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砷镉复合污染稻田修复研究进展

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摘　要　我国农田土壤重金属污染严重，其中稻田土壤砷镉（As/Cd）复合污染形势严峻。相较于其他粮食作物，水稻具有更强的积累 As/Cd 能力，稻米中 As/Cd 通过食物链进入人体会对人类健康带来危害。因此，修复 As/Cd 复合污染稻田土壤，降低稻米 As/Cd 含量，对保障我国粮食安全生产意义重大。对农田 As/Cd 复合污染现状及危害进行综述，讨论 As/Cd 有效性影响因素及水稻中 As/Cd 吸收转运机制，详细探讨稻田中 As/Cd 污染的修复方法，最后针对目前修复技术提出存在的不足和展望，以期为 As/Cd 复合污染稻田土壤修复提供一定的指导。

关键词　稻田土壤；砷镉复合污染；水稻；As/Cd 有效性；土壤修复

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Research Progress on Remediation of Cadmium and Arsenic Compound Contaminated Paddy Soil

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Abstract　In China, heavy metal pollution in agricultural soils is very serious, especially arsenic and cadmium (As/Cd) co-contamination in paddy soil. Compared with other food crops, rice has a stronger ability to accumulate As/Cd, and As/Cd in rice enters human body through the food chain can causes harm to human health. Therefore, it is of great significance to repair As/Cd compound contaminated paddy soil and reduce rice As/Cd content in rice to ensure the safety of food production. In this paper, the current situation and hazard of As/Cd co-contamination pollution in farmland were summarized, the influencing factors of As/Cd effectiveness and the absorption and transport mechanism of As/Cd in rice were discussed, and then the remediation methods of As/Cd pollution in paddy fields were discussed in detail. Finally, the shortcomings and prospects of the current restoration technology were proposed. This paper aim to provide some guidance for the remediation of As/Cd compound contaminated paddy soil.

Keywords　paddy soil; cadmium and arsenic co-contamination; rice; As/Cd effectiveness; remediation of soil

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因工业采矿、冶炼和农业农药、化肥的使用等人为活动导致大量的重金属进入土壤环境，农业土壤中砷(As)和镉(Cd)的污染已成为一个持久的环境问题[1-2]。As 和 Cd 不是植物必需元素，被国际癌症研究机构定为Ⅰ类致癌物质[3]。As/Cd 可以通过食物链在人体中积累，并对人类健康构成巨大威胁，因此被认为是土壤优先污染物[4]。水稻，是全球一半以上人口的主要粮食作物[5]，可以有效地从土壤中吸收 As 和 Cd，并在根、茎、叶及籽粒进行累积[3,6]。因此，为了改善农业生态和人类健康，减少 As/Cd 从土壤中向水稻籽粒中的累积非常必要[7-8]。

水稻对 As/Cd 的吸收与其在土壤中的迁移和转化有关，受土壤 *Eh*、pH 值、有机质等因素的影响，改变土壤这些性质已成为修复 As/Cd 污染土壤的重要措施。在稻田中，淹水和排水交替可以极大地改变土壤 *Eh* 和 pH 值等土壤性质，从而影响 As/Cd 的迁移性[9]。调节土壤 pH 值和 *Eh* 具有成本低、效果好的特点，是控制水稻土中 As/Cd 有效性的常用方法。然而，As 和 Cd 的生物有效性因其独特的化学行为，往往会朝向相反的方向[9-10]。在淹水环境中，通过静电排斥、与氢氧化物竞争性吸附、吸附的铁氧化物溶解等过程使 As(Ⅴ)被还原为 As(Ⅲ)，As 的有效性和毒性提高[11-12]；相反，由于低 *Eh* 和高 pH 值，氢氧化物和 S^{2-} 的沉淀作用会降低 Cd 的生物有效性[13]。因此，同时降低水稻对土壤中 As 和 Cd 的吸收是一个艰巨的挑战。本文综述了我国农田土壤 As/Cd 复合污染现状及其危害、土壤中砷镉生物有效性及影响因素、水稻砷镉吸收转运机制和稻田砷镉污染修复方法，并对已有修复方法的研究不足进行了总结，以期为我国砷镉污染稻田的修复研究提供一定的理论依据和技术支持。

1 农田土壤砷镉复合污染现状及其危害

1.1 土壤 As、Cd 复合污染来源

土壤 As 的来源可分为自然来源和人为来源，As 的自然来源是指在生物地球化学因素的作用下在自然中释放到土壤所致。据估算，由岩石风化侵蚀作用释放到环境中的 As 通量大约为 4.5×10^4 t/a，且大部分在水力作用下迁移至水体和土壤中[14]。人类活动是稻田土壤 As 污染的重要来源之一。人为来源包含污水灌溉、农药、化肥、畜禽粪便、废弃污泥、采矿、冶金、化石燃料、电子垃圾等[15]。

土壤 Cd 污染一般与其土壤母质、大气沉降、工业活动、农业活动有直接关系。土壤 Cd 的来源也可以分为人为来源和自然来源，自然来源主要是干湿沉降和母岩的风化，后者是 Cd 进入土壤的一个重要途径[16]；土壤 Cd 的人为来源包含燃烧排放、污水污泥、交通运输、采矿活动和化肥的过度施用等[17](图 1)。

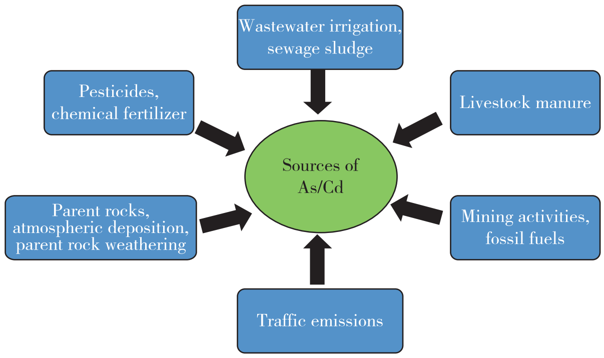

图 1 土壤砷镉的来源

Figure 1 Diagram of arsenic and cadmium sources in soils.

1.2 土壤 As、Cd 复合污染现状

据 2014 年《全国土壤污染状况调查公报》显示，Cd 和 As 的点位超标率分别为 7.0% 和 2.7%，位于第一位和第三位。从整体分布情况看，南方土壤污染程度高于北方；长江三角洲、珠江三角洲、东北老工业基地等部分区域土壤污染问题较为突出。部分地区受 As/Cd 复合污染影响，存在一定程度的健康风险。我国 As/Cd 复合污染土壤呈现出明显的地域分布差异，整体上，As/Cd 复合污染集中在华中华南和西南地区[18]。

1.3 土壤 As、Cd 复合污染的危害

稻米中砷镉污染问题已被广泛关注，As 和 Cd 不仅对水稻植株的生长代谢存在毒性作用，也对人类健康产生潜在危害。CHEN 等[19-20]报道了水稻植株中 Cd 积累的毒性作用，水稻植株中 Cd 积累，显著降低植物生长、抗氧化酶活性、养分利用率和光合作用色素，最终导致产量下降。人体长期接触 Cd 会导致严重的健康问题，如肾脏疾病、肝癌和皮肤癌[21]。As 的累积会导致植物的形态、生理和生化属性的损伤[22]，出现褪绿、枯萎、坏死、抑制生长及光合作用、扰乱代谢途径[23]。人体长期暴露可能导致呼吸系统疾病、免疫学疾病、生殖紊乱、内分泌紊乱、皮肤癌和胃癌等问题[24]。

2 砷镉有效性及影响因素

2.1 土壤 pH 值和 *Eh*

Cd^{2+}、亚砷酸盐(As^{3+})和砷酸盐(As^{5+})的阳离

子是土壤中 Cd 和 As 的主要组分[25-26],容易被水稻植株吸收。当土壤 pH 值较高时,H^+ 值降低,土壤胶体的负电荷增加,提高土壤胶体吸附土壤中带正电荷的 Cd^{2+} 的能力[9],从而降低土壤 Cd 的生物有效性。然而,高 pH 值可以促进土壤溶液中 As^{3+} 和 As^{5+} 的更大解吸,从而提高土壤 As 的生物有效性[27]。当土壤 pH 值较低时,土壤中 Al^{3+} 和 H^+ 浓度较高,会与 Cd^{2+} 竞争土壤表面的吸附点位,同时会降低土壤表面所带负电荷量,增强与 Cd^{2+} 间的静电排斥作用,从而提高其迁移性和溶解性[28];而 pH 值较低时,As 则向结合态的形式转化,迁移性和有效性降低[29]。

土壤氧化还原电位(*Eh*)是反映土壤氧化还原状况的重要指标,能够显著影响 As/Cd 在土壤中的赋存形态和生物有效性[30-31]。在 *Eh* 较低条件下,S 会还原成 S^{2-},易与 Cd^{2+} 形成难溶性的 CdS 沉淀,降低其生物有效性;而在 *Eh* 较低条件下,As(Ⅴ)则会被还原为毒性和有效性更强的 As(Ⅲ),且铁锰氧化物在还原性条件下有再次溶解的可能,会造成原本吸附固定在铁锰氧化物的 As 释放,从而提高其溶解性[32]。在 *Eh* 较高条件下,CdS 沉淀会发生氧化反应,提高土壤 Cd 的迁移性和生物有效性[33],而 As(Ⅲ)则会被氧化为 As(Ⅴ)和铁锰氧化物固定,其有效性降低。

2.2　土壤有机质(SOM)

土壤有机质含量是影响土壤重金属有效性重要的因素之一。有机质含量高的土壤 Cd 的吸附量大,SOM 可以通过络合作用与土壤 Cd 结合,降低有效性[34-35];但也有研究表明,SOM 为土壤溶液提供螯合剂,提高了 Cd 的生物有效性[33]。SOM 通常带负电荷与土壤 ${AsO_4}^{3-}$ 或 ${AsO_3}^{3-}$ 会竞争吸附土壤颗粒表面的吸附点位,降低土壤 As 的有效性和活性;但 SOM 的分解或者水解成为小分子的溶解性有机质(DOM)过程,也可能促进土壤 As 还原,增加其活性和毒性[36]。可见,调控 SOM 会改变土壤 As/Cd 有效性,但尚未形成统一的结论。

2.3　土壤微量元素(Fe、Mn、Si、Ca)

铁(Fe)和锰(Mn)的氧化物及氢氧化物通过对金属强烈的专性吸附作用降低土壤 As/Cd 有效性,土壤 Fe/Mn 形态的变化会导致 As/Cd 的铁锰氧化物结合态的变化[37];在根际环境中,Fe^{3+} 氧化物还原产生的 Fe^{2+} 可以被氧化成水稻根表面的 Fe 膜,限制了 As/Cd 进入根系[18];此外,水稻 Cd 和 Fe 在植株中具有相同的吸收途径,水稻根际 Fe 可能通过竞争 Cd 转运通道来降低 Cd 的吸收[38]。水稻植株中 As 和 Si 存在竞争转运通道,提高土壤有效 Si,可减少籽粒 As 积累[39];此外,土壤中的 Ca 离子可与 As 形成不溶性沉淀,降低 As 的生物有效性[40]。

2.4　土壤微生物

土壤微生物活动影响 As/Cd 的形态和生化反应,从而导致其有效性的改变[41],微生物可通过影响土壤氧化还原反应进而影响 As/Cd 的有效性;水稻根际微生物可分泌有机物和胞外酶,这些物质改变了 As/Cd 在根际的分布和迁移,从而影响了 As 和 Cd 在水稻中的吸收和积累;土壤微生物还可以通过影响水稻根表 Fe 膜的形成来影响其对 As/Cd 吸收和转运[42-43](图 2)。

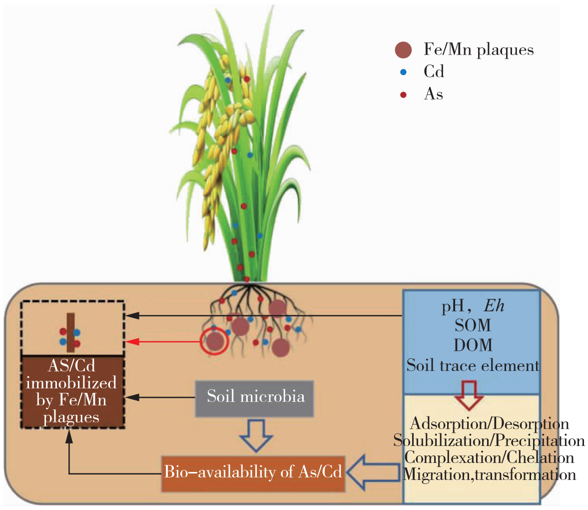

图 2　土壤砷镉有效性影响因素

Figure 2　Factors affecting the availability of arsenic and cadmium in soil.

3　水稻砷镉吸收转运机制

3.1　水稻 As 吸收转运机制

在淹水缺氧环境下,水稻本身会表现出较高的吸收和转运 As(Ⅲ)的能力,此外,水稻植株具有较高的降低根中 As(Ⅴ)的能力,并将 As(Ⅲ)有效上传到木质部组织中[44]。而在水稻细胞中对 As 的吸收主要取决于 As 种类,如砷酸盐[As(Ⅴ)],使用磷酸盐转运体(如 Pht1 转运体),因为 P 与 As(Ⅴ)化学性质相似[45]。亚砷酸盐[As(Ⅲ)]使用硅(Si)转运体,如水通道蛋白 NIP2;1 及 Lsi1 和 Lsi2 转运体,因为 As(Ⅲ)和 Si 相似[46]。As 的减少主要发生在根细胞中,然后再运输到木质部和植物的其他部分[47]。有研究表明无论水稻品种如何,植株 As 在根系中的积累分别是籽粒和地上部的 75 倍和 28 倍[48],根表面

形成的铁锰氧化膜会吸附 As，并限制其进一步向植物组织转运[49]。另有研究表明，水稻韧皮部细胞中发现了一种液泡转运体(OsABCC1)，促进 As 在液泡中积累，减少 As 在籽粒中的转运[50]。

3.2　水稻 Cd 吸收转运机制

土壤中的 Cd 被水稻根细胞吸收后，转移到地上茎叶部，最终在籽粒中积累[46]。木质部在 Cd 从根到茎叶的运输中起着主要作用，韧皮部是将 Cd 从茎运输到籽粒的主要途径[51]。有报道称，OsIRT1 和 OsIRT2 转运体在水稻根系吸收 Cd 的过程中发挥了重要作用[51]，此外，水稻植株中 Cd 的累积主要是由 NRAMP5 转运体促进的[52]，类似于铁转运体的 OsNRAMP1 在植物质膜中也作为 Cd 内流转运体[53]。Cd 从水稻根到茎的运输主要由 OsHMA2 转运体转运[54]，茎叶中的 Cd 再由 OsLCT1 转运体运输到籽粒中[55](图 3)。

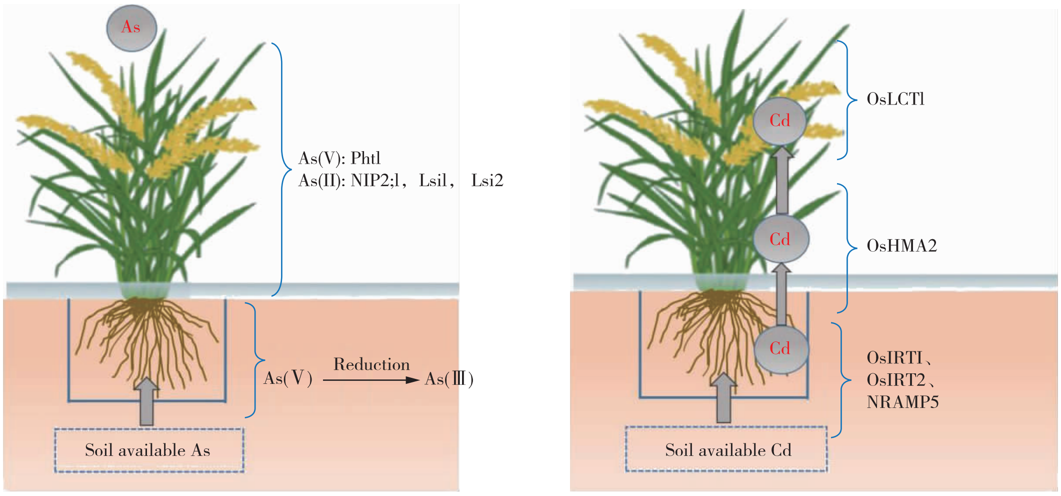

图 3　水稻砷镉吸收转运机制

Figure 3　Mechanisms of arsenic and cadmium uptake and translocation in paddy soil-rice systems.

4　稻田砷镉污染修复方法

As/Cd 复合污染稻田土壤修复是全球性的难题，其修复策略探讨是全球研究的热点之一[56-57]。稻田污染土壤修复与其他类型的污染土壤修复在修复目标、修复对象和修复需求方面的巨大差异性，往往要兼顾生产与修复，因此可供选择的稻田土壤修复技术有限。现阶段主要修复策略从减小土壤 As 和 Cd 的总量、降低土壤 As 和 Cd 的有效性和抑制水稻对 As 和 Cd 的吸收和转运这三个方面展开。具体使用的修复方法包括物理方法、化学方法、生物方法、农艺措施和基因工程等(图 4)。

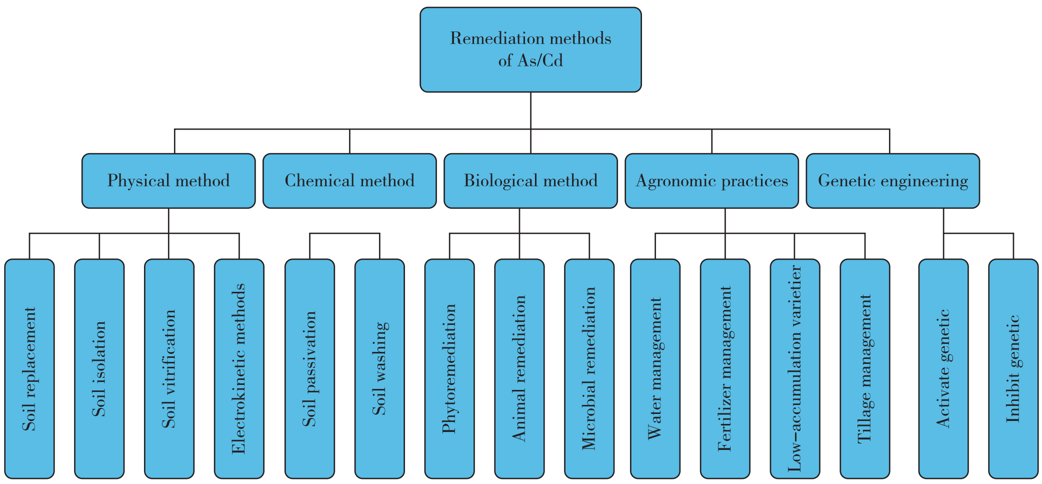

图 4　土壤砷镉修复技术

Figure 4　The remediation methods of asenic and cadmium contaminated soil.

4.1　物理方法

物理修复方法主要包括换土法、隔离法、玻璃化法和电动力学方法[58-59]。换土法是用非污染土壤替代或部分替代污染土壤,该方法能有效减少重金属对环境的影响,但是工程量大、费用高,同时也会对环境产生一定风险[58]。土壤隔离法是指将金属污染土壤与未污染土壤分离,通过将金属元素限制在指定区域内来阻止其向场外移动[60-61]。该方法可以暂时隔离污染区域,以在现场评估和修复期间防止重金属的迁移[62]。玻璃化法是指通过污染场所的高温作用,土壤系统内金属元素的运动减少,导致玻璃体物质的形成[57]。该方法能耗大、破坏土壤有机质,改变土壤原生态结构且对易挥发元素产生的蒸气不易回收,造成二次污染。电动力学修复技术是指向土壤施加直流电场,在电解、电迁移、扩散、电渗透、电泳等作用的共同作用下,使土壤溶液中的离子向电极附近富集从而被去除的技术[63]。该方法具有治理效果较好、时间短等特点[64],但是工程量相对较大,消耗人力物力,费用昂贵,还会引起土壤结构混乱,肥力降低。

4.2　化学方法

化学修复方法主要包括化学钝化和化学淋洗[58]。化学钝化法是指通过在受污染的土壤场地中添加化学试剂来降低土壤中重金属的迁移性和生物有效性,通过吸附、络合和沉淀作用固定化土壤金属元素[17]。土壤金属钝化作用通常是通过使用无机和有机改良剂来实现的[65],常见无机改良剂包括磷酸盐、沸石、水泥、黏土矿物,常见的有机改良剂包括秸秆、有机堆肥、天然腐殖质、泥炭、生物炭等[62]。该方法优点是快速、高效、便捷,可实现“边生产,边修复”,同时存在成本高、修复时间长、总量降低效果不明显,存在二次污染的风险等不足[17,58]。化学淋洗技术通常指的是利用不同的淋洗剂和萃取液去除土壤中的重金属[66]。将土壤从污染场地挖出,加入合适的淋洗剂与土壤中的金属离子发生沉淀、螯合或离子交换等将其转移至渗滤液中。该方法主要去除土壤表层的有毒物质,去除效率较高。但因为淋洗剂的加入,很有可能会对土壤产生二次污染,同时也有土壤营养元素流失、肥力降低等风险[67]。

4.3　生物方法

生物修复重金属污染土壤的治理法是指利用植物、动物以及微生物的生命代谢活动,降低土壤环境中重金属浓度或通过生物作用改变其在土壤中的化学形态而降低重金属的迁移性或毒性的技术[68]。生物修复技术与传统的处理技术相比具有成本低、易于操作、修复效果好,对轻微至中度污染土壤来说,是一种环境友好型和资源节约型的修复方法[69-70],因而越来越受到人们的关注,成为了土壤污染修复方面研究的热点。主要包括植物修复、动物修复和微生物修复[71-72]。

植物修复:通常指的是利用自然界中的一些植物通过吸收、提取、转化和固定等方式来对土壤进行修复,这些植物通常对土壤中的一种或几种重金属离子具有富集效应[73-75]。植物修复是一种很有前途的技术,与传统的物理化学修复方法相比,该方法高效、简单、无侵入性、成本低、生态友好,已逐渐被采用[76-77]。然而因其存在生物量低、生长缓慢、植物吸收金属的能力有限,其大规模应用受到限制[77],因此,需要采取提高其修复效率的补救措施。

动物修复:旨在通过土壤中的蚯蚓、潮虫、黑水虻等动物对污染物进行降解[78]。动物在土壤中的新陈代谢、生长发育、繁衍等活动会直接或间接的影响土壤的物质组成和分布;同时,动物的消化道微生物也会对污染物进行降解或转化,进而减少污染物对土壤的危害性[67]。蚯蚓作为生态系统工程师,在改善土壤质量,提高土壤肥力方面起着至关重要的作用[79-80];蚯蚓可以通过改变土壤理化性质及微生物群落结构影响金属有效性[81],还可以通过摄食和皮肤吸收改变土壤中金属的含量和迁移性[82-83]。近年来,蚯蚓作为修复生物对污染土壤进行生物修复及其扩展技术引起了广泛关注,大量研究表明蚯蚓对土壤重金属污染具有良好的修复效果[84-86]。

微生物修复:主要是指利用微生物促进土壤中重金属元素的沉淀、吸附和氧化还原[46],微生物修复法成本低、效率高,可以在耕作时间同时进行,可在原位进行修复,对土壤环境的影响较小,甚至可以起到改善作用,是一种安全的修复方法。但微生物修复技术也存在对重金属固定能力有限、代谢水平不佳、微生物流逝或吞噬现象等问题[87]。所以,微生物在强化修复以及联合修复方法中更具备应用优势。LIU 等[88]研究表明,丛枝菌根真菌提高了植物生物量和 Cd 的浓度,增加了植物对 Cd 的富集。

4.4　农艺措施

常用的几种农艺措施可通过改善土壤理化性质对 As/Cd 起到固定化作用,主要包括水分管理、施肥管理、筛选低累积水稻品种及耕作管理等。水分管理主要是通过调节土壤 Eh 和 pH 值来影响 As 和 Cd 的有效性,HONMA 等[89]在水稻抽穗期前后

三周以淹水为对照进行不同水分管理调控，发现水稻土中溶解态 As 和 Cd 的含量与土壤中 *Eh*、pH 值及溶解 Fe(Ⅱ)浓度的变化相关，且控制 As/Cd 浓度的最佳土壤 *Eh* 为－73 mV，pH 值为 6.2。吴佳等[90]采用盆栽实验研究了不同水分管理方式对水稻 As 和 Cd 吸收转运的影响，结果发现湿润灌溉和间歇灌溉创造的氧化与还原条件的交替是修复 As/Cd 复合污染土壤的有效措施。

施用含磷(P)、铁(Fe)和硅(Si)的肥料可以改变土壤性质，从而影响 As/Cd 的有效性。施用 P 肥可提高了土壤 pH 值，增强土壤中 Cd 的吸附，从而降低土壤中 Cd 的生物有效性[91]；磷酸盐肥料施入土壤后可通过络合反应等实现对 As 的钝化[92]。含 Fe 类肥料主要通过专性吸附和非专性吸附作用降低土壤中 As 的生物有效性[93]；Fe 和 Cd 属于同族重金属元素，同时存在土壤中时，由于对吸附点位的竞争会导致出现拮抗竞争作用，因此在植物生长过程中 Fe^{2+} 会与 Cd^{2+} 竞争吸附位点，从而抑制植物对 Cd 的吸附，降低 Cd 的生物有效性[94]。含 Si 类肥料也被广泛应用于 As/Cd 复合污染稻田土壤修复，Si 肥可以提高土壤 pH 值，增强土壤对 Cd 的吸附，从而降低 Cd 的迁移性，还可以增加土壤中有效硅(SiO_2)含量，进而与土壤中不稳定态 Cd 络合形成牢固的 Si-Cd 络合物[93]；As(Ⅲ)和硅酸是大小相似的四面体，且水稻根系细胞吸收 As(Ⅲ)和 Si 的转运体系统是相同的[95]，因此水稻在吸收和转运 Si 和 As(Ⅲ)之间会产生竞争。此外，Si 肥的加入还可以促进水稻根表铁膜的形成，增加对土壤 As/Cd 的吸附作用，抑制水稻根系对 As/Cd 的吸收与积累[96]。

水稻 As/Cd 低累积品种的选择和育种是降低稻米中 As/Cd 积累的有效方法之一。研究表明在 Cd 污染土壤中种植低 Cd 水稻品种能使稻米 Cd 含量低于国家限量标准(0.20 mg/kg)[97]，NORTON 等[98]在孟加拉等地的 300 多个水稻品种大田实验中，筛选出了 76 个 As 低累积品种。在低、中等程度 Cd 污染土壤中，粳稻品种比籼稻[99]更不易累积 Cd，旱季品种比雨季品种[100]更耐亚砷酸盐或砷酸盐污染的土壤。此外，耕作方式也会影响稻米 Cd 的累积，油菜等高积累性作物的间作可以降低水稻的 Cd 含量[101]；减少耕作方式确保了土壤中较高的有机质含量，从而增强了 Cd 等金属的吸附和络合作用[102]。

4.5 基因工程

已有研究表明，通过基因工程激活或抑制某些基因的表达，可降低了 As/Cd 对水稻的毒性[103-105]。例如，OsHMA3 过表达可抑制 Cd 向水稻茎叶的运输及籽粒中的积累[106]；抑制 OsNRAMP5 的表达可抑制 Cd 在籽粒中累积[107]。过表达 OsNIP1；1 和 OsNIP3；3 可以减小籽粒中 As 的累积[108]；过表达 OsABCC1 和启动 PvACR3；1 表达可增加 As 在液泡中的滞留，减小在籽粒中的累积[109]。因此，基因工程可以成为降低水稻 As/Cd 水平的潜在技术。

5 总结与展望

As/Cd 复合污染土壤中元素间相互作用复杂，在不同氧化还原电位和 pH 值的土壤环境中，容易出现 As 和 Cd 活性此消彼长的现象，因此单独类型的钝化技术很难解决 As/Cd 复合污染问题，耦合技术将成为 As/Cd 复合污染水稻土钝化修复未来研究的重点。目前 As/Cd 复合污染修复技术仍存在许多不足之处，有待进一步研究。

1)目前，砷镉复合污染稻田的修复主要关注的是对土壤 As/Cd 进行钝化，降低其活性，减少其被水稻吸收量，对土壤 As/Cd 总量去除效果并不明显。

2)大部分稻田 As/Cd 污染的处理技术还处于实验室或田间实验和示范阶段，真正的工程应用较少。

3)对于修复材料及修复生物的回收和处理有待强化，许多修复材料可能会出现二次污染等现象。

4)由于土壤性质的差异，能够大面积推广的通用技术很少。

针对上述存在问题提出以下几点建议：

1)将不同的修复方法联合使用，探索有效去除 As/Cd 总量的修复方法和修复技术。

2)因地制宜，不同土壤探究不同修复方法，多支持鼓励开展大田实验探究，将修复技术切实应用到农业生产中。

3)应用稳定的同位素作为示踪剂，更好地探究 As/Cd 转运的具体机制。

4)农业、环境和医学多学科研究人员之间需要密切联系，以真实有效评估 As/Cd 在土壤-植物-动物-人类系统中的健康影响。

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