

doi: 10.12029/gc20220702001

赵鑫娜, 杨忠芳, 余涛. 2023. 矿区土壤重金属污染及修复技术研究进展[J]. 中国地质, 50(1): 84–101.

Zhao Xinna, Yang Zhongfang, Yu Tao. 2023. Review on heavy metal pollution and remediation technology in the soil of mining areas[J]. Geology in China, 50(1): 84–101(in Chinese with English abstract).

矿区土壤重金属污染及修复技术研究进展

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摘要:【研究目的】土壤重金属污染是造成土壤生态环境质量下降、农作物重金属超标,甚至危及人体健康的主要因素。采矿及冶炼活动是土壤重金属污染的主要来源,中国矿山贫矿多、难选矿多、共生矿多,矿山开采过程中产生了大量的有毒有害重金属进入土壤,由于采矿活动造成土壤重金属污染严重。目前,寻找合适的方法修复矿区重金属污染土壤是国内外环境科学领域研究的热点和难点。【研究方法】本文通过查阅大量矿区重金属污染土壤修复技术方向的文献,综述了国内外关于矿区土壤中重金属污染的特点、危害及修复技术最新研究进展。【研究结果】目前矿区土壤重金属污染的修复方法包括:(1)工程修复技术:客土法;(2)物理修复技术:电动修复法、玻璃化法、热处理法等;(3)化学修复技术:土壤淋洗法、固定/稳定化法等;(4)生物修复技术:植物修复法、微生物修复法等。【结论】目前客土法是使用较广、最有效的处理技术,植物修复技术是应用前景最好的技术。在实际矿山重金属污染土壤修复中,单一的修复技术往往存在一定的局限性而不能满足修复需求,因此可采用多种修复技术相结合的联合修复方法相互补充,以达到最佳修复效果。

关键词: 矿区; 土壤; 重金属污染; 修复技术; 环境地质调查工程

创新点: (1) 本文综述了矿区土壤重金属污染特点,对今后有关矿区土壤重金属污染研究有一定的帮助;(2) 本文较为全面地整理了矿区重金属污染修复技术,提出关于矿区土壤重金属污染修复技术选择建议,希望能有效提高未来矿区土壤治理水平。

中图分类号: X53 文献标志码: A 文章编号: 1000-3657(2023)01-0084-18

Review on heavy metal pollution and remediation technology in the soil of mining areas

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收稿日期: 2022-07-02; 改回日期: 2022-10-26

基金项目: 中国地质大学(北京)地质调查成果转化基金(202001, 2020002)、中国地质调查局项目(DD20211574, DD20211414)、秦巴山区健康地质调查与评价(No.202201)联合资助。

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Abstract: This paper is the result of environmental geological survey engineering.

[Objective] Soil heavy metal pollution is the main factor that causes the decline of soil ecological environment quality, crop heavy metal exceeds the national standard and even endangers human health. Mining and smelting activities are the primary source of soil heavy metal pollution. China's domestic metal mines have relatively low-grade and are expensive to process. The mining process produces a large amount of toxic and harmful heavy metals into the soil, which causes severe soil heavy metal pollution due to mining activities. At present, finding suitable methods to remediate heavy metal contaminated soils in mining areas is a hot issue and complex research area in environmental science. **[Methods]** This paper reviews the characteristics, hazards and remediation technology of heavy metal pollution in mining soils by many relevant literatures. **[Results]** The current remediation methods for soil heavy metal pollution in mining areas include: (1) engineering remediation technology: guest soil method; (2) physical remediation technology: electro kinetic remediation, vitrification method and heat treatment method; (3) chemical remediation techniques: soil leaching and fixation; (4) bioremediation technology: phytoremediation and microbial remediation method. **[Conclusions]** The guest soil method is the most widely used and effective treatment technique, and phytoremediation technology is the best application prospect. A single remediation technique often has certain limitations in the actual remediation of heavy metal contaminated soil in mines. It cannot meet the remediation needs, so a combined remediation method combining multiple remediation techniques can complement each other to achieve the best remediation effect.

Key words: mining; soil; heavy metal pollution; remediation technology; environmental geological survey engineering

Highlights: (1) This paper summarizes the characteristics of soil heavy metal pollution in mining areas, providing a reference for future research on such areas. (2) This paper comprehensively sorted out the remediation technology of soil heavy metal pollution in mining areas and provided some suggestions on the selection of remediation technology which would effectively improve the level of soil treatment in mining areas in the future.

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Fund support: Supported by China University of Geosciences (Beijing) Geological Survey Achievement Transformation Fund (No.202001, No.2020002), the project of China Geological Survey (No.DD20211574, No.DD20211414) and Health Geological Survey and Evaluation in Qinling-Bashan Mountain region (No.202201).

1 引言

土壤作为人类赖以生存的资源,在人类的生产生活中占据至关重要的位置。然而许多国家,尤其是发展中国家在大力推进工业化的同时,环境正遭受着一定的污染破坏,其中土壤的重金属污染问题较为严重。据估计,中国耕地约1/6左右受到了土壤重金属污染,经济损失严重(宋伟等,2013;林苾等,2021)。土壤的重金属污染,不仅降低了土壤的质量,同时也会对人的身体健康造成很大的危害,土壤重金属污染治理已经成为当今社会环境保护不可避免的内容(余春浩,2019;刘利等,2020;綦崢等,2020)。

土壤中重金属元素主要有自然来源和人为输入两种途径。土壤自然来源的重金属元素主要指

继承于成土母质或在成土过程中次生富集形成的重金属元素。通常情况下,源于自然源的土壤重金属,在含量特征与空间分布上对应于一定的地质体,如土壤中Cd、As、Hg、Cu、Zn等重金属异常区,即地质高背景区,与黑色岩系、煤系地层、金属矿床、磷矿等分布相一致(刘晓媛等,2019;郭超等,2019;何振嘉,2020;杨琼等,2021;崔潇等,2021)。中国相当一部分受重金属污染的土壤分布在地质高背景区,特别是在金属硫矿床、磷矿等富含重金属元素的矿山分布的地质高背景区(Cheng et al., 2014; Yang et al., 2014; 骆永明和滕应,2018),在金属矿山开采和资源利用过程中,所造成的土壤重金属污染元素种类多、异常范围广、污染程度高,且易在植物体内积累,并通过食物链富集到动物和人体中,诱发癌变或其他疾病,生态危害大(鲍丽然等,

2020; Liu et al., 2021), 如铅中毒会影响人的神经系统、造血系统和消化系统等, 镉中毒则会引起骨痛病等(徐友宁等, 2013; Wen et al., 2020; 朱丹尼等, 2021)。

目前由于采矿活动世界许多国家都面临着不同程度的矿区土壤重金属污染问题。土壤修复技术的研制成为国际上的研究热点问题, 在矿山污染土壤修复技术研制与管理方面, 国外起步较早, 早在20世纪初期, 欧美等国家就开始了矿区的土地复垦和生态恢复工作(张永双等, 2017)。和欧美发达国家相比, 虽然中国矿山修复工作起步较晚, 但随着规章制度的日渐完善及科学技术的不断进步, 矿区土壤重金属污染修复工作也将逐渐规范化、系统化。本文系统地总结了矿区土壤重金属污染特点、现状和修复技术, 并探讨了未来发展趋势, 对于矿区土壤重金属污染防治具有重要的参考价值。

2 矿区土壤重金属污染特点

矿区土壤重金属污染多来源于富含有害重金属的矿山采选活动, 土壤重金属污染具有重金属元素富集程度高、复合污染明显、污染面积大和生态危害显著的特点。

各类矿床中, 重金属元素一般以硫化物矿物形式存在。在这些矿床的开采、冶炼过程中, 会产生大量的含有Pb、Cd、As、Cu、Zn等重金属硫化物矿物的尾矿废石, 在地表环境中极易氧化, 一方面产生酸性废水, 加速了矿物的溶解, 一方面则使矿区中的重金属元素活化, 以离子形态随地表水迁移到矿区周边的农田土壤, 由于长时间的重金属累积效应, 导致金属矿区土壤中重金属含量远远超过其背景值。邬光海等(2020)发现内蒙古赤峰市废弃钨钼矿区周围土壤重金属元素明显高于矿区周边背景值, As、Cd、Pb、Zn、Cu、Pb和Mo高含量区集中分布在尾矿周围, 受矿山采选活动影响较大。

矿区土壤重金属污染常为多种重金属复合污染。由于矿山废石中常含有As、Pb、Zn、Hg、Cd、Cu、Co、Ni等多种化学元素, 对矿区土壤造成污染的重金属元素种类较多, 多是几种、甚至是十几种重金属元素交织的复合污染。Timofeev et al. (2018)对贝加尔湖钨钼矿周围土壤调查发现, 在矿山废弃地周围土壤存在着Cd、Cu、As、Pb、Zn、Mo等

重金属元素不同程度交织的污染状况。表1列出了不同类型金属矿山的矿业活动造成的土壤重金属污染种类, 土壤重金属复合污染是其突出的特点。

矿区土壤重金属污染面积大。在世界范围内, 随着矿山的开采和矿山废弃地的产生, 已经使数以百万计的矿区土壤受到了重金属污染(Asensio et al., 2013; Mahar et al., 2016)。据《2018中国环境统计年鉴》统计, 截至2017年, 中国因矿山开采累计破坏土地面积2607489 hm², 当年废弃地恢复面积仅75778 hm²(周连碧等, 2021)。

矿山土壤重金属污染生态危害显著。目前金属矿山的开采、冶炼及矿山废弃地的产生是造成土壤重金属污染、粮食重金属含量超标和生态风险增加的重要原因(Yang et al., 2019; 程睿, 2020)。当土壤中重金属浓度过高时, 不仅会损害土壤质量、引起大量的农作物减产甚至绝收、导致树木和动物死亡, 甚至会危害矿区附近居民的身体健康(杨威杉等, 2018; Liu et al., 2020)。如广东韶关市上坝村就是由于村民长期食用在大宝山矿山开采时受到镉污染的稻米而成为癌症村(蓝楠, 2014; 陈强等, 2020)。

3 中国矿区土壤重金属污染现状

中国矿产资源丰富, 种类齐全, 分布广泛, 是世界主要矿业大国之一(图1)。总体上看, 中国矿产资源禀赋较好, 以开采铁、铝、铅、锌、铜等金属矿产和石料、磷矿、建筑用砂等非金属矿产为主。据中国地质调查局统计, 全国约有大型矿山4800多个, 生产矿山7.8万个, 矿山总面积约1.04×10⁴ km²(李国政, 2019)。中国目前共有300多个以矿业为主导的矿业城市, 开采规模居世界前列(黄永泉等, 2012; 关铎, 2016; 李小龙和王雪冬, 2018; Wang et al., 2018), 随着中国的矿产资源被大量开采, 产生了大量的矿渣, 给环境带来了相当大的压力, 矿区土壤重金属污染分布广泛, 各省均存在不同程度重金属污染, 空间分布上东部较西部严重, 南部较北部严重, 金属矿山造成的土壤重金属污染在中国西南、华东、华南地区尤为突出(何芳等, 2010)。

据全国矿山地质环境调查数据统计, 截至2010年底, 中国矿山破坏土地面积约386.80×10⁴ hm², 而恢复治理面积只有49.60×10⁴ hm², 恢复治理率仅为

表1 世界上一些受污染矿区土壤中某些重金属元素的浓度

Table 1 Summary of concentrations of some heavy metals in some contaminated mining areas in the world

矿区位置	主要矿产资源	主要金属污染元素	浓度(mg/kg)	文献来源
摩洛哥 Oued Zem 市 Ait Ammar 铁矿区	Fe	Fe	435490.0	Nouri et al., 2014; Nouri and Haddioui, 2016
		Cd	20.399	
		Cr	222.162	
		Pb	9200.00	
		Zn	153.3	
塞浦路斯 Limni 矿区	Cu	Cu	9653	Christou et al., 2017
		Zn	1081	
		Ni	118.16	
		Pb	20.35	
		Cd	2.12	
突尼斯 Jebel Ressay 的 Zn-Pb 矿区	Zn-Pb	Zn	42400.0	Boughattas et al., 2016
		Pb	14500.0	
		Cd	184	
		Mn	413	
陕西潼关金矿区	Au	Pb	1295	Xiao et al., 2017
		Zn	852	
		As	22	
土耳其 Kutahya 地区的 Gumuskoy 银矿区	Ag	As	14662	Yildirim and Sarwar, 2016
		Ag	127	
		Pb	13556	
重庆裕溪锰矿区	Mn	Mn	120566.00	Hao and Jiang, 2015; Wu et al., 2016
		Cd	6.82	
		Cu	138.43	
		Pb	126.55	
		Zn	398.55	

12.80%(何芳等, 2013)。2014 年环境保护部和国土资源部发布的《全国土壤污染状况调查公报》指出在全国范围内调查的 70 个矿区的 1672 个土壤点位中,超标点位占 33.4%,其中,有色金属矿区周边土壤镉、砷、铅等污染较为严重,工矿业废弃地土壤环境问题突出。2016年,中国公布的《中国土壤修复技

术与市场发展研究报告(2016—2020)》指出:目前中国需要环境恢复与治理的矿山中重金属矿山占 30%,土壤重金属污染严重。修复治理矿区污染土壤、恢复并提升矿区土壤生态功能是解决我国土壤环境问题的当务之急(Zhao et al., 2015)。同年,国务院颁布《关于印发土壤污染防治行动计划的通知》,即“土十条”,旨在明确监管重点并加强空间布局管控。

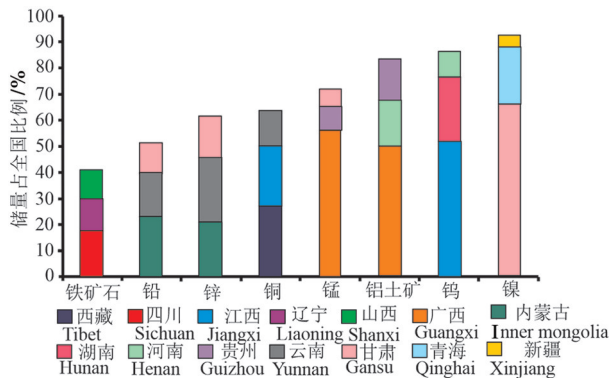


图1 中国主要矿产储量地区分布(据中国矿产资源报告(2021)修改)

Fig.1 Regional distribution of major mineral reserves in China (modified from Report of China Mineral Resources (2021))

4 矿区土壤重金属污染修复技术

在对矿区重金属污染土壤的修复技术进行研究时,须根据其污染特点制定合理且行之有效修复方法。目前矿区土壤重金属污染治理途径归纳起来主要有 3 种:一是改变重金属在土壤中的赋存状态,使其稳定或固定,降低其在环境中的迁移性和生物可利用性;二是利用各种技术从土壤中去掉重金属,达到回收和减少土壤中重金属的双重目的;三是利用水泥、黏土等各种防渗材料,将污染地区与未污染地区隔离,以减少或阻止重金属的迁移和扩散,由此各国相继研究出物理法、化学法及生物法(陈桂荣等, 2010)。在修复技术上,矿区土壤重

金属修复治理一般包括工程修复技术、物理修复技术、化学修复技术、生物修复技术等。

4.1 工程修复技术

土壤的工程修复技术主要包括换土法、客土法与深耕翻土法等。换土法是指将被污染的土壤运走,在原本污染区域填入无污染的土壤;客土法是指在被污染的土壤上覆盖上无污染的土壤;深耕翻土是指翻动土壤上下层,将下层干净无污染土壤与上层被污染土壤调换位置从而改善表层土壤质量的方法(侯李云等,2015;曲磊和石琛,2019)。客土法是一种最为传统的土壤改良技术,它可以在较短的时间内显著改善矿区土壤状况,加快生态修复速度(刘永光等,2012;侯李云等,2015)。一般选择质地较好或肥力较高的壤土、沙壤土等作为客入的无污染土壤,此外客入土层的厚度还要能满足植物根系的健康生长需求,以避免其伸到污染土层。目前许多国家都已经利用客土法对矿区土壤进行了实地修复,并取得了不错的效果,但该方法整体工程量偏大,需耗费大量劳力,一般只适用于土壤污染面积较小的情况(闫德民等,2013)。

20世纪前半叶日本富山县周边的矿业公司向神通川流域的河道中排放了大量含镉废水,造成周边地区土壤、水稻中镉含量普遍超标,严重危害了当地居民的生命健康。据此,富山县政府宣布采用客土法修复污染土壤。1983—2002年,客土法修复受污染稻田,水稻籽粒Cd含量显著下降,如图2中字母B到G所示(Aoshima,2016),未受污染的参考区(A区)水稻中Cd的平均含量稳定在 $0.1 \mu\text{g/g}$ 左右,而Cd污染的神通川流域水稻中Cd平均含量在进行土壤修复后也逐渐稳定在 $0.1 \mu\text{g/g}$ 左右(B~G区),除F区未进行复垦外,其他受污染地区水稻Cd含量均有显著下降。

4.2 物理修复技术

物理修复是指通过各种物理过程把土壤中的污染物分离或去除的技术,目前物理修复的常见方法有电动修复法、玻璃化法、热处理法等。

4.2.1 电动修复法

电动修复技术(electrokinetic remediation, EKR)是指在污染土壤两侧施加直流电压(电动修复装置如图3所示),在电场的驱动下使土壤重金属活化并通过电迁移、电渗流和电泳三种方式定向运动,促

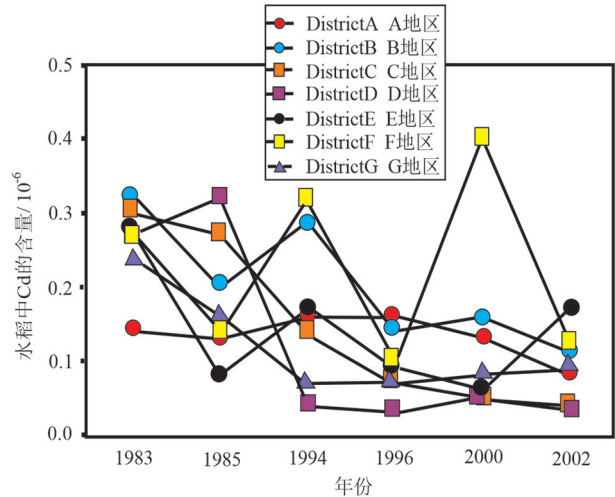


图2 1983—2002年神通川流域受污染水稻(B~G区)和参考区(A区)水稻稻(Cd)平均浓度的变化(据 Aoshima, 2016 修改)

Fig.2 Variation of average cadmium (Cd) concentration (geometric mean) in home-grown rice from the Jinzu River basin (Districts B-G) and the reference area (District A) from 1983 to 2002 (modified from Aoshima, 2016)

使重金属向两电极邻近区域富集,最后通过抽离电解质等方式实现重金属的去除,达到清洁污染土壤的目的(Vocciant e et al., 2017; Wu et al., 2021)。1993年, Acar and Alshawabkeh (1993)率先提出将电动力学技术用于重金属污染土壤的修复,目前,电动修复技术已应用于去除土壤中如铜、铅、锌和镉等多种重金属污染物(Isosaari et al., 2012; Asadollahfardi et al., 2021; 陈敏洁等, 2021)。土壤理化性质、电极材料、土壤pH、电解质、处理时间等均会影响电动修复效率(张小江等,2021)。电极材料是影响电动修复的重要因素,蔡宗平等(2016)采用不同电极材料(石墨、不锈钢、钛板)对尾矿附近的铅污染土壤进行电动修复,发现48小时后,石墨电极去除效果最好,总铅去除效率达到了77%。在进行电动修复之前,还可向土壤中添加络合剂或酸性溶液等辅助试剂调控土壤pH以提高土壤中重金属的去除率,Ryu et al.(2011)在去除金属冶炼厂附近土壤中铜等重金属元素的实验工作发现用酸性溶液预处理土壤后,土壤中Cu和Pb的最大去除效率提高至60.1%和75.7%。

土壤系统的复杂性使该方法具一定的局限性,目前电动修复多停留在实验室模拟或比较小的尺度污染修复上,未大范围地进行应用推广。电动修

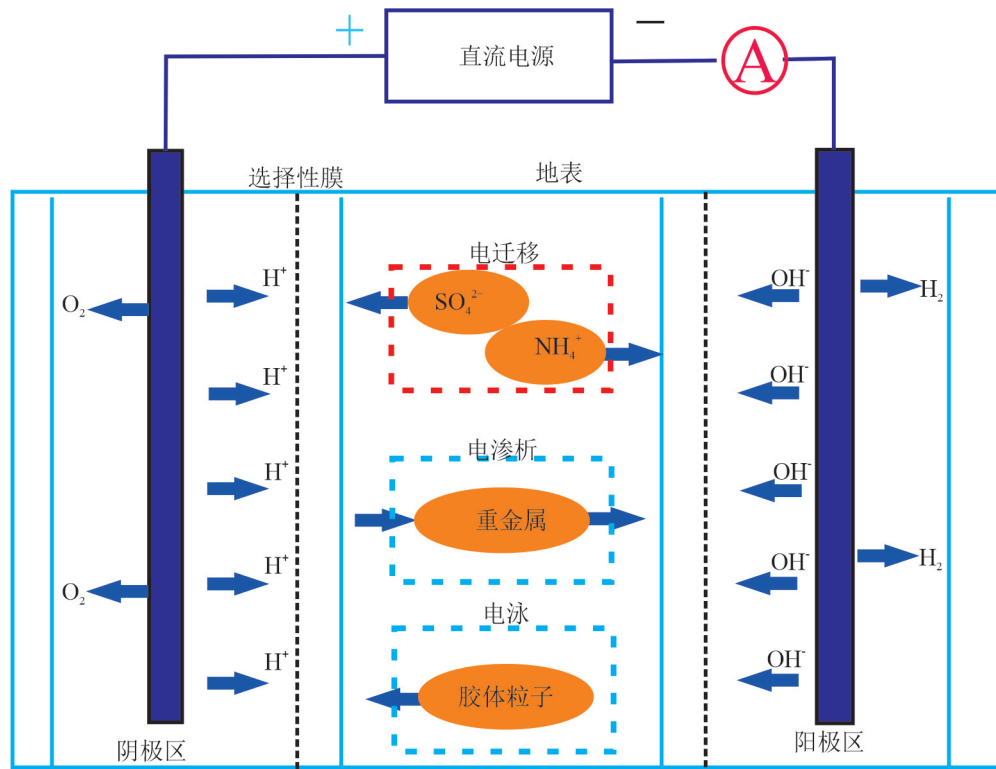


图3 电动修复原理示意图(据张小江等, 2021 修改)

Fig.3 Schematic diagram of electrokinetic remediation (modified from Zhang Xiaojiang et al., 2021)

复在去除土壤重金属同时,还可能影响到土壤中氮、磷、钾等营养物质含量,Zhou et al.(2015)研究发现,电动修复污染土壤后,从阳极到阴极,土壤有效氮和磷含量逐渐减少,土壤有效钾含量逐渐增加。因此,电动修复后的耕地,须采取有效措施,增加土壤肥力、改善土壤结构,才能用于农业耕作。

4.2.2 热处理法

对挥发性的重金属如汞,可使用热处理技术,通过加热的方法将其从土壤中解吸出来,然后再回收利用,目前通常在高温(例如600~800℃)下修复汞污染土壤(Yao et al., 2012)。实施时,需严格设计并操作加热和蒸汽收集系统,以防污染物扩散而产生二次污染。高温热处理技术设备成本高且能耗大,近年有人提出使用太阳能代替传统的不可再生能源,如Navarro et al.(2009, 2014)利用太阳能供热修复汞污染土壤发现,土壤残留总汞和包括汞在内的重金属离子浸出浓度均有显著下降(Wang et al., 2012)。时间和温度是影响热处理效率的重要因素。Sierra et al.(2016)对Almadén矿区汞污染土壤进行热处理修复实验发现随着处理温度的升高,

总汞含量不断下降,750℃时去除效率最高,土壤中残余汞含量降低到0.88 mg/kg。有学者研究贵州省某汞矿附近污染土壤时发现土壤中残留汞含量随着处理温度的升高和处理时间的增加而减少,但单纯增加处理时间并不能一直有效地降低残留汞含量,相同温度下处理60 min和90 min土壤残留汞含量几乎不变(Ma et al., 2014)。

热处理修复技术在采矿活动导致的土壤汞污染治理方面具有突出的应用价值(王泓博等, 2021)。热处理技术处理速率高、修复周期短、普适性强,但它能耗较高,成本高昂,只能用于小范围的土壤重金属污染修复。加入活性炭、纳米材料、生物炭、粉煤灰等可以提高热处理修复的效率,降低热处理温度。Ma et al.(2014)发现在添加FeCl₃后,即使热处理温度低至400℃时土壤中汞含量仍降低到0.8 mg/kg。此外,热处理技术温度过高时,会破坏土壤中的有机质等营养物质,造成土壤理化性质发生变化,限制了该技术的应用(Huang et al., 2011)。

4.2.3 玻璃化法

玻璃化是通过加热将污染的土壤熔化,冷却后

形成比较稳定的玻璃态物质,借助玻璃体的致密结晶结构,使其转化为稳定状态(Mallampati et al., 2015; Liu et al., 2018)。1991年美国爱达荷州工程试验把各种重金属废物(As、Ag、Ba、Cd等)填埋于0.66 m地下后,使用玻璃化法固定了重金属污染物,证明了该技术的可行性(王新和贾永锋, 2007)。玻璃化技术加热过程导致了玻璃相的形成,玻璃相可能吸收了土壤中存在的金属。温度是影响玻璃化技术固定化效果和工艺成本的关键因素(Liu et al., 2018)。Navarro et al.(2013)对西班牙某汞矿的采矿废料在1350℃进行玻璃化处理,发现土壤中Fe、Mn、Ni、Cu和Zn的含量降低且处理后的样品具有简单的矿物学特征。

玻璃化技术对土壤的修复效率较高,修复效果良好。玻璃化技术形成的玻璃类物质结构稳定,一般试剂很难降解,这使得玻璃化技术实现了对土壤重金属的永久固定,处理后对环境危害较小。但是该技术的应用需要相关设备的支持,能耗较大、成本较高,因此玻璃化技术目前仍停留在实验尺度(曲永军, 2021)。

4.3 化学修复技术

化学修复是指通过各种化学过程把土壤中的污染物分离或去除的技术,目前化学修复的常见方法有固化/稳定化技术和化学淋洗技术等。

4.3.1 固定/稳定化法

固定/稳定化技术(S/S)是指向土壤中添加固化/稳定剂将土壤中的有害污染物固定起来,或者将污染物转化成化学性质不活泼的形态,阻止其在环境中迁移、扩散等过程,从而降低污染物质毒害程度的修复技术(郝汉舟等, 2011)。目前常用的S/S固定剂包括:(1)水泥;(2)石灰/粉煤灰;(3)有机堆肥、城市污泥等有机物料;(4)硫酸亚铁、磷酸盐等药剂;(5)海泡石、沸石等矿物材料(赵述华等, 2013; Derakhshan et al., 2018)。水泥是一种无机胶结材料,加水后能发生水化反应生成坚硬的水泥固化体。由于水泥原料来源丰富、处理费用低、效果好、操作简单,所以在固定/稳定化应用中使用最为广泛。在水泥的水化过程中,重金属可以通过吸附、沉降、离子交换、钝化等多种方式与水泥发生反应,最终以氢氧化物或络合物的形式停留在水泥水化形成的水化硅酸盐胶体表面,同时水泥的加入也能提高土壤的pH,抑制重金属的浸出与迁移(Amin et al.,

表2 固定/稳定剂的研究与应用

Table 2 Research and application of solidification/stabilization agents

类型	土壤重金属 污染种类	参考文献
水泥+沸石	Pb、Cu、Cd、Zn	Ok et al., 2011
水泥+粉煤灰+生石灰	Pb、Cd、Zn	关亮等, 2010
赤泥	Pb、Cd、Zn	田杰等, 2010
家禽粪便堆肥	Cd	Chen et al., 2010
铁盐类、钙基类药剂	As、Pb	董慧等, 2021
粘土矿物(钠基膨润土)	Cu、Cd、Zn	杨秀敏等, 2009
沸石	Hg	张新艳和王起超, 2009

2012; Wang et al., 2014)。固定剂的种类和用量是影响固定/稳定化效果的重要因素,不同固定/稳定剂对不同重金属元素污染土壤修复效果不同,表2总结了一些目前重金属固定/稳定剂的研究与应用。

Fernández-Caliani et al.(2021)在西班牙一硫化物铜矿区使用甜菜石灰(sugar beet lime,一种糖提纯过程的副产品,可以缓解土壤酸性的石灰材料)和有机固体堆肥作为固定剂,通过深耕将其彻底混合到深度约为50 cm土壤中,发现经修复后矿区污染土壤中重金属含量有所降低(图4所示中Cu、Zn的中位数增加,可能是由于废石堆酸性径流中可溶性硫酸盐矿物的短暂沉淀所致)。固定/稳定化技术快速、有效、经济,在国外已经形成较完善的技术体系(谭科艳等, 2017),但它只改变了重金属元素在土壤中存在的形态,金属元素仍保留在土壤中,并不是一种永久的修复措施,修复后场地的后续利用可能使固化材料老化或失效,从而影响其修复效果(O'Day and Vlassopoulos, 2010)。

4.3.2 化学淋洗技术

化学淋洗技术是指通过淋洗剂或能促进土壤中重金属溶解和迁移的溶剂,将重金属从固相的土壤提取出来,再把富含重金属的废水进一步回收处理的土壤修复方法。目前常用的淋洗剂包括酸性溶液、盐溶液、螯合剂、表面活性剂、无机淋洗剂等(Sharma et al., 2018)。何苏祺等(2021)用无患子皂苷与柠檬酸复配淋洗剂修复铅锌矿区重金属污染土壤,发现在pH为5.0、淋洗时间为12.0 h时,土壤中Cd和Pb的去除率可达75.2%和41.6%。一般来说,酸性强或氧化性强或螯合能力强的淋洗剂对重金属阳离子的去除效果好,且淋洗剂浓度越高淋洗

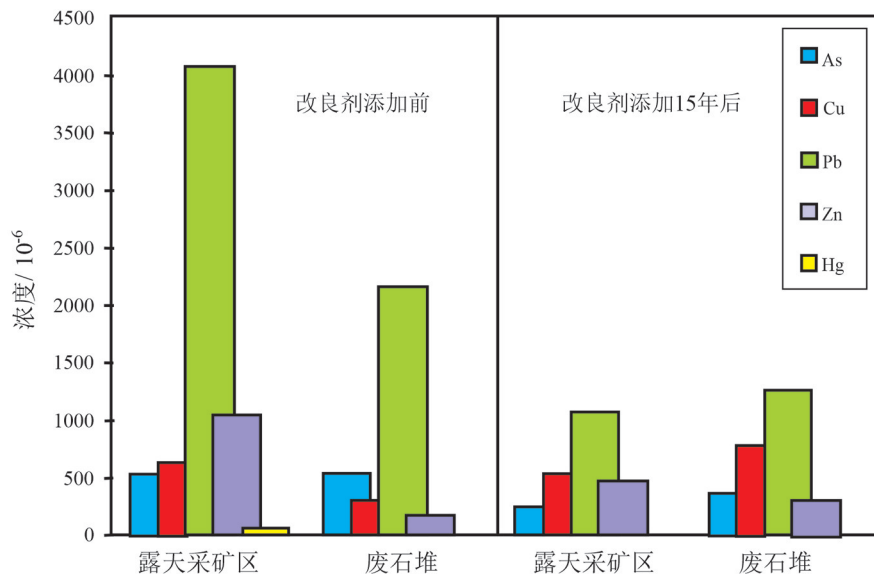


图4 修前后土壤重金属元素总浓度中位数比较柱状图(据Fernández-Caliani et al., 2021修改)

Fig. 4 Bar charts comparing the median total concentrations of trace elements in soil before and 15 years after the mine land reclamation (modified from Fernández-Caliani et al., 2021)

效果越好(朱健洸等, 2021),同时土壤的理化性质、淋洗时间、淋洗方式等也是影响土壤化学淋洗效果的重要因素。莫良玉等(2013)采用3种淋洗剂(硝酸铵、磷酸二氢铵、草酸铵)对受尾矿重金属污染严重的土壤进行修复,发现草酸铵去除土壤Zn能力最强,且随着淋洗次数和淋洗液浓度的增加,淋洗液中锌浓度不断降低。

化学淋洗技术效率高、易于操作、治理范围广、见效快(Baldissarelli et al., 2019)。但淋洗修复土壤时需要消耗大量的淋洗剂,不仅成本高,而且会产生大量的淋洗废液,对其处理和回收成为一大问题。其次,淋洗剂在淋出重金属的同时,势必会将土壤中其他元素洗脱出去,造成土壤中营养元素的流失,导致土壤肥力的下降(周智全等, 2016)。目前该技术关键是寻找一种既能提取各种形态的重金属,又不破坏土壤结构、降低土壤肥力,同时不会对环境造成二次污染的新型淋洗材料(孙涛等, 2015; Morillo and Villaverde, 2017; 曹明超等, 2019)。

4.4 生物修复技术

生物修复是指通过各种生物过程把土壤中的污染物分离或去除的技术,目前生物修复的常见方法有动物修复技术、植物修复技术和微生物修复技术。

4.4.1 动物修复技术

动物修复技术是指利用自然界某些动物的生

命代谢活动降低矿区污染土壤中重金属的含量,或通过改变重金属在土壤中的赋存形态从而降低其毒性(水新芳等, 2021)。目前对于可修复重金属污染土壤的动物研究较多的为蚯蚓、鼠类等,蚯蚓粪表面积较大,吸附能力较强,对Pb、Cd等重金属有一定的吸收能力(敬佩等, 2009; Singh et al., 2020)。矿区栖息蚁种的生态系统工程活动具有改善土壤质量、降低土壤重金属含量的潜力, Khan et al. (2017)等发现某煤矿采场长脚蚁(*Cataglyphis longipedem*)和短腹蚁(*Camponotus compressus*)两蚁穴栖息地附近土壤的总重金属含量和有效重金属浓度均显著低于其他土壤。

4.4.2 植物修复技术

植物修复技术是指在受污染土壤中科学种植适合植物,利用植物提取、吸收、分解、转化和固定土壤中有毒有害污染物,从而实现改善土壤理化性质、降低土壤重金属含量的目的(黄益宗等, 2013; Mahar et al., 2016; Khalid et al., 2017; Shah and Daverey, 2020)。植物修复技术包括植物固定技术、植物提取技术、植物挥发技术、根系过滤技术等(图5)。本文主要介绍植物提取技术和植物固定技术。

超富集植物是指相对于普通植物能在重金属浓度较高的土壤中生存,并对土壤中重金属有良好的吸附和富集效果的植物(Baker et al., 1994; 何玉

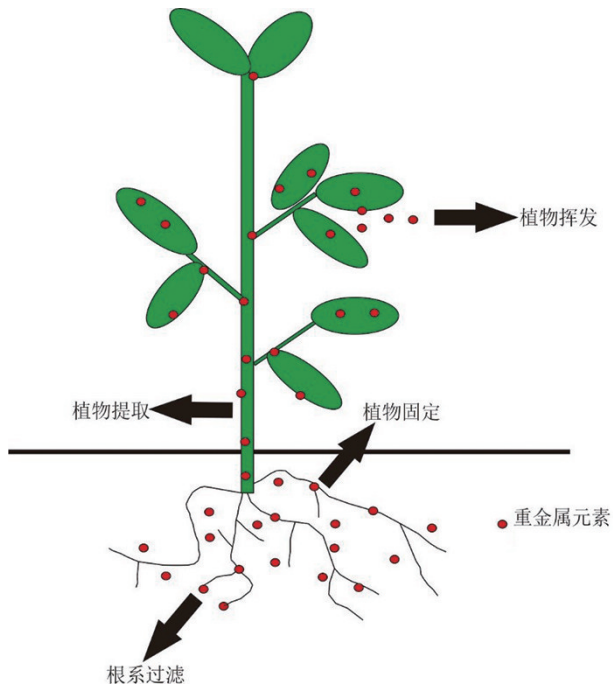


图5 植物修复土壤重金属污染示意图

Fig.5 Schematic diagram of phytoremediation of heavy metal contamination in soil

君等, 2020)。植物提取就是利用超富集植物对重金属污染物的吸收并在地上茎叶部分的积累, 通过收割地上部分并对其进行专门处置降低土壤重金属含量(沈振国和陈怀满, 2020; 赵云峰等, 2020)。现今世界上共发现了400多种超富集植物, 表3给出了一些已发现的重金属超富集植物。超富集植物通常都只能对一种重金属元素表现出富集能力, 仅少部分可以超富集吸收两种或两种以上的重金属元素(Wilson-Corral et al., 2012), 植物提取的关键在于因地制宜地筛选出超富集植物。中国在利用超富集植物提取重金属的研究领域工作开展较晚, 目前国内报道的主要为As、Zn、Cd、Mn等元素的超富集植物(韦朝阳等, 2002; Qiu et al., 2006; Zhang et al., 2010; 郑太辉等, 2015)。Chen et al. (2002)在生长于湖南省石门雌黄矿矿区的大量植物中筛选并发现蜈蚣草对As具有超强的富集能力, 通过刈割可以提高其对砷的去除能力, 蜈蚣草是国际上首次报导的砷超富集植物, 在砷污染土壤的修复和植物学研究中具有重要价值。

植物固定是指通过建立耐受性植物覆盖层, 将重金属固定或沉淀在土壤周围, 不向地上部分迁移

表3 Zn、Cd等重金属的超富集植物

Table 3 Hyperaccumulator of Zn, Cd and other heavy metals

金属元素	超富集植物	文献来源
Zn	Thlaspi caerulescens(天蓝遏蓝菜)	陈一萍, 2008
	Festuca arundinacea(高羊茅)	
Cd	Cardamine hupingshanensis(壶瓶碎米荠)	李非里等, 2021
	Solanum nigrum(龙葵)	
Ni	Leersia hexandra Swartz(李氏禾)	张学洪等, 2006
	Thlaspi caerulescens(天蓝遏蓝菜)	
As	P. vittata(蜈蚣草)	Chen et al., 2002
	Pteris cretica(羽叶鬼针草)	
Pb	Bidens maximovicziana(黑麦草)	陈三雄等, 2011
	C. ambrosioides(土荆芥)	
Mn	Polygonum hydropiper(水蓼)	王华等, 2007
	Phytolacca acinosa(商陆)	
Cr	Dicoma niccolifera(尼科菊)	杨晓琼, 2017
	Sutera fodina Wild(铬线蓬)	

(Sylvain et al., 2016; 谢东等, 2019)。用于植物稳定的植物不仅要耐重金属, 并且从根到地上部分的迁移率应该相对较低。由于土壤理化性质的不同和矿区污染类型的差异, 要有针对性地选取特定的植物进行固定修复, 表4给出了一些不同矿区重金属污染土壤耐受植物及一些使用植物固定矿区污染土壤中重金属元素所需的改良剂。

植物提取技术成本高、修复时间长, 主要应用于土地价值高及可以等待较长时间恢复的地区。植物固定技术成本低、易操作, 但植物固定并不是从污染土壤中去掉重金属, 所以当土壤环境发生变化时, 稳定的重金属可能被重新活化(Bolan et al., 2011; Ali et al., 2013)。植物修复作为一种不破坏土壤结构、不引起二次污染的土壤污染治理技术, 在重金属污染土壤治理方面有广阔的发展前景(Shah and Daverey, 2020; 肖鹏飞和吴德东, 2021)。但单一的植物很难完成对复合污染的修复, 因此寻找富集重金属种类更多、累积能力更强的植物或植物组合是植物修复技术的研究重点。

4.4.3 微生物修复技术

微生物修复是指微生物通过吸附、沉淀、淋滤、氧化还原等方式参与土壤中重金属的转化, 降低土壤中重金属的浓度和活性, 改变土壤中重金属的赋存状态, 将重金属转化为无毒或低毒的化合物形式的修复方法(郭维君等, 2010)。微生物修复技术包

表4 植物固定修复土壤重金属所采用的植物及相应的改良剂

Table 4 List of hyperaccumulator plant species for phytoextraction and corresponding amendments

植物	重金属污染	改良剂	位置	参考文献
Artemisia sacrorum Ledeb(白莲蒿)	Pb	-	白音诺尔铅锌矿区	赵磊, 2009
Cynodon dactylon L.(狗牙根)、 Neyraudia reynaudiana(Kunth)Keng. (类芦)、Pennisetum purpureum Schum. (狼尾草)、Eucalyptus robusta Smith. (大叶桉)	Cu、Pb、Zn、Cd	石灰、鸡粪	广东大宝山矿区	Yang et al., 2016
Festuca rubra(紫羊茅)	Pb、Zn	家禽粪便、造纸污泥与灰泥 混合	西班牙巴斯克地区一废弃铅 锌矿	Burges et al., 2016
Pistacia terebinthus(黄连木)、Pinus brutia(土耳其松)	Cu	葡萄酒废料、鸡粪	塞浦路斯Skouriotissa铜矿区	Johansson et al., 2005
Echinops gmelina Turcz(砂蓝刺头)、 Crepis flexuos(弯茎还阳参)、 Caragana densa Kom(密叶锦鸡儿)	Cu、Ni、Co	-	金昌镍铜矿区	廖晓勇等, 2007

括生物稳定、生物薄膜吸附、生物淋滤等方式。生物稳定是利用细菌活动将土壤中溶解态重金属元素转化为更加稳定的结合态,降低其活性和生物利用度的方法(黄春晓,2011)。研究发现从香蒲根际中分离出的一些菌株能钝化固定土壤中的Cu和Cd,降低它们在土壤中的可交换态含量(Tiwari et al., 2008)。生物薄膜是一种高效的生物修复工具,有研究发现利用微藻生物膜的吸附作用可以去除矿山废石堆渗滤液中的Zn(Li et al., 2015)。生物淋滤是利用特定微生物或其代谢产物的氧化、还原、络合、吸附或溶解作用,将土壤中重金属浸出并再回收的方法(Solisio and Lodi, 2002; Kumar and Nagendran, 2007)。经微生物修复后的土壤理化性质基本保持不变,对环境影响很小。但微生物修复技术也具有一定的局限性,微生物需要一定的生长条件和生活环境,且微生物修复技术的周期相对较长,这限制了微生物修复的大范围运用或推广;同时加入到修复现场中的微生物可能会与土著菌株竞争或难以适应环境从而导致实际作用与实验结果有较大出入(余天红和黎华涛, 2014)。

5 联合修复技术

与单一的物理修复、化学修复和生物修复技术相比,联合修复技术(如植物-微生物联合修复、化学-植物联合修复、化学-微生物联合修复等)同时具备多种修复技术的优势和特点,修复效率相对会比较,尤其针对重金属复合污染严重的

矿区土壤修复治理效果突出。邓敏等(2021)以硫酸盐还原菌为修复微生物,以低分子有机酸为络合剂,对攀西矿区典型重金属污染土壤开展了化学-微生物联合修复,结果表明:与单一的化学修复技术相比,化学-微生物联合修复技术对矿区土壤中铅、镉含量有着显著的钝化效果,土壤中镉交换态含量最大降低了60.7%。叶攀骅等(2016)利用改良剂和超富集植物对锰污染土壤进行联合修复,发现添加改良剂(海泡石和沸石)能显著提高植物茎叶中的Mn含量,表明海泡石等改良剂可以与植物联合使用,改良重金属土壤修复效果。崔照豪(2018)采用植物-微生物联合技术降低尾矿的重金属含量,发现接种土著微生物菌剂能够促进地肤、波斯菊和紫羊茅这3种重金属耐受植物的生长,从而有效提高重金属的去除效率。目前运用多种修复技术结合的联合修复技术是土壤生态恢复新的研究方向,同时,随着遥感技术的研究进步,遥感监测重金属污染土壤修复的研究及应用也会有很大的提升(宋文等,2021),它将为矿区重金属污染土壤的高效、低耗修复提供更优解法。

6 结论

矿区土壤重金属污染形式日益严峻,生态风险不断增加,迫切需要采取土壤修复措施,降低土壤重金属含量或活性。过去数十年间,世界各国已陆续研发出了多种修复技术。本文综述了这些修复技术的原理、制约因素、优缺点和适用范围等。目前,传统的修

复技术相对常见,物理修复方法耗时短、效率高,对土壤重金属复合污染修复较为有效;化学修复方法方便实用,发展前景广阔。但部分物理化学修复技术如电动法、淋洗法等,也会存在破坏土壤的理化性质和营养元素等问题,因此在进行实际操作时,要结合重金属污染土壤的性质与未来土地规划,选择最合理的修复技术。生物修复技术虽然安全环保,不会造成二次污染,但修复时间过长、植物耐受的重金属元素过于单一、修复过程不确定因素过多等都成为制约其进一步发展的阻碍。因此,在现有修复技术基础之上,因地制宜地发展适用范围广、二次污染小、高效实用、成本低新型土壤修复技术,合理运用多手段结合的联合修复技术是未来土壤重金属污染修复领域的研究热点。

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