system	
USA	Smith Ranch-Highland Operation Lost Creek project	Collectively 2900	2000 2013	$CO_2$ - $O_2$ leaching $CO_2$ - $O_2$ leaching	[24,74,169]
China	Erdos Sandstone-hosted Uranium Deposit	Unknown	2020 (trial test)	CO <sub>2</sub> -O <sub>2</sub> leaching	[24]
	Songliao Sandstone-hosted Uranium Deposit	Unknown	2023 (trial test)	CO <sub>2</sub> -O <sub>2</sub> leaching assisted by RTM simulation	
India	Tummalapalle Mine	Unknown	2017	Alkaline leaching	[24,170]
Finland	Terrafame Mine (formerly Talvivaara Mine)	Unknown	2024 (trial test)	Bioleaching	[24,75,171]

Table 3. Representative in situ leaching uranium projects worldwide.

Australia possesses the world's largest uranium reserves and has a history of utilizing open-cut and underground mining in regions like South Australia, Queensland, and the Northern Territory. The transition to in situ leaching began around 2000, primarily at the Beverley Uranium Deposit [38]. This deposit officially began operations in 2001, signifying Australia's first uranium mine using the in situ leaching technique with sulfuric acid as the leaching solution. In 2005, the significant discovery of the Four Mile Uranium Deposit [78], located a few kilometers northwest of Beverley, contributed substantially to the uranium

resources in the region, amassing reserves of 28,000 tons of uranium oxide. It has officially implemented weak acid leaching for uranium mining since 2014. Collectively, these two deposits are known as the Beverley and Beverley North Uranium Deposit. The Honey-moon Uranium Mine is another representative in situ leaching project for uranium mining currently operating in Australia [172]. It is a sandstone-hosted paleochannel uranium deposit, marking the second deposit in Australia to adopt in situ leaching technology. The mine officially commenced production in 2011 after two series of acid in situ leaching trials. Following a period of suspension for care and maintenance, it resumed production in 2019.

Kazakhstan, currently the world's leading uranium-producing country, achieved a production of 21,819 tU in 2021 [24]. The country hosts 13 uranium mining projects, with 6 in commercial operation [24]. Among them, the Katco Mine stands out as the largest in situ leaching uranium mine globally, boasting an annual production ranging from 3200 tU to 4000 tU since 2019 [35]. In an effort to maximize the extraction of remaining uranium resources in this deposit, a 3D reactive transport model simulation was implemented in 2019, covering 2394 wells across 39 production blocks. Remarkably, the simulation's predictions demonstrated less than a 10% deviation from the actual production results 16 months later [35], underscoring the feasibility of this method in large-scale industrial production and its promising prospects for broader future applications. The in situ acid leaching project at the Zarechnoye Deposit in Kazakhstan stands out as another significant venture [24]. Extensive exploration work conducted in 2020 extended the identified uranium reserves of this deposit, prolonging its feasible lifespan of in situ acid leaching. Kazatomprom [173], the national atomic company overseeing the project, implemented advancements in the acid leaching technique. They introduced nanofiltration technology to separate rare metals, including scandium, rhenium, and vanadium, from the uranium co-dissolving in the acid leaching solution. These rare metals are recovered as by-products in the form of scandium oxide, ammonium perrhenate, and vanadium during the processing of the pregnant solution. This enhancement substantially increases the value of the uranium deposit, contributing to heightened economic benefits for the overall uranium mining project.

Canada started operating its first in situ leaching project at the Phoenix Uranium Deposit in 2023. The deposit, characterized as a high-grade unconformity-type uranium deposit with proved reserves of 59.7 million pounds of uranium oxide, was discovered in the Athabasca Basin in 2008 [166]. Since 2021, comprehensive assessments of hydraulic conductivity, permeability, leachability, and containment parameters have been conducted for this deposit through commercial-scale in situ leaching tests. The acid leaching solution has been adopted with an additional neutralization step, introducing a mild alkaline solution to counteract residual acidity in the leaching zone [167]. During trial production in late 2022, the mine successfully recovered and processed 14,400 pounds of uranium oxide [166]. Upon full production, the Phoenix Uranium Deposit is expected to achieve an average annual production of 8.4 million pounds of uranium oxide for the initial five years, followed by an average annual production of 3.0 million pounds of uranium oxide [166], demonstrating substantial economic extraction benefits.

Russia has integrated its indigenously developed Smart ISL Site digital mining system [24] into the operational framework of the Dular Mine and Khiagda ISL operation plant [168]. The system is centrally managed from a control complex, facilitating the monitoring and administration of hydrodynamic processes. It conducts a comprehensive analysis of geological and production data through automated data collection, automatically optimizing extraction capacity. Furthermore, the system promptly detects wells in need of repair or restoration, ensuring a smooth production procedure. The implementation of this system has significantly enhanced the effectiveness of in situ leaching at both uranium mines. It has resulted in significant reductions in operating expenses by increasing extraction rates, shortening extraction times, and using fewer reagents [24].



**Figure 8.** Overview of uranium resources and production capacities of countries utilizing in situ leaching. (a) Identified recoverable uranium resources of countries. (b) Uranium production capacities of countries for 2021. Note: The decline in uranium production in the United States was significant and attributed to the temporary suspension of mine operations at several facilities due to an unfavorable market. These data do not accurately reflect the actual production capacity of uranium mines in the United States. (c) Uranium production capacities of countries for 2030.

The United States' uranium resources generally contain a significant amount of carbonate. As a result, in situ alkaline leaching was the predominant method for early uranium mining. In recent years, the  $CO_2$ - $O_2$  leaching technique has replaced alkaline leaching as the primary technology for in situ uranium mining in the country, offering new possibilities for efficient uranium extraction under specific geological characteristics. Currently, in situ leaching uranium mines in Nebraska and Wyoming significantly contribute to the United States' uranium production [24]. In the early 1960s, the Gas Hills and Shirley Basin uranium districts in Wyoming [169] pioneered in situ alkaline leaching technology and have now transitioned to  $CO_2$ - $O_2$  leaching [74]. Present-day operational in situ  $CO_2$ - $O_2$  leaching uranium projects in the district include the Smith Ranch-Highland operation, the Lost Creek project, and the Nichols Ranch project. Among these, the Smith Ranch-Highland operation, a sandstone-hosted uranium project, has maintained stable and relatively high annual uranium production since 2000. The combined annual uranium production of Smith Ranch-Highland and Lost Creek projects reaches as high as 2900 tU/year [24].

China's uranium resources exhibit characteristics such as low endowment, small deposit sizes, and low grades. Due to the sustained low uranium prices, Chinese uranium companies underwent reorganization in 2017 and 2018, emphasizing a domestic industry with a focus on in situ leaching in northern China to align with ecological goals [24]. Recent proactive exploration efforts have confirmed substantial uranium reserves in hydrocarbonbearing basins [2,7,9,11], notably the Erdos Basin and Songliao Basin. Consequently, these sandstone-hosted uranium deposits have become the focal point of uranium mining in China. Given the high carbonate, high iron–aluminum content, and high mineralization level characteristics of these two sandstone-type uranium deposits, the  $CO_2$ - $O_2$  in situ leaching technique has been adopted and is currently in the trial test stage. In the test units of the Songliao Basin, a comprehensive understanding of the complex phenomena during CO<sub>2</sub>-O<sub>2</sub> leaching under the conditions of the uranium deposit has been achieved. This involves practical sample collection, laboratory testing and analysis, and integration with a 3D multicomponent reactive transport model simulation. Neutral  $CO_2-O_2$  leaching not only provides a more efficient approach for these low-permeability, low-grade, and high-carbonate-content uranium deposits but also contributes to achieving greenhouse gas emission reduction goals through the utilization and underground storage of CO<sub>2</sub> [2,98,101].

India's Tummalapalle uranium mine, which is situated within the South Cuddapah Basin, is a typical alkaline in situ leaching project [24,170]. This mine constitutes approximately 49% of the nation's uranium reserves. The uranium deposit is found in carbonate-hosted rock formations, predominantly composed of massive limestone, dolostone, and intra-formational conglomerate [170]. Consequently, in situ leaching at this mine employs an alkaline leaching solution primarily composed of sodium carbonate and sodium bicarbonate. In recent years, optimization endeavors within this in situ alkaline leaching project have predominantly concentrated on post-mining processing of the leaching solution, aiming to attain enhanced efficiency in the extraction of uranium resources [170].

Finland's Terrafame Mine serves as a typical example of a mine employing bioleaching technology [171]. The mine is regarded as an unconventional resource of uranium [24], characterized by metamorphosed black shale-hosted deposits containing uranium and accessory minerals such as Cu, Co, Ni, Zn, and Mn. Due to the high content of components like pyrite (FeS<sub>2</sub>), sphalerite ((Fe,Zn)S), pentlandite ((Fe,Ni)<sub>9</sub>S<sub>8</sub>), and chalcopyrite (CuFeS<sub>2</sub>) in its ore [75], it is particularly suitable for the application of bioleaching techniques in extraction. Currently, uranium bioleaching is undergoing testing using shake flasks containing samples of rock and ore deposits from four locations within this mine [75].

## 6. Challenges and Future Directions

Currently, approximately 63% of the world's natural uranium resources are mined through the in situ leaching technique [24,35]. Continuous advancements in in situ leaching techniques for uranium mining have been witnessed in recent years. These improvements have led to significant reductions in energy and material consumption, a substantial increase in productivity, and considerable decreases in direct mining costs. As a result, the uranium

mining industry has experienced noteworthy economic benefits. However, despite these advancements, in situ leaching still faces certain challenges.

For low-grade uranium deposits, larger amounts of leaching solution, extended leaching periods, and subsequent intricate concentration processes are typically required due to their lower uranium concentration, significantly impacting their economic benefits. To address this challenge, the research and development of pre-leaching beneficiation techniques for low-grade uranium deposits represent a potential avenue. Currently, Elevate Uranium Ltd is dedicated to developing a pre-leaching in situ uranium concentration enhancement process known as "U-pgrade" [24]. This initiative is specifically tailored for the low-grade uranium deposits inherent to the Namibian Marenica Uranium Deposit, primarily composed of a clay matrix, carbonate, black mica, feldspar, quartz, and other constituents. The ongoing research and development of analogous pre-leaching beneficiation processes, adaptable to various low-grade uranium deposits featuring diverse compositions, hold promise for enhancing the economic efficiency of in situ leaching mining in these deposits.

Reactive transport models have proven advantageous in the study of the in situ leaching process, providing profound insights into accurate predictions of fluid flow, liquid-rock geochemistry, and various aspects tailored to the characteristics of each reservoir. Nevertheless, it is crucial to recognize that the repeated use of reactive transport models may impose a substantial computational burden. Therefore, exploring alternative approaches, particularly iterative models based on machine learning (ML-based surrogate models), holds the promise to replace reactive transport models in predictive simulations for in situ leaching. This emerging direction represents a compelling research avenue, offering the potential for enhanced computational efficiency and predictive accuracy in the field of in situ leaching.

In the pursuit of sustainable development, a paramount goal in uranium resource mining is to maximize resource utilization while minimizing environmental impact during the in situ leaching process. This requires meticulous attention to issues such as pore plugging, waste generation, and post-mining ecological restoration associated with in situ leaching. Addressing the risk of pore plugging involves crucial steps, such as optimizing leaching solution formulations and refining the circulation injection system. Waste generation can be significantly reduced by researching and developing more advanced, efficient, economical, and environmentally friendly ion exchange resin materials for the concentration process of uranium-bearing pregnant solution. Post-mining ecological restoration of uranium deposits necessitates comprehensive actions in soil restoration, vegetation restoration, water resource restoration, and other aspects. Incorporating advanced ecological and environmental science techniques, along with sustained long-term monitoring and assessment, is indispensable to ensure the success of ecological restoration efforts [174].

Furthermore, it is noteworthy that the efficient extraction of uranium resources is impacted not only by the mining technologies explored in this study but also by related significant technologies, including uranium tailings enrichment [175,176], extraction of uranium and other valuable metal resources from enriched tailings [177,178], and techniques for reducing ore loss [179]. Through comprehensive research and enhancement of these technologies, further advancements can be achieved in addressing challenges associated with mineral resource wastage and environmental preservation.

The effectiveness of in situ leaching technology is closely associated with the selection of optimal schemes for the opening productive formations, the pattern of injection and production wells, the modes of pumping solutions, and the means of solution lifting [57]. When preparing uranium deposits for in situ leaching, the consideration of preparation parameters and methods in the mining plan is crucial. It is necessary to take into account the morphological characteristics of the deposits being mined, the hydrogeological conditions of the productive horizon, and the related geotechnical parameters to select appropriate preparation parameters, thereby achieving optimal mining results [57].

# 7. Conclusions

Uranium, as the cornerstone of nuclear energy, attracts significant attention for its pivotal role in clean and efficient energy conversion. In contrast to fossil fuels, nuclear energy generates no greenhouse gases like carbon dioxide during usage, making it a positive contributor to climate change mitigation. In the context of global clean energy initiatives, uranium resources emerge as a crucial component for achieving sustainability. Uranium mining techniques encompass open-pit mining, underground mining, and in situ leaching. Compared to the former two conventional approaches, in situ leaching technique stands out for its high extraction efficiency, environmental friendliness, and economic advantages.

The in situ leaching technique for uranium mining is classified into acid leaching, alkaline leaching, neutral leaching, and bioleaching based on the distinct properties of uranium deposits. Acid leaching is suitable for uranium deposits with low carbonate content, alkaline leaching is applicable to those with high carbonate but low pyrite content, and neutral leaching is widely applicable. Bioleaching, being a versatile method, is particularly effective for uranium deposits with high pyrite and other sulfide content.

In recent years, the in situ leaching technology for uranium mining has experienced multifaceted innovations. The blasting-enhanced permeability technique has been shown to effectively establish well-connected fracture networks with sustainable and large-scale seepage channels within low-permeability uranium deposits. This enables the utilization of in situ leaching in uranium deposits that were previously unsuitable for this method, thus facilitating the economically and environmentally sustainable extraction of uranium from such deposits. Reactive transport models showcase exceptional accuracy in predicting complex fluid flow and geochemical reactions, specifically tailored to the distinct characteristics of various uranium deposits. Furthermore, the integration of data-driven and intelligent information technologies offers intelligent optimization of the extraction process, holding promising potential for heightened production efficiency.

Globally, prominent uranium mines in Australia, Kazakhstan, Russia, Canada, the United States, China, India, and Finland serve as successful models for the effective application of in situ leaching technology. Australia, Kazakhstan, Canada, and Russia primarily rely on acid leaching with integrated technological advancements. Uranium deposits in the United States and China are characterized by high carbonate content, and they both emphasize the utilization of the neutral  $CO_2$ - $O_2$  leaching method. This allows for the economical extraction of uranium resources while effectively utilizing carbon dioxide, thereby contributing to clean energy projection and mitigating the negative environmental impact of greenhouse gas emissions. Uranium deposits with high carbonate content in India are mined using alkaline in situ leaching, while the bioleaching technique is employed in the uranium deposit characterized by high iron and sulfur content in Finland. The varied in situ leaching methods tailored to the primary composition of uranium deposits worldwide underscore the remarkable achievements of in situ leaching in improving extraction efficiency, reducing costs, and adapting to diverse geological conditions. These outcomes, serving as a robust foundation of experiences, can be applied to uranium deposits worldwide with similar characteristics, contributing to the sustainable and thriving development of the global uranium mining industry.

In situ leaching, while a promising technology for uranium extraction, confronts several challenges. To address the economic constraints of mining low-grade uranium deposits, research and development in pre-leaching beneficiation techniques represent a promising avenue. While reactive transport models are valuable for in situ leaching studies, their significant computational load poses challenges. Future directions involve exploring alternative methods, particularly iterative machine learning-based surrogate models, to enhance computational efficiency and predictive accuracy in in situ leaching simulations. Addressing environmental sustainability concerns, including pore plugging, waste generation, and post-mining ecological restoration, requires optimizing leaching solutions, employing advanced ion exchange resin materials, and applying ecological

methods with continuous monitoring to ensure the sustainability of in situ leaching for uranium mining. Moreover, advancements in tailings enrichment, strategies to minimize ore loss, and the careful consideration of preparation parameters when selecting optimal schemes are imperative for the efficient extraction of uranium resources.

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