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Review

Chelating Agents in Assisting Phytoremediation of Uranium-Contaminated Soils: A Review

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Abstract: Massive stockpiles of uranium (U) mine tailings have resulted in soil contamination with U. Plants for soil remediation have low extraction efficiency of U. Chelating agents can mobilize U in soils and, hence, enhance phytoextraction of U from the soil. However, the rapid mobilization rate of soil U by chelating agents in a short period than plant uptake rate could increase the risk of groundwater contamination with soluble U leaching down the soil profile. This review summarizes recent progresses in synthesis and application of chelating agents for assisting phytoremediation of U-contaminated soils. In detail, the interactions between chelating agents and U ions are initially elucidated. Subsequently, the mechanisms of phytoextraction and effectiveness of different chelating agents for phytoremediation of U-contaminated soils are given. Moreover, the potential risks associated with chelating agents are discussed. Finally, the synthesis and application of slow-release chelating agents for slowing down metal mobilization in soils are presented. The application of slow-release chelating agents for enhancing phytoextraction of soil U is still scarce. Hence, we propose the preparation of slow-release biodegradable chelating agents, which can control the release speed of chelating agent into the soil in order to match the mobilization rate of soil U with plant uptake rate, while diminishing the risk of residual chelating agent leaching to groundwater.

Keywords: chelator; soil uranium; mobilization; phytoextraction; slow release

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1. Introduction

Nuclear energy is a major source of low-carbon electricity and plays an important role in the achievement of carbon neutrality. According to the World Nuclear Performance Report 2021, there are 443 operational reactors with a total capacity of 3.94×10^5 MWe, and 57 reactors with a total capacity of 5.88×10^4 MWe under construction worldwide [1]. The global nuclear power generation reached 2.55×10^3 TWh in 2020, meanwhile the annual global demand for uranium (U), a primary element used in nuclear energy, has increased to 6.25×10^4 t [1,2]. At the same time, soil contamination with U has become a serious environmental issue.

There are two main sources of U in soils, i.e., natural source and anthropogenic source. U naturally occurs in soil parent materials. Due to different accumulation levels of U in different soil parent materials, the concentration of U in soils can vary greatly. For example, the average background concentration of soil U in the United States (US) is 3.5 mg/kg, and that in China is 3.13 mg/kg, whereas Portugal reaches 25.1 mg/kg [3]. The background concentration of U in the environment is usually low. However, anthropogenic activities (e.g., industrial and agricultural production and military activities) have emitted a large amount of U into the environment, resulting in distinct accumulation of U in soils [4–9]. U tailings are the main source of U in contaminated soils. For example, U contamination in the soil and groundwater within the catchment of a U mill tailings pond can be caused by the hydrological cycle around the tailings pond [10].

Although U has a long half-life and relatively weak radioactivity, it may induce chemical and radiological toxicity in organisms after long-term exposure to U-contaminated soils or U accumulation in the organism through the food chain [11–17]. On the one hand, U can induce ionic toxicity similar to that of cadmium (Cd), copper (Cu), zinc (Zn), and other heavy metals. The ionic toxicity of U increases with U concentration in the contaminated environmental substrate. On the other hand, the radiation from the decay of U can cause oral necrosis, anemia or chronic diseases of lung and kidney in mild cases, and central nervous system damage or death in severe cases [17,18]. In addition, chronic U exposure can cause chromosome aberrations and increase cancer risk [19,20]. Therefore, it is of great importance to clean up U-contaminated soils.

Among common remediation technologies for soil contamination with heavy metals and radionuclides, phytoextraction is a popular in situ technology for large-scale soil remediation due to its low cost and environmental friendliness. Although several plants (e.g., broad bean (*Vicia faba* L.), willow (*Salix smithiana* Willd.), radish (*Raphanus sativus* L.), Indian mustard (*Brassica juncea* L.), and sunflower (*Helianthus annuus* L.)) have the ability to absorb U from soils [21–25], hyperaccumulator plant species of U have not yet been discovered. In addition, plants for remediation of U-contaminated soils usually have low extraction efficiency of U, which results in a long-term remediation period and consequently hinders large-scale phytoremediation of U-contaminated soils. Apart from searching for U hyperaccumulators, numerous studies have showed that chelating agents can mobilize heavy metals and radionuclides in soils and therefore enhance plant uptake of these contaminants [26–28]. Due to the lack of U hyperaccumulators, chelating agent-assisted phytoremediation can be an effective approach for U-contaminated soil remediation.

It should be noted several previous studies found that the mobilization rate of soil U by chelating agents exceeded the plant U uptake rate, consequently causing groundwater contamination with soluble U through leaching processes as well as transient phytotoxicity [26,28]. In fact, U contamination in groundwater is also a global environmental issue. It is reported that the groundwater in India, South Korea, the US, and Zambia has been contaminated with U to a varying extent [29]. In Nambe, New Mexico, US, the maximum groundwater U concentration reached 1200 ng/mL, which is conspicuously greater than the drinking water standard (20 ng/mL) recommended by the US Environmental Protection Agency (US EPA) [30]. U in contaminated groundwater can enter into the human body via groundwater drinking pathway and consequently pose a risk to human health. Epidemiological studies found that U mainly accumulates in kidney. Long-term consumption of U-contaminated groundwater can induce kidney tubular cell death, resulting in kidney diseases [30,31]. In addition, chronic drinking of U-contaminated groundwater can cause other detrimental health effects, such as hindering bone growth [32], disrupting metabolic processes in liver [33], affecting germ cell growth [34,35], and triggering inflammatory responses in nervous system [36]. Therefore, it is necessary to tackle the secondary pollution issue due to the short-term rapid mobilization of soil U so as to achieve effective phytoremediation and ensure groundwater safety.

This review elucidates the interactions between chelating agents and U ions, and then explains the phytoextraction mechanisms and summarizes recent progresses in synthesis and application of chelating agents in assisting phytoremediation of U-contaminated soils. The major factors influencing phytoextraction efficiency are given. The environmental risks of existing chelating agents are also discussed. Finally, recent advances in slow-release technology are reviewed, with synthesis and application of slow-release chelating agents for slowing down metal mobilization in soils in order to match plant uptake rate presented. This review aims at shedding light on the promise of slow-release biodegradable chelating agents for enhancing phytoextraction of soil U. This novel approach is considered as a green and sustainable remediation technology for U-contaminated soils.

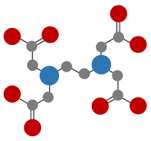
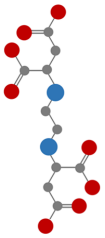
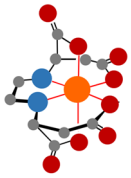
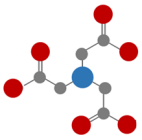
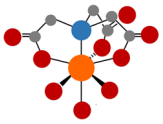
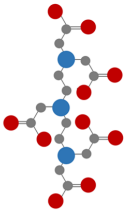
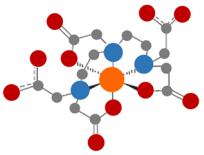
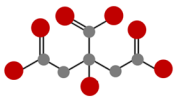
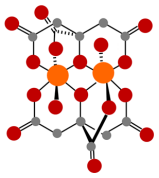
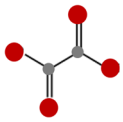
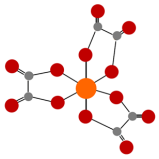
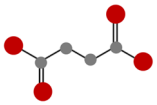
2. Interactions between Chelating Agents and U Ions

2.1. Mechanisms of Chelation

The atomic number of U is 92. There are 92 protons and 146 neutrons in the nucleus, forming four electron-filled layers (K, L, M, N) and three under-filled electron layers (O, P, Q). The six electrons ($5f^3 6d^1 7s^2$) on the three underfilled electron sublayers are valence electrons, which determine the chemical properties of U [11]. After losing six valence electrons, the outermost energy level of U is filled (S^2P^6), forming noble gas structure, which has a strong affinity with oxygen. Therefore, U usually exists in the forms of oxides and oxysalts in the crust and soil. Specifically, U is mainly present in the form of uranyl ion (UO_2^{2+}) in soils, which has a strong complexing ability to form stable complexes with phosphate (PO_4^{3-}), carbonate (CO_3^{2-}), hydroxyl (OH^-), sulfate (SO_4^{2-}), etc., and consequently reduces the mobility and bioavailability of U in soils [37,38].

Chelating agent refers to ligand which can form a cyclic structure complex with central ions (usually metal ions). Once in the soil, it can chelate with heavy metals and radionuclides to form water-soluble and exchangeable metal-chelating agent complexes, which can increase their mobility and bioavailability, and consequently promote plant uptake of these contaminants [39]. Chelating agents can be divided into two main categories including aminopolycarboxylate chelating agents (APCAs) (e.g., aminotriacetic acid (NTA), ethylenediaminetetraacetic acid (EDTA), ethylenediaminedisuccinic acid (EDDS), and N, N-Bis(carboxymethyl)-glutamic acid (GLDA)) [40,41] and low molecular weight organic acids (LMWOAs) (e.g., citric acid (CA), oxalic acid (OA), and succinic acid (SA)) [42,43]. In addition, APCAs can be classified as biodegradable and non-biodegradable [44], whereas LMWOAs are biodegradable [26,45]. The chemical structures and biodegradability of commonly used chelating agents are shown in Table 1.

Table 1. Structures and biodegradability of commonly used chelating agents, and structures and stability of chelates formed by UO_2^{2+} with these chelating agents.

Category	Chelator	Chelator Structure	Biodegradability	U Chelate Structure	logK_U *
Aminopolycarboxylate chelating agents (APCAs)	Ethylenediaminetetraacetic acid (EDTA)		Half-life > 40 d [46]		7.4 (UO_2^{2+}:EDTA = 1:1) 17.87 (UO_2^{2+}:EDTA = 2:1)
	Ethylenediaminedisuccinic acid (EDDS)		Completely degraded after 54 d [47]		10.7
	Nitrilotriacetic acid (NTA)		75% degraded after 21 d [48]		9.5
	Diethylenetriaminepentaacetic acid (DTPA)		Similar to EDTA [49]		11
Low molecular weight organic acids (LMWOAs)	Citric acid (CA)		69% degraded after 20 d [50]		8.96
	Oxalic acid (OA)		Hard to degrade in nature due to the formation of Ca-oxalate [21]		6.36
	Succinic acid (SA)		Half-life < 2 min and 200 h in soil solution and for microbial mineralization, respectively [51]		3.87

Notes: N C O UO_2^{2+} ; * Data was sourced from [26,52,53].

Cyclic chelate formed by metal ion with ligand through coordination bonds has strong stability. Table 1 shows the structures of chelates formed by UO_2^{2+} with commonly used chelating agents. The stability constant (K) is used to indicate the stability of metal chelate, which increases with K value. K value is calculated using the following equation [54–57]:

$$K_{ML}^{T} = \frac{C_{ML}}{C_M C_L}$$

where C_{ML}, C_M, and C_L are concentrations of metal chelate, metal ion, and ligand, respectively; K_{ML}^{T} is the stability constant.

It is worth noting that EDTA has the highest K values for various heavy metal chelates compared to other chelating agents (Table 1) and therefore has been widely used in mobi-

lization of metal contaminants in soils and soil washing over past decades [58]. However, the tertiary amine in EDTA molecule makes it difficult to be biodegraded. As a result, long-term retention of non-degradable EDTA in the environment has posed a critical risk to the ecology and human health [59–63]. As shown in Table 1, biodegradable EDDS has a high *K* value for U chelate. Moreover, its chemical structure is similar to that of EDTA. Thus, EDDS has become an ideal substitute for EDTA [64–67]. The *K* value of NTA for U chelate is slightly lower than that of EDDS. However, the use of NTA should be restricted due to its carcinogenic effect [44,68]. For example, iron (Fe)-NTA can induce renal cell carcinoma, which is widely used to perform oxidative tissue damage and carcinogenesis experiments [69–71]. CA, which is a natural LMWOA, also has a high *K* value for U chelate. Previous studies showed that CA increased the bioavailability of U in soils more effectively compared to other chelating agents (i.e., OA, EDDS, and NTA) [26,72].

2.2. Mobilization of Soil U by Chelating Agents

Soil texture, pH, and redox potential (Eh) are dominant factors influencing the effect of chelating agents on mobilizing U in soils [26,73–75]. Table 2 presents the mobilization efficiency of soil U by different chelating agents. The dosages of chelating agents ranged from 0.5 to 50 mmol/kg. The experimental soil pH values ranged from 5.6 to 7.3. The soil U concentration reached nearly 1000 mg/kg. Among the chelating agents listed in Table 2, the effect of CA on mobilizing U in soils, in particular, in sandy soil, was the most significant, with the exception of NH_4-citrate > CA in the loamy sand and heavy clay in the study by Duquène, et al. [26]. A possible explanation for the different effects of CA on mobilizing U in soils with contrasting textures is that the soil containing higher silt and clay contents has a larger specific surface area, which can result in stronger adsorption of contaminants and consequently reduce the mobilization efficiency of soil U by chelating agents [76,77]. In addition, there are a large number of functional groups (e.g., amino and carboxyl groups) in soils with high organic matter (OM) content, which has a strong adsorption capacity for U ions [78–80]. This can also affect the effectiveness of chelating agents in mobilization of soil U. A similar trend was observed in Jiang, et al., which showed that the mobilization efficiency of chromium (Cr) in contaminated soils by CA was in the following decreasing order: sandy soil > loam > clay soil [81]. The mobilization efficiency of soil heavy metals by chelating agents tends to increase with increasing soil acidity [82]. For example, Yang, et al., found that the mobilization efficiency of heavy metals in the soil solution with low pH by EDDS was significantly higher than that with high pH [83]. Likewise, Wang, et al., showed that the mobilization efficiency of heavy metals by chelating agents (i.e., GLDA and NTA) in the strongly acidic soil was 4-times higher than that in the alkaline soil, and the chelating agents were more effective in mobilizing heavy metals in the sandy soil compared to the clay soil at a wide soil pH range of 4 to 10 [82]. The leaching experiment of Kantar and Honeyman showed that the lower pH of eluent facilitated the mobilization of soil U by CA [84]. In addition, Eh can significantly influence the chemical forms of U present in soils and therefore control the solubility of U in the soil [73]. Specifically, at low Eh, U is present in solid phases, whereas at high Eh, U carbonates are transformed into soluble phases, which tend to migrate with water. To sum up, chelating agents can transform soil U from solid phase to soluble phase by forming high *K* value complex compounds with UO_2^{2+}. In particular, sandy soil with low pH, high Eh, and low OM content is favorable for the mobilization of U by chelating agents.

Table 2. Effects of different chelating agents on mobilizing U in soils with different textures.

Item	Reference						
	[85]	[72]	[86]	[26]			[27]
Soil characteristics							
Texture	Silty loam	Loam	Sandy loam	Sand	Loamy sand	Heavy clay	Medium clay
OM (%)	3.4	4.2	3.47	4.9	7.8	9.2	12.1
pH	6.8	7.3	5.58	6.9	7.2	6.7	6.67
Total U (mg/kg)	909	280	272	14	13	41	18
Source of U contamination	Industrial	Industrial	U mine site	Industrial	Industrial	Uraniferous shale	$UO_2(NO_3)_3 \cdot 6H_2O$
Experimental design							
Chelator	CA and OA	CA, EDTA, and MA	CA, EDTA, and EDDS	CA, EDDS, OA, NTA, and NH_4-citrate			CA and EDDS
Chelator dosage (mmol/kg)	2, 10, and 20	0.5, 1, 5, 10, and 20	0.5, 2, 2.5, 5, 10, 15, 25, and 50	5			5, 5 + 5, 10, and 10 + 10
Test conditions	Soil incubation	Soil incubation and pot experiment	Column leaching test	Soil incubation	Soil incubation	Soil incubation	Soil incubation and pot experiment
Result of most effective mobilization of soil U							
Chelator and dosage (mmol/kg)	CA 20	CA 20	CA 50	CA 5	NH_4-citrate 5	NH_4-citrate 5	CA 10 + 10
Days taken to reach the maximum U concentration in soil solution	1	1	6	1	1	1	8
Maximum U concentration in soil solution and increment compared to control	775 mg/kg, increased by 140 times	240 mg/L, increased by 200 times	2000–2400 Bq/kg, increased by 356 times	5019 μg/L, increased by 479 times	1106 μg/L, increased by 368 times	733 μg/L, increased by 366 times	1463.6 μg/L, increased by 215 times

3. Chelating Agent-Assisted Phytoremediation of U-Contaminated Soils

3.1. Mechanisms of Phytoextraction of U from Soils

Phytoextraction is a common technology for phytoremediation of U-contaminated soils, which transfers U from the soil to plant roots, stems, and leaves, and therefore cleans up the soil. Specifically, plant uptakes U^{4+} or U^{6+} ions through epidermal cells [3,87]. U cations (e.g., UO_2^{2+}) can be absorbed by roots through the same carrier or ion channel as calcium (Ca^{2+}), Fe^{3+}, magnesium (Mg^{2+}), and other necessary elements. U anions (e.g., $UO_2(CO_3)_2^{2-}$) can pass through plant cell membranes in a way similar to CO_3^{2-} [88]. For most plants, the majority of U is enriched in roots [89,90], with a small proportion of plants, such as sesbania (*Sesbania rostrata*) and water lily (*Nymphaea tetragona* Georgi) can further transfer the absorbed U upward to stems, leaves, and fruits [3].

The dominant factors affecting root uptake of soil U include chemical forms of U, soil environmental conditions (e.g., pH and Eh), and plant species [3,72,89–92]. U is mainly present in the form of UO_2^{2+} in soils. PO_4^{3-} and CO_3^{2-} are common ligands for U ions and can form uranyl phosphate, uranyl carbonate or triple uranyl-calcium carbonate complexes. The formation of uranyl phosphate and uranyl carbonate in soils usually depends on the contents of PO_4^{3-} and CO_3^{2-} in the soil [90]. Moreover, soil pH can influence U solubility and sorption or desorption in soils, and consequently affect the bioavailability of soil U. Soil OM is rich in functional groups (e.g., carboxyl, hydroxyl, and aromatic groups) containing lone pairs of electrons, which can cause the formation of different U compounds by altering soil Eh [93]. Moreover, when U is taken up by root cells, it is translocated to the mid-column before being released into the xylem [3]. The transpiration and expression of transporter protein genes can influence the translocation of U to the aboveground parts. For example, three genes including IRT1, FRO2, and FIT1 were found to affect U translocation in *Arabidopsis thaliana* L. [94,95].

3.2. Mechanisms of Assisted Phytoextraction of U from Soils

The principle of chelating agent-assisted phytoremediation of U-contaminated soils is to use chelating agents to desorb U ions from soil particle surface into soil solution, and

therefore increase the solubility of soil U. The chelating agent-mobilized U can be readily available for plant uptake and facilitate phytoextraction of U from the soil. Table 3 lists the effects of commonly used chelating agents on phytoextraction of U from soils. It can be seen that although CA has a relatively lower *K* value for U chelate than EDDS (Table 1), CA was the most effective in enhancing the phytoextraction of soil U in all phytoremediation trials presented in Table 3. This can be related to the following three reasons: (1) CA can reduce soil pH value, which may be favorable for the mobilization of soil U [72,96]. Lozano, et al., found that CA had the most significant effect on the dissolution of U with acidic pH, whereas the maximum dissolution of U with the addition of EDDS, EDTA or no chelating agent was under alkaline conditions [86]. (2) CA can form U-CA complexes, which increase the solubility and bioavailability of U in soils [91]. (3) CA can improve plant tolerance to the bioavailable U in soils [97]. Rong, et al., found that 5 mmol/kg CA addition could alleviate cell damage and improve the tolerance of perennial ryegrass (*Lolium perenne* L.) to soil U [98]. They claimed that the optimal dosage of CA can avoid adverse effects (e.g., etiolation, withering, and even death) of CA in excess amounts on plant growth as well as improve the buffer capacity of cytosol, enhance the photosynthesis of perennial ryegrass, and decrease the electrical conductivity (EC) and malondialdehyde (MDA) content while increasing the contents of soluble proteins and enhancing the activities of antioxidant enzymes in the shoots and roots. Likewise, a significant increase in antioxidant enzyme activities in the leaves of *Macleaya cordata* grown in the CA-treated soils has been noted, thereby mitigating the oxidative stress induced by U and chelating agent and facilitating the phytoextraction of soil U [27]. As shown in Table 3, the application rates of chelating agents in assisting phytoremediation of U-contaminated soils ranged from 0.5 to 25 mmol/kg. At a high application rate, the toxic effect of EDDS on plant growth was more pronounced than that of CA and OA [99]. In addition, compared to the single application at a high rate, multiple consecutive applications of chelating agents at a relatively low rate can alleviate the toxicity of mobilized U to plants and increase phytoextraction efficiency of soil U. It is well known that hyperaccumulator plant species play an important role in phytoremediation. At present, the Global Hyperaccumulator Database records 759 hyperaccumulators. However, there is still a lack of U-related hyperaccumulator plant species [100]. Therefore, the enhancement of phytoextraction of soil U as assisted by chelating agents can be a possible approach for filling the current gap of U hyperaccumulator plant species.

Table 3. Effects of different chelating agents on assisting phytoextraction of U from soils.

Chelator	Plant	Total U (mg/kg)	Source of U Contamination	Incubation Period	Dosage (mmol/kg)	Application Frequency	Result of Most Effective Mobilization of Soil U	Ref.
AA [1], CA, and MA	10 species [6]	280 and 750	Industrial	28 d of growth prior to chelator addition and harvested 7 d after addition	20	Single	CA was the most effective in enhancing U accumulation in plants. After 20 mmol/kg CA addition, shoot U concentrations in four plant species (*B. juncea*, *B. chinensis*, *B. narinosa*, and amaranth) increased by more than 1000 times compared to control within a few days.	[72]
CA, CDTA [2], DTPA, EDTA, HEDTA [3], and OA	Sunflower (*Helianthus annuus* L.) and Indian mustard (*Brassica juncea* L.)	300	Additional $UO_2(NO_3)_3 \cdot 6H_2O$	28 d of growth prior to chelator addition and harvested 42–56 d after addition	1–25	Single	CA was the most effective in desorption of U ions and enhancing plant accumulation of U. 20 mmol/kg CA addition to loamy acid soil resulted in the highest U concentration in sunflower shoots, being 150 times greater than control.	[89]

Table 3. *Cont.*

Chelator	Plant	Total U (mg/kg)	Source of U Contamination	Incubation Period	Dosage (mmol/kg)	Application Frequency	Result of Most Effective Mobilization of Soil U	Ref.
AC [4], CA, EDDS, NTA, and OA	Indian mustard (*Brassica juncea* cv. Vitasso) and ryegrass (*Lolium perenne* cv. Melvina)	14 41	Industrial U-contaminated soil (sand, labelled as BK1) Natural U-enriched soil (heavy clay, labelled as BI)	28 d of growth prior to chelator addition and harvested 4–14 d after addition	5	Single	The Indian mustard shoot U concentration was in the following decreasing order: EDDS > AC > CA > OA > NTA in BK1; and CA > AC > EDDS > OA = NTA in BI. The ryegrass shoot U concentration was in the following decreasing order: CA > AC > OA > EDDS > NTA in BK1; and CA = AC > OA = EDDS > NTA in BI.	[21]
AC and CA	Kochia (*Kochia scoparia* L. Schrad.), sunflower (*Helianthus annuus* L.), and sweet corn (*Zea mays* L.)	78.7	Depleted U-contaminated soil	45 d of growth prior to chelator addition and harvested 6 d after addition	20	Single	AC was as effective in enhancing plant uptake of U as CA. The kochia leaves U concentration was in the following decreasing order: CA > AC, whereas that in the stems: AC > CA. The sunflower leaves U concentration was in the following decreasing order: AC > CA, whereas that in the stems: AC = CA. Corn had the lowest U uptake capacity and was considered as a negative control.	[101]
CA and MA	Indian mustard (*Brassica juncea* L.)	100	Additional $UO_2(NO_3)_3 \cdot 6H_2O$	66 d of growth prior to chelator addition and harvested 7 d after addition	5, 10, and 20	Single	A total of two restorations were carried out. In the first round, the maximum plant accumulation of U was noted in 10 mmol/kg CA treatment after 45 d and in 20 mmol/kg CA treatment after 55 d. In the second round, the maximum plant accumulation of U was noted in 20 mmol/kg MA treatment after both 55 and 65 d.	[102]
CA and mixture of CA, MA, OA, and LA [5] (labelled as Mix)	Mustard (*Brassica juncea var. tumida*)	47.75	Additional $UO_2(NO_3)_3 \cdot 6H_2O$	60 d of growth prior to chelator addition and harvested 7 d after addition	5 for CA and molar ratio of CA: MA: OA: LA in Mix = 2.5:2.31:1.15:0.044	Single	Mix was more effective in enhancing U accumulation in mustard.	[103]
CA, EDDS, and OA	Red wetter grass (*Zebrina pendula* Schnizl)	150	Additional $UO_2(CH_3CO_2)_2 \cdot 2H_2O$	90 d of growth prior to chelator addition and harvested 7 d after the last addition	2.5, 5, and 7.5	Three consecutive applications every three days	CA was the most effective in enhancing U accumulation in *Zebrina pendula* Schnizl. Addition of 5 mmol/kg CA resulted in the highest U concentration in the plant, being 5.7 times greater than control.	[99]
CA, EDDS, and OA	*Macleaya cordata*	18	Additional $UO_2(NO_3)_3 \cdot 6H_2O$	Chelator addition at the beginning of flowering and harvested 14 d after the last addition	5 and 10	Except for 10 mmol/kg EDDS, all other chelators were applied both in a single application and two consecutive applications.	The enhanced U uptake was in the following decreasing order: CA > EDDS > OA. Two consecutive applications of 10 mmol/kg CA resulted in the most significantly promoted solubilization of soil U, being 215 times greater than control.	[27]
CA, EDDS, and OA	Sunflower (*Helianthus annuus* L.)	15	Additional $UO_2(CH_3CO_2)_2 \cdot 2H_2O$	60 d of growth prior to chelator addition and harvested 7 d after the last addition	2.5, 5, and 7.5	Three consecutive applications every three days	5 mmol/kg CA addition resulted in the highest U phytoextraction efficiency, being 1.78 times greater than control.	[28]

Notes: [1] AA: Acetic acid; [2] CDTA: Trans-1,2-diaminocyclohexane-N,N,N′,N′-tetraacetic acid; [3] HEDTA: N-hydroxyethlenediaminetriacetic acid; [4] AC: Ammonium citrate; [5] LA: Lactic acid; [6] 10 species: Amaranth (*Amaranth cruentus* L.), Indian mustard (*Brassica juncea* L.), bush bean (*Phaseolus vulgaris* L.), Chinese cabbage (*Brassica chinensis* L.), Chinese mustard (*Brassica narinosa* L.), corn (*Zea mays*), cow pea (*Pisum sativum* L.), field pea (*Pisum sativum* L.), sunflower (*Helianthus annuus* L.), and winter wheat (*Triticum aestivum* L.).

3.3. Environmental Risks

Chelating agents can rapidly increase the mobility of soil heavy metals and radionuclides through chelation once applied to the soil. However, during the short period of rapid mobilization of soil heavy metals and radionuclides, plants may only be able to absorb a small proportion of the mobilized metal ions in the soil, whereas the remaining large amount of mobilized metal ions exists in the soil and may cause contamination to

subsurface soil and groundwater through leaching [60,62,63,104]. In addition, a sudden short-term release of bioavailable and substantial heavy metals and radionuclides in the soil may cause transient phytotoxicity and consequently inhibit plant growth [97,105–108]. For example, Chen, et al., compared the growth of sunflowers in soils treated with EDDS, CA, and OA at high (7.5 mmol/kg), medium (5 mmol/kg), and low (2.5 mmol/kg) application rates [28]. They found that all chelating agents reduced sunflower biomass, and a greater reduction was noted in soils treated with higher dosage of chelating agents. Whereas Römkens, et al., found that although EDTA increased the concentrations of bioavailable heavy metals in the soil, the plant uptake of heavy metals did not exhibit an increasing trend after EDTA application [109]. This could be attributed to the limitations in plant uptake capacity for and tolerance to U, beyond which plant growth and uptake of soil bioavailable U will be affected. Similar results were obtained by Hou, et al., which showed that the germination of tomato, cabbage, and radish seeds was significantly inhibited at a soil U concentration greater than 320 mg/kg [24]. Therefore, plants have a limited uptake capacity for soil U, and excess amounts of mobilized U in soils in a short period can be toxic to plants.

In addition, chelating agents can influence the soil ecosystem by affecting soil microbial metabolism. For example, additional chelating agents (e.g., EDTA and EDDS) can affect the activity of soil microorganisms, especially dehydrogenase activity and basal respiration [110]. Lee and Sung found that EDTA inhibited the microbial activity in heavy metal-contaminated soils [111]. Soil microorganisms depend directly or indirectly on soil solutions to absorb food and water, and increased concentrations of bioavailable metals in soils may poison microorganisms [109]. Nevertheless, Cao, et al., found that biodegradable chelating agents (e.g., EDDS and methylglycinediacetic acid (MGDA)) could alleviate soil heavy metal stress and therefore benefit the bacterial community in the soil [112]. Thus, varying chelating agents may bring different effects on the soil ecosystem.

Barona, et al., found that EDTA increased the leachability of heavy metals in the soil and led to a weak adsorption of metal ions by soil compounds, which contributed to phytoextraction [113]. The study of Udovic and Lestan showed that the mobility of residual lead (Pb) in the EDTA-washed soil increased during the soil aging process [114]. The column leaching experiment of Wu, et al., also demonstrated that Cu, Zn, and Pb migrated into the leachate with rainwater after EDTA application to the soil, and the heavy metal concentrations in the leachate increased linearly with increasing rainfall [60]. This indicated that the migration of soil heavy metals increased by EDTA, which was conducive to phytoextraction. However, if the mobilized heavy metals in soils were not absorbed by plants in time, they may migrate to groundwater with rainwater, and consequently cause more serious environmental problems. Huang, et al., found that DTPA contributed more significantly to the leakage of soil Pb than acid rain under planting conditions, posing a great risk of groundwater pollution [115]. Nowack, et al., elucidated the unavoidability of chelated metal leaching by preferential flow processes during the chelating agent-enhanced phytoextraction [104]. They pointed out that the application of chelating agents to soils should be limited to areas where the connection to groundwater has been broken, or where groundwater contamination is not an issue. Chang, et al., found that CA enhanced the accumulation of U in crop plants (i.e., edible rape, Indian mustard, canola, and sunflower), whereas additional CA could lead to groundwater pollution with downward U migration [97]. Additionally, CA promoted the phytoextraction of U from the soil in the short term, whereas the additional U-CA complexes could be converted into less available forms (e.g., U trioxide) after dissociation in the long term, which could consequently reduce the phytoextraction efficiency after the first few croppings [97]. Thayalakumaran, et al., found that Cu leaching was much greater than herbaceous uptake after EDTA application [116]. Therefore, when using chelating agents to assist phytoremediation of U-contaminated soils, it is necessary to consider the reasonable application rate and precipitation during the application.

4. Slow-Release Chelating Agents

4.1. Slow-Release Technology

Slow-release technology, which aims at extending the release time of a substance, was first used in the pharmaceutical industry and has since been applied to synthesize a wide range of slow-release materials such as slow-release fertilizers, slow-release pesticides, and food packaging [117–119]. Controlled-release technology is also a way to extend the release time, which allows the core material to be released at a specific rate or concentration level [120,121]. There is a slight difference between the two technologies, but essentially both extend the release time of core material, so this paper does not make a detailed distinction between the two. Slow-release fertilizers are designed to control and slow down the release speed of nutrients so as to reduce fertilization as well as nutrient loss and increase crop yield [119]. In the food packaging industry, the quality of food products for long-term storage is maintained or improved by slow release of substances such as antimicrobials, antioxidants, enzymes, and spices [122–124]. Although used in different industries, slow-release materials usually consist of three parts, i.e., core material which is the active substance to be released, wall/carrier material which can regulate the release speed of core material, and binding agent which forms the core material and wall/carrier material into a cohesive whole.

There are four common approaches which can be used to achieve slow-release effect, including blending, multilayer composite-controlled release, multi-hole adsorption carrier, and microencapsulation [125,126]. To be specific, the blending approach uses the principle of chaotic convection in fluid mechanics to prepare different forms of polymers and films [127]. The multi-layer composite controlled-release approach originated from the layer-by-layer coating (LBL) approach in the pharmaceutical field. Han and Floros first proposed the concept of multi-layer composite controlled-release approach, which is mainly composed of internal controlled-release layer, intermediate active film layer, and external barrier layer [128]. The inner layer is used to control the diffusion rate of active substance to food surface, the intermediate matrix layer contains the active substance, and the outer layer serves to prevent the extravasation loss of active substance. Multi-hole adsorption carrier controls the drug release speed by modulating and modifying the porous material to allow selective adsorption of target molecules. Microencapsulation uses coating material to encapsulate the core material to form particles less than 1000 μm in diameter [129,130]. Microencapsulation is popular due to its simple operation and inexpensive equipment. The common methods for preparing slow-release microcapsules include emulsion solvent evaporation [131], spray drying [105], emulsion dispersion [132], chemical crosslinking, ion exchange, and complex coalescence [133].

Slow-release technology has recently received growing attention in environmental pollution treatment. In in situ chemical remediation, slow-release technology can control the release of active compounds, reduce the non-selective consumption of oxidants, and maintain the treatment effect for a longer period [134]. For example, Christenson, et al., prepared a slow-release oxidant by heating the mixed paraffin and potassium permanganate and cooling them in the mold [135]. Their five-year field-scale test showed that the slow-release oxidant gradually reduced trichloroethene (TCE) in a low permeable aquifer by 89% and was only refurbished yearly. Tang, et al., used chitosan and urea as carriers to prepare sodium persulfate slow-release material, which can prolong the generation time of free radicals and improve the degradation of OM [136]. In bioremediation, slow-release technology can continuously provide substrate, improve the removal efficiency of pollutants, and maintain a longer period of anaerobic dechlorination [134]. For example, Tsai, et al., reported that the slow-release material prepared by vegetable oil, cane molasses, and surfactants, provided nutrients for microorganisms and changed the subsurface environment to anaerobic conditions, which were conducive to reductive dechlorination. The removal efficiency of TCE in the contaminated groundwater reached 99% after 50 d [137]. In the study of Yeum, et al., slow-release precipitating tablets and slow-release floating tablets,

both prepared from hydroxypropyl methylcellulose, were applied to in situ biological denitrification systems to provide a continuous carbon source for microorganisms [138].

4.2. Slow-Release Chelating Agent-Assisted Phytoremediation

To address the current bottleneck problem that the rapid mobilization rate of soil heavy metals and radionuclides by chelating agents does not match the plant uptake rate of bioavailable metal ions in the soil [28,105], slow-release chelating agents have been developed to control the release speed of chelating agent so as to avoid a sudden increase in soil bioavailable heavy metals and radionuclides which exceed plant tolerance and consequently cause phytotoxicity. The mechanisms of slow-release chelating agent-assisted phytoremediation of heavy metal- or radionuclide-contaminated soils are illustrated in Figure 1. As shown in Figure 1, the chelating agent as core material is wrapped by degradable wall material. After being applied to the soil, the wall material will gradually degrade meanwhile the internal chelating agent can be released to form chelates with metal ions in the soil, which can increase their solubility and therefore facilitate plant uptake of the contaminants.

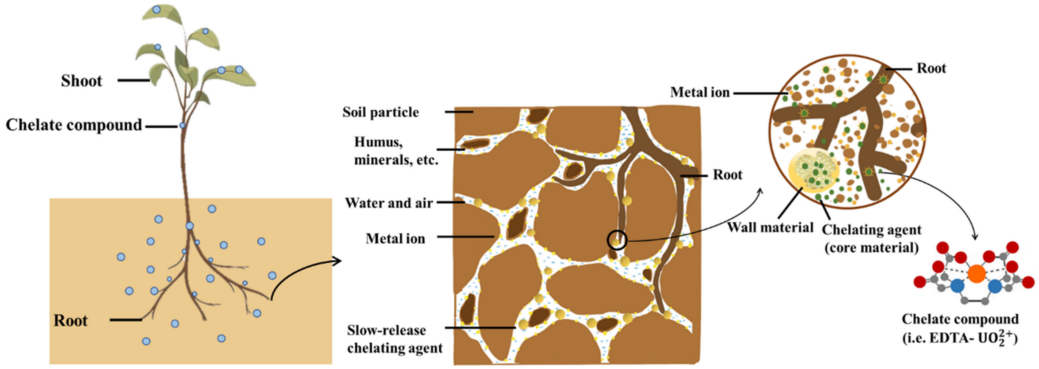


Figure 1. Mechanisms of slow-release chelating agent-assisted phytoremediation of heavy metal- and radionuclide-contaminated soils.

Slow-release chelating agents and slow-release fertilizers have conspicuous similarities in their slow-releasing behaviors. Based on the comparatively mature preparation methods for slow-release fertilizers, slow-release chelating agents can also be prepared in two main ways, i.e., one method is to synthesize slow-release carrier and then use diffusion mechanism to import chelating agent into the carrier in liquid form (e.g., [139–141]). The other method is to use granular solid chelating agent as core material, and polymer organic or inorganic materials is selected as wall material to encapsulate chelating agent particles inside the slow-release chelating agent (e.g., [105,131,142,143]).

Until now, most studies have used synthetic organic polymers (e.g., polyethylene, polycaprolactone, and ethylene-vinyl acetate copolymer) as wall/carrier material to prepare slow-release chelating agents. However, these synthetic materials can be harmful to the environment. In contrast, biodegradable natural organic polymers (e.g., starch, cellulose, and chitosan) and organic-inorganic hybrid materials (e.g., acrylamide + montmorillonite and cellulose + zeolite) are ideal wall/carrier materials, which have been used to synthesize slow-release fertilizers. Current research on slow-release chelating agents with biodegradable materials as wall/carrier material is relatively limited. Very recently, Wang, et al., prepared a slow-release chelating agent with chitosan and cyclodextrin as carrier materials and CA as core material [141]. In addition, considering the similarity between slow-release chelating agents and slow-release fertilizers, some wall/carrier materials applied to slow-release fertilizers (e.g., starch, cellulose, and chitosan) may also be ideal for the synthesis of slow-release chelating agents. Furthermore, slow-release materials can cause a burst-release effect after application in the water and soil environment [144,145]. The burst-release effect can be induced when a large amount of core material is released after the shell of slow-release material with a shell-core structure degrades or the shell

breaks down due to the increased internal pressure. To mitigate the above disadvantage, polymers, such as polyvinyl alcohol (PVA) and gelatine, can be added to increase the toughness of outer wall and enable it to withstand a certain amount of internal pressure. Zhang, et al., prepared a biodegradable carboxymethylcellulose and PVA blended film. They found that the water permeability of the film could be adjusted by changing the ratios of PVA and formaldehyde [146]. If this film was applied to slow-release chelating agents, the slow-release period could be prolonged. Han, et al., prepared a blended film from starch and PVA that is biodegradable and has good compatibility [147], which may also be used as a slow-release carrier for slow-release chelating agents.

It is worth noting that the aforementioned organic polymers and inorganic materials have been used in preparation of adsorbents [148–152], suggesting that these materials may promote the adsorption of some metal ions onto the wall/carrier material, leading to a temporary passivation effect. Kos, et al., used EDDS and EDTA in conjunction with acrylamide hydrogel to assist the phytoextraction of soil Pb [153]. They found that 10 mmol/kg EDDS addition significantly increased the leachable Pb, whereas 5 mmol/kg EDDS addition not only increased the phytoextraction efficiency of soil Pb, but also effectively controlled the leaching of soil Pb. Hydrogel can be a promising carrier for the synthesis of slow-release chelating agents as the chelating agent can be released into the soil through water loss from the hydrogel. Moreover, hydrogel can maintain the soil nutrient balance by retaining water in the soil. Hydrogel-based slow-release fertilizers have also been reported by other researchers [154–158]. If the functions of nutrient supply, water retention, and controlled release of chelating agent were incorporated into hydrogel-based slow-release chelating agents, chelating agent-assisted phytoextraction technology could be significantly advanced.

Until now, only a few studies have examined the effectiveness of slow-release chelating agents in assisting phytoremediation of heavy metal-contaminated soils, whereas its application in phytoremediation of U-contaminated soils has been scarce. Table 4 lists slow-release chelating agents which have been used to remediate heavy metal- and radionuclide-contaminated soils in recent years. As shown in Table 4, the particle sizes of slow-release chelating agents ranged from 5–20 μm to 3–5 mm. EDTA is the most commonly used chelating agent in these studies. The slow-release period of these chelating agents ranged from several days to months, which is mainly related to the particle size of chelating agent and properties of wall/carrier material. Among the chelating agents listed in Table 4, those with higher molecular weight organic compounds as the wall/carrier material possessed a longer slow-release period. Recently, the first biodegradable slow-release chelating agent with CA as core material has been prepared by Wang, et al., who used carboxymethyl-β-cyclodextrin (CMCD) and hydroxypropyl chitosan as carriers [141]. They found that the novel slow-release chelating agent had a noticeable slow-release effect on CA, which could reduce the impact of soil bioavailable U on Indian mustard (*Brassica juncea* L.) growth and promote the absorption and accumulation of U in the plant. However, compared with the slow-release chelating agent (i.e., microencapsulated EDTA) in Xie, et al., the release rate of the aforementioned slow-release CA in water is faster [131]. In addition, due to the lack of long-term soil incubation experiment, the dynamic interactions between the slow-release CA and U as well as other metals in the soil remain unclear. Therefore, the slow-release period of slow-release CA prepared by Wang, et al., in soils is hard to be estimated [141].

Table 4. Components, synthesis methods, particle size, and slow-release performance of slow-release chelating agents applied in remediation of heavy metal- and radionuclide-contaminated soils.

Wall/Carrier Material	Core Material	Synthesis Method	Particle Size	Slow-Release Period	Contaminant	Dosage (mmol/kg)	Test Conditions	Ref.
Silicate	EDTA-Na_2	Spray drying	3–5 mm	Release of EDTA from slow-release EDTA was still significantly lower than from uncoated solid EDTA in soil after 18 d.	Pb and Zn	13	Pot experiment	[105]
Talc, polyethylene, ethylene-vinyl acetate copolymer, ethylene-octene-1 copolymer, and polyoxyethylene monomethyl ether	EDTA-Na_2	Coating	-	Release of 75% EDTA ranged from 3 to 210 d in water	Pb	4	Pot experiment	[142]
Diethylenetriamine and hexamethylene diisocyanate	EDTA-Na_4	Interfacial polymerization	5.78 μm in average	Release of 85% EDTA in water after 5 d	-	-	Pot experiment	[143]
Polycaprolactone	EDTA	Solvent evaporation	65 ± 15 μm	Release of 93% EDTA in water after 30 d	Cu and Pb	3 and 6	Pot experiment	[131]
Chitosan and its derivatives	EDTA-Na_2	Spray drying	5–20 μm	Release of EDTA from slow-release EDTA was still significantly lower than from non-microencapsulated EDTA in water after 3 d.	Cd, Cu, and Pb	4 and 8	Pot experiment	[159]
Hydroxypropyl chitosan-graft-carboxymethyl-β-cyclodextrin	CA	Spray drying	-	Cumulative release of 80% CA in water after 2 d	U	5	Pot experiment	[141]

Application rate of slow-release chelating agents is one of the most important factors influencing mobilization of soil U. As summarized from Table 3, the application rates of chelating agents alone in assisting phytoremediation of U-contaminated soils ranged from 0.5 to 25 mmol/kg. In addition, previous studies reported that chelating agents (e.g., EDDS, EDTA, and DIPA) reduced plant biomass and even caused plant mortality at application rates greater than 5–7.5 mmol/kg [27,28,160,161]. The impact of chelating agents on plant growth is also related to the types of chelating agent and plant. For example, Chen, et al., found that 7.5 mmol/kg EDDS addition significantly inhibited the growth of *Zebrina pendula* Schnizl and sunflower (*Helianthus annuus* L.) [28,160]. Hu, et al., also noted some toxic symptoms such as chlorosis and necrotic spots occurred during *Macleaya cordata* growth after the addition of 5 and 10 mmol/kg EDDS [27]. In contrast, under the same experimental conditions, the aforementioned plants exhibited much higher tolerance to CA treatments. As shown in Table 4, the application rates of slow-release chelating agents in assisting phytoremediation of heavy metal and radionuclide-contaminated soils ranged from 3 to 13 mmol/kg, which is close to the above application rate range of chelating agents alone. Moreover, when comparing the effectiveness of chelating agents and slow-release chelating agents in enhancing phytoextraction of soil contaminants, the amount of core material in the slow-release chelating agent should be equal to that of chelating agent alone. At present, research on the use of slow-release chelating agents in assisting phytoremediation of U-contaminated soils is still at juvenile stage. The evaluation of their effectiveness in enhancing phytoextraction of soil heavy metals and radionuclides reported in scientific literature are exclusively obtained from laboratory pot experiments with no field applications (Table 4). Hence, it is recommended that pilot-scale tests should be conducted to investigate the effect of slow-release chelating agents on enhancing phytoextraction of soil U under field conditions. This effort can provide a better evaluation of slow-release performance. In

addition, bench-scale accelerated aging tests (e.g., wet–dry cycling and freeze–thaw cycling) can be carried out to investigate the effect of slow-release chelating agents on controlling the release speed of core material under simulated natural aging conditions, which can make a contribution to elucidation of long-term slow-release mechanisms.

4.3. Evaluation of Slow-Release Performance

The slow-release performance of slow-release chelating agents can be indicated by the increasing rate of core material (i.e., chelating agent) content in the soil with time after application. The common methods for evaluating slow-release performance of slow-release materials include hydrostatic dissolution, soil incubation, and soil column leaching [162–166]. In the hydrostatic dissolution method, slow-release material is added to deionized water. The mixture is then maintained at a constant temperature. The amount of core material released into the water is measured regularly to calculate its release speed with time. The method is simple, fast, and well reproducible. However, it ignores the effect of soil environmental conditions on the slow-release performance. The soil incubation method simulates the release of core material in the soil and is closer to natural environmental conditions compared to the hydrostatic dissolution test. In the soil column leaching method, slow-release material is subjected to leaching test, and the concentration of core material in the leachate collected with time is measured. Among the above methods for evaluating slow-release performance, hydrostatic dissolution method has been widely used (e.g., [141,167–169]) due to its simple operating procedure, controllable environmental conditions, and convenient comparison between different slow-release materials. Considering the complexity of actual contaminated site environment, it is necessary to evaluate the slow-release performance of slow-release chelating agents in soils. Li, et al., first synthesized a slow-release chelating agent with EDTA as core material and silicate as coating material [105]. They demonstrated that the change of DOC concentration in the leachate could reflect the slow-release performance of coated EDTA. In contrast to the inorganic wall material in Li, et al., Xie, et al., used polyethylene as the wall material of microcapsule, and the concentration of EDTA in aqueous solution released from the microcapsule was determined by an ion chromatography (IC) [143]. Likewise, Shibata, et al., prepared polymer-coated EDTA and also characterized the slow-release property based on the variation of EDTA concentration in distilled water using a high-performance liquid chromatography (HPLC) [142]. The EDTA concentration in the solution can be determined by HPLC after pretreatment with an extractant containing Cu or Fe ions [170], and Shibata, et al., used a $CuSO_4$ solution as extractant. However, due to the lack of details about the operating procedures of the above methods, it is difficult to compare the slow-release results obtained from different studies. The longest release period of slow-release chelating agent recorded in literature is a polymer-coated EDTA synthesized by Shibata, et al., who reported that 75% of the EDTA in the slow-release chelating agent was released into the water after 210 d [142]. The pot experiment showed that among the five slow-release chelating agents with different wall materials, the EDTA coated with 40% talc, 45% polyethylene, 10% ethylene-octene-1 copolymer, and 5% polyoxoethylene monomethyl ether (0.72 mmol/g) were the most effective in reducing Pb and EDTA concentrations in the soil solution and increasing Pb enrichment in Sorghum bicolor *L.* seedlings with a release period of 80 d [142].

5. Conclusions and Future Perspectives

With the rapid development of nuclear energy and on-going U mining, cleaning up of U-contaminated soils has become a major global challenge. At present, no U hyperaccumulator plant species have been found, whereas plants for soil remediation have low extraction efficiency and long-term remediation period for U-contaminated soils. Both APCAs (e.g., EDTA and EDDS) and LMWOAs (e.g., CA and OA) can enhance phytoextraction of soil U by increasing bioavailable U in the soil via forming soluble chelate compounds. CA, which can be generated by organisms and is biodegradable, has a pronounced effect on mobilizing U in soils, and therefore assists phytoextraction of U from the soil. It should be noted that

after chelating agents are applied to the soil, the mobility and bioavailability of soil U can increase rapidly in a short period, resulting in the problem that the increasing rate of soil bioavailable U does not match the plant uptake rate of and tolerance to U. The majority of mobilized U may not be absorbed by roots and remained in the soil, posing a non-negligible contamination risk to groundwater through leaching processes. Slow-release chelating agents synthesized by slow-release technology can control the release speed of chelating agent so that the mobilization rate of soil U matches plant uptake rate of U.

Currently, research on synthesis and application of slow-release chelating agents in assisting phytoremediation of U-contaminated soils is still in its infancy. The slow-release CA synthesized by Wang, et al., is the only slow-release chelating agent which was used to enhance phytoextraction of soil U [141]. Meanwhile most of the existing slow-release chelating agents designed for phytoremediation of heavy metal-contaminated soils contain non-biodegradable EDTA as core material, which would cause secondary pollution. In addition, the effective performance of the aforementioned slow-release CA was noted in laboratory pot experiments, whereas its effectiveness in assisting phytoremediation of U-contaminated soils in the field remains unclear. Future work can mainly include: (1) selecting and developing environmentally friendly wall/carrier materials and cost-effective preparation methods. Natural materials (e.g., starch, cellulose, and chitosan) and simple preparation methods with low cost (e.g., interfacial polymerization and spray drying) should be screened for synthesis of slow-release chelating agents prior to large-scale application. (2) Some existing wall/carrier materials (e.g., chitosan and its derivatives) of slow-release chelating agents have high adsorption capacity for heavy metals and radionuclides. They can have a passivation effect on these contaminants in soils. Therefore, it is necessary to eliminate the above negative impact of wall/carrier materials of slow-release chelating agents on mobilizing soil U. This can be achieved by optimizing the ratio of wall/carrier material to core material and modifying the surface porous structure of slow-release chelating agent. (3) Existing slow-release chelating agents have been mainly added to soils in the form of solutions. Synthesis of hydrogel-based slow-release chelating agent can be a promising research direction because hydrogel as a carrier for chelating agents has the potential to combine controlled release of chelating agent into the soil with soil and water conservation, thus improving the soil environment. (4) The present systematic review showed that CA is the most effective in enhancing phytoextraction of U from contaminated soils compared to other commonly used chelating agents. However, CA is easily decomposed in soils. Thus, it is encouraged to evaluate the long-term effect of CA on mobilization of soil U as well as explore methods for maintaining the effectiveness of CA during natural aging processes. (5) Soil microorganisms play an important role in the formation of soil fertility, transformation of contaminants as well as nutrient elements [171]. Therefore, it is also necessary to investigate the effect of microbial metabolic activities in rhizosphere soils on the slow-release performance of slow-release chelating agents and explore the transport mechanisms of complex compound formed by chelating agents with U ions at the plant-soil interface. This endeavor can contribute to the combination of chelating agent- and microorganisms-assisted phytoremediation of U-contaminated soils. (6) The studies of Saleh, et al.,e. demonstrated that aquatic plants including *Eichhornia crassipes*, *Ludwigia stolonifera*, and *Myriophyllum spicatum* have an enormous accumulation capacity of heavy metals (e.g., Cd, Cr, Cu, and Pb) and radionuclides (e.g., cesium (Cs) and cobalt (Co)) [172–177]. Therefore, apart from the above on-going research relevant to slow-release chelating agents, the effect of chelating agents on enhancing the accumulation of U and other radionuclides in these aquatic plants in contaminated paddy soils and riparian zones can be investigated.

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