



含铬污水的生物修复技术研究现状及展望

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[摘要] 六价铬被广泛应用于工业生产。含铬污水排入环境, 在污染周边水源和土壤的同时, 还可通过生物富集和生物放大作用威胁人类健康, 因此高效处理含铬污水是世界环境科学领域亟待解决的问题。传统的铬修复技术一般采用物理化学处理方法, 但其具有处理程序复杂、成本较高、效率低、容易导致二次污染等缺点。生物修复技术则利用植物、动物和微生物, 通过吸收、降解、转化等途径减少污水中的铬, 具有经济、无二次污染、修复效率高、过程安全等优点, 因此被认为是处理含铬污水的资源节约型、环境友好型技术。就微生物还原, 微生物吸附, 植物、纳米材料与微生物联用等现有处理含铬污水的生物修复技术的作用机制、反应机理及优缺点进行了归纳总结, 揭示其应用潜力, 并对其未来的发展趋势进行展望。

[关键词] 含铬污水; 生物还原; 生物吸附

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Research status and prospects on bioremediation technologies of chromium-containing wastewater

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Abstract: Hexavalent chromium is commonly used in industrial manufacturing. The outflow of chromium-containing wastewater into the environment not only pollutes the surrounding water source and soil but also endangers human health through bioaccumulation and biomagnification. Therefore, the efficient treatment of chromium-containing wastewater has emerged as a critical issue in environmental science around the world. Traditional chromium remediation technology is physical and chemical treatment method, but it has drawbacks such as a complex treatment procedure, high cost, limited efficiency, and the potential for secondary contamination. Bioremediation technology uses plants, animals and microorganisms to reduce chromium in sewage through absorption, degradation and transformation. Because of the benefits of economy, no secondary pollution, high restoration efficiency, and process safety, it is regarded as a resource-saving and environmentally sustainable technology for dealing with chromium-contained wastewater. In present work, the mechanism, reaction theory, advantages and drawbacks of current bioremediation technology for hexavalent chromium pollution, such as microbial reduction, microbial adsorption, phyto-microbial remediation, and nano-microbial remediation were summarized. Its implementation potential and future development pattern was prospected.

Key words: chromium-containing wastewater; bioreduction; biosorption

环境中的 Cr(VI) 主要源于金属制品、冶金、皮革制造、电镀、木品保藏和染料生产等行业的污水排放^[1-2]。根据中国环境监测总站发布的《2020 中国海洋生态环境状况公报》可知, 我国 2019 年工业污水

中 Cr(VI) 排放量达 489.9 kg。这些含 Cr(VI) 污水排入环境, 严重污染周边水源和土壤。

Cr(VI) 具有高水溶性, 可在自然界中长期存在^[3], 即便是低浓度的 Cr(VI) 也可通过生物富集和

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生物放大作用对人类健康产生威胁^[4]。由于Cr(VI)具有较高的致突变性、致癌性和致畸性^[5],国际癌症研究机构(IARC)和联合国世界卫生组织(WHO)已将其列为一类致癌物。可见,含Cr(VI)污水已严重威胁生态环境和公众健康,高效处理含Cr(VI)污水已成为世界环境科学领域亟待解决的问题。

目前,污水中的Cr(VI)主要通过物理化学方法被转化为Cr(III)后,形成溶解度较低的Cr(OH)₃沉淀^[6]而被去除。但该过程处理程序复杂、成本高、效率低,且容易导致二次污染^[3]。生物修复技术是指利用植物、动物和微生物,通过吸收、降解、转化等途径减少水体中污染物的环境污染治理技术。因经济、无二次污染、修复效率高、过程安全等优点,生物

修复技术在处理含Cr(VI)污水方面展示出巨大潜力。笔者就现有含Cr(VI)污水生物修复技术进行归纳总结,详细地分类阐述了其作用机制、反应机理,进一步分析了不同修复技术的优缺点及其应用潜力,并指出未来该领域可能的发展方向。

1 微生物还原

Cr(VI)的微生物还原是指微生物通过氧化还原反应将Cr(VI)还原为Cr(III)。目前,已经有大量具有Cr(VI)还原能力的菌株被筛选出来,部分代表性菌种如表1所示。根据细菌还原机理的不同,可将其大致分为3类,即铬还原菌(Chromium reducing bacteria, CRB)、硫酸盐还原菌(Sulfate-reducing bacteria, SRB)和铁还原菌(Iron reducing bacteria, IRB)。

表1 部分具有Cr(VI)污染修复潜力的微生物
Table 1 Some microorganisms with potential to repair Cr(VI) pollution

物种	微生物名称	修复机制	修复能力	温度/ ℃	pH	其他	参考文献
细菌	大肠杆菌(<i>Escherichia</i>)	还原、吸附	可还原小于400 mmol/L的Cr(VI)	35	5	主要通过ChrR和YieF还原Cr(VI)	[7]
	普罗维登斯菌(<i>Providencia</i>)	还原	100%还原100~300 mg/L的Cr(VI)	37	7	还可耐受镍、锌、汞、铅、钴等	[8]
	布鲁氏菌(<i>Brucella</i>)	还原	还原94.1% 100 mg/L的Cr(VI)	37	7	还可耐受镍、锌、汞、铅、钴等	[9]
	节杆菌(<i>Arthrobacter</i>)	还原、吸附	24 h可去除88.6% 100 mg/L的Cr(VI)	28	4	最佳接种量为5 g/L	[10]
	芽孢杆菌(<i>Bacillus</i>)	还原	48 h可还原90% 200 mg/L的Cr(VI)	37	7.5	可耐受镉、铜、钼、镍、铅等金属	[11]
	纤维素微菌(<i>Cellulosimicrobium funkei</i>)	还原、吸附	120 h内100%还原200 µg/L的Cr(VI)	35	7	可耐受0~2%的NaCl;细菌还原产物对斑马鱼的毒性作用较小	[12]
真菌	青霉菌(<i>Penicillium</i>)	还原、吸附	2 d内去除89.1% 164.6 mg/L的Cr(VI)	30	3.6	pH先升高到4.2再降低至2.9	[13]
	黄曲霉菌(<i>Aspergillus flavus</i>)	还原、吸附	72 h内100%还原100 mg/L的Cr(VI) 72 h内对250 mg/L的Cr(VI)吸附量达7.51 mg/g	30	5.1	最高可耐受500 mg/L Cr(VI);对250 mg/L Cr(VI)内化量达6.17 mg/g	[14]
	拟青霉菌(<i>Arthrobacter viscosus</i>)	还原、吸附	7 d可去除96% 50 mg/L的Cr(VI)	28	4	最佳接种量为38 mg/L	[15]
酵母	南极酵母隐球菌(<i>Cryptococcus laurentii</i>)	吸附	可100%去除10 mg/L的Cr(VI)	22	5	pH先升高到5.3再降低至2.5;还可吸附汞、镍、铅等其他重金属	[16]
	假丝酵母(<i>Candida</i>)	吸附	2 h内可100%去除20 mg/L的Cr(VI)	30	2	最佳接种量为3.60 g/L	[17]
藻类	小球藻(<i>Chlorella</i>)	吸附	48 h可去除50.24% 27.48 mg/L的Cr(VI)	28	3	最佳接种量为1×10 ⁶ 细胞/株	[18]
	绿藻门(<i>Chlorophyta</i>)	吸附	90 min可吸附96.81% 150 mg/L的Cr(VI)	30	1	最佳接种量为6.0 g/L	[19]

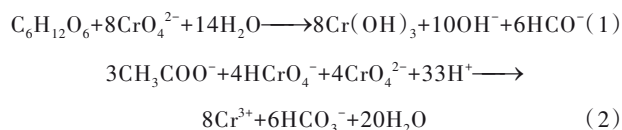
1.1 铬还原菌

在好氧环境下,CRB主要通过ChrR和YieF两种还原酶将Cr(VI)还原为Cr(III)。这些酶通常存在于大肠杆菌(*Escherichia*)、假单胞菌(*Pseudomonas*)、芽孢杆菌(*Bacillus*)和肠杆菌(*Enterobacter*)中^[20]。ChrR和YieF的氨基酸衍生序列中均具有NADH₂蛋白家族的特征序列,这些氨基酸可能参与黄素单核苷酸(FMN)的结合,使得CRB能够利用NAD(P)H作为电

子受体将Cr(VI)还原为Cr(III)^[7]。

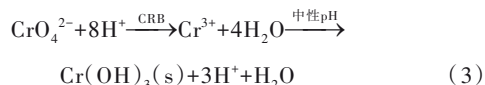
在厌氧环境下,CRB则主要通过细胞色素c,以Cr(VI)作为终端电子受体还原铬酸盐。细胞色素c通过电子传递系统催化电子沿着呼吸链穿梭,以Cr(VI)作为电子受体从而将其还原为Cr(III),并在菌体表面形成不溶性铬酸盐沉淀^[21]。无氧条件下,在呼吸链中可作为Cr(VI)电子供体的物质种类很多,包括碳水化合物、脂肪、氢、NAD(P)H和其他内源性电子

储备物质,如泛醌、细胞色素、金属离子、生物素等。其中以葡萄糖作为电子供体的还原反应见式(1),以醋酸作为电子供体的反应见式(2)^[22-23]。



部分菌种在好氧和厌氧环境下均表现出对Cr(VI)的还原能力^[23],如荧光假单胞菌LB300(*Pseudomonas fluorescens* LB300)、无色菌(*Achromobacter*)、二歧假单胞菌(*Pseudomonas ambigua*)、普罗维登斯菌(*Providencia*)、布鲁氏菌(*Brucella*)、芽孢杆菌(*Bacillus*)等。

事实上,虽然CRB还原Cr(VI)的机制各不相同,但都依托于胞内具有还原功能的酶或胞外聚合物进行氧化还原反应。发生氧化还原反应时会消耗大量质子,导致本底pH升高,从而有利于Cr(OH)₃沉淀的形成。其还原机制可总结为式(3)^[22]。



1.2 铁还原菌和硫酸盐还原菌

上述的CRB通过酶促反应或功能蛋白传递电子直接还原Cr(VI),而IRB和SRB则分别通过产生的代谢副产物Fe(II)和H₂S间接还原Cr(VI),且二者还原速度为CRB的100倍^[22]。

IRB主要为希瓦氏菌(*Shewanella*),其中*Shewanella oneidensis* MR-1的还原效率最为突出^[24],菌株将体系中Fe(III)还原为Fe(II),然后Fe(II)再将Cr(VI)还原为Cr(III)。SRB主要为脱硫弧菌和假单胞菌等,其主要在无氧的情况下还原Cr(VI)^[25]。在希尔登伯勒普通脱硫弧菌(*Desulfovibrio vulgaris* Hildenborough)中,存在着具有还原Cr(VI)能力的氢化酶[Fe]、[NiFe]、[NiFeSe],其中[Fe]的还原效率最高,约为细胞色素的10倍^[26];而施氏假单胞菌KC(*Pseudomonas stutzeri* KC)则是以吡啶-2,6-二硫代羧酸作为硫酸盐转运体,先消耗硫酸盐生成HS⁻,再通过HS⁻将Cr(VI)还原为CrS₃或Cr(OH)₃沉淀^[27]。

2 微生物吸附

微生物吸附是金属离子通过与羧基、羟基、巯基、酚醛、胞外聚合物间的相互作用而吸附在微生物表面的现象^[28]。该过程不依赖于微生物的生物代

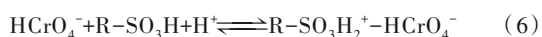
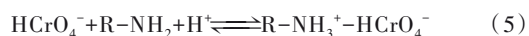
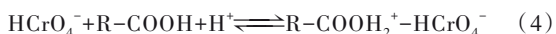
谢,是一个被动的过程,故无需考虑微生物的活性状态^[29]。因此,微生物吸附剂在工业污水处理中有广阔的应用前景。目前,真菌、细菌、酵母和微藻类均表现出了吸附Cr(VI)的能力。

真菌对Cr(VI)有较高的耐受能力,因此成为主要的Cr(VI)微生物吸附剂。例如念珠菌(*Candida* sp.)、拟青霉菌(*Paecilomyces lilacinus*)、曲霉菌(*Aspergillus* sp.)、根霉菌(*Rhizopus rhizus*)、青霉菌(*Penicillium* sp.)等均对Cr(VI)有显著的吸附性^[28]。S. VAJPAI等^[14]通过SEM-EDX、TEM-EDX检测发现黄曲霉菌(*Aspergillus flavus*)可吸附Cr(VI)于菌丝表面并在胞质内蓄积,经FTIR结果证实其参与吸附的主要成分为胺、氢氧化物、磷脂、羧基、蛋白质和碳水化合物等,类似的结果也出现在其他真菌中^[13, 30]。

除真菌外,细菌和酵母也可以吸附Cr(VI)。相对于真菌,细菌有更快的生长速度,酵母则对营养物质的需求更低。黏节杆菌(*Arthrobacter viscosus*)、枯草芽孢杆菌(*Bacillus subtilis*)及中型假丝酵母(*Candida intermedia*)、毕赤酵母(*Pichia guilliermondii*)和异常威克汉姆酵母(*Wickerhamomyces anomalus*)均被证实对Cr(VI)具有吸附能力^[31-32]。通过FTIR分析显示,细菌的细胞膜表面有羧基、烷烃、酰胺、胺、羟基、磷酸盐、磺酸盐、羰基、咪唑、磷酸二酯等多种成分,它们是肽聚糖、脂多糖等大分子的组成部分^[33]。在革兰氏阴性菌中,脂多糖层下有一层薄薄的肽聚糖,这两层均参与Cr(VI)的吸附,而革兰氏阳性菌细胞膜上只有一层厚厚的肽聚糖^[34]。经SEM-EDX和FTIR检测发现,细菌吸附Cr(VI)后,其表面分子组成会发生变化,引起细胞的聚集,并促进细胞外表面脂多糖的分泌,这可能源于细菌对Cr(VI)的自我防御机制^[12]。相对于细菌的表面结构,酵母的“三明治”结构具有较大的比表面积,能够通过静电黏附、氧化还原和配位螯合等方式捕获并结合具有活性位点的Cr(VI)。Baoyan HE等^[17]通过AFM图像发现酵母在吸附Cr(VI)后菌体聚集形成网状结构,经FTIR检测其表面有—OH、N—H、C—H、C—C、C—O、C—N、C=O、O=C—N、C—O—S和PO₂⁻等化学键或化学基团,表明主要由碳水化合物、蛋白质、磷脂、甲壳素等成分参与酵母对Cr(VI)的吸附^[16]。

关于藻类吸附Cr(VI)的研究较少,主要集中在小球藻(*Chlorella*)、斜生栅藻(*Scenedesmus*)、聚孢子藻(*Synechocystis*)和螺旋藻(*Spirulina*)上^[35-36]。J. JAAFARI

等^[37]发现,通过优化工艺参数,*Chlorella*可有效吸附97.8%的Cr(VI)。王岩等^[19]在Cr(VI)溶液中培养绿藻,其在酸性条件下,180 min内可以去除92.10%的Cr(VI)。Y. A. R. LEBRON等^[38]通过SEM-EDX检测发现藻类呈均匀光滑的球形或楔形,经H₃PO₄和ZnCl₂等化学修饰剂修饰后,其表面变得更加粗糙多孔,在增加其吸附面积的同时,还会增加其表面碳水化合物、脂类和蛋白质的含量,从而增强其对Cr(VI)的吸附性。ATR-FTIR测定显示,藻类表面主要存在羟基、羧基、胺基、酰胺基和木质素等,其中羧基和木质素在藻类吸附中起到关键作用,这就使得藻类可以首先通过细胞外多糖、羧基、羟基、硫酸盐和磷酸基将Cr(VI)吸附于细胞壁上,然后通过细胞膜的主动运输使Cr(VI)与蛋白质和其他细胞内成分结合,其反应的主要机制见式(4)~(6)^[18]。



微生物吸附处理含Cr(VI)污水的主要优势是经济、方便和便于回收Cr(VI)。吸附Cr(VI)的主要官能团均存在于微生物的细胞壁,微生物对Cr(VI)的抗性不是必要条件,因此无需考虑Cr(VI)浓度对微生物存活率的影响。但该技术在商业化应用时,需要配套相应的生物反应器及其处理工艺,以解决生物修复剂运输不便和需单独培养等问题。

3 复合生物修复

3.1 植物-微生物联用

植物修复可利用植物和根系微生物清除Cr(VI),阻止含铬污水中Cr(VI)迁移到地下水或食物链中,是一种比较新的技术,也被称为“绿色修复”^[39-40]技术。目前发现符合要求的模式物种种类繁多,据报道,狼尾草、折臂草、芦苇、菹草、李氏禾、雷公藤、刺荨麻、油菜、印度芥菜和玉米等植物已纳入实验室盆栽实验^[41-43]。虽然较细菌等其他微生物而言,植物修复可以通过根系吸收Cr(VI),并在地表以上的器官中积累100~1 000倍浓度的Cr(VI),但依旧无法避免其生长周期长的限制^[44]。植物生长促进菌(Plant-growth promoting bacteria, PGPB)可以通过改变植物的根系结构促进植物的生长,提高植物对Cr(VI)的吸收能力,降低Cr(VI)对植物的毒性,同时植物也会分泌碳水化合物、氨基酸、黄酮类化合物等来刺激PGPB,使其活性提高10~100

倍,这使得植物修复应用的潜力大大提高^[45-46]。因此,植物-微生物联用来提高植物修复效率的修复技术受到越来越多的关注。

植物-微生物联合修复相比传统修复技术的优势在于其具有较好的成本效益、舒适性和大规模的适用性,并且可以保持生物群的完整。但目前这项技术并未投入应用,依然处在实验室研究状态。

3.2 纳米材料-微生物联用

纳米材料因其体积小、溶解度好、比表面积大、表面成分丰富而成为环境修复的理想候选材料^[47]。但单一运用纳米材料处理含Cr(VI)污水面临着材料回收困难、易被氧化,存在二次污染,具有生物毒性等诸多问题,因此将微生物与纳米材料结合修复污染的概念被提出^[48]。K. V. G. RAVIKUMAR等^[49]将纳米零价铁(nZVI)复合膜和SRB相结合,在厌氧条件下最高可去除65.19%的Cr(VI)。微生物虽然对Cr(VI)有良好的吸附和还原能力,但在高浓度Cr(VI)环境中活性受到抑制,因此可以先利用纳米材料良好的吸附能力降低水体中的Cr(VI)浓度,从而保证微生物的存活,进一步提升其去除Cr(VI)的能力。纳米材料-微生物联用技术已经处于发展阶段,未来具有一定的应用前景。

4 结语

生物修复技术具有经济、无二次污染、修复效率高、过程安全且便于操作等诸多优势,已成为处理含Cr(VI)污水领域的研究热点。现今生物修复技术理论研究已趋于完善,未来的研究重心将会逐渐向实际应用方面转移,可能的发展方向包括:

(1)开展基础研究。探究生物修复菌株功能基因及相关蛋白,进一步为生物修复Cr(VI)的机制研究和提高生物修复技术对Cr(VI)的处理能力提供理论基础。

(2)开展优化实验。探究生物所需营养物质及其必要元素,并优化其理化参数,寻找合适的培养溶剂,使其在投入应用后能够维持正常的生长并达到最佳修复治理条件。

(3)开展应用研究。构建高效表达功能蛋白的工程菌株,或提取胞外聚合物直接应用,或将多种生物修复技术联合应用,高效处理含Cr(VI)污水。

(4)开展工艺研究。对生物修复反应器及其处理工艺进行研究,保证生物在可以达到处理效果的前提下也能与现有污水处理工艺良好结合,最大限度地降低运营成本。

(5)开展安全评价。对生物进行安全性评价,避免其本身或代谢产物对生态环境及人群健康产生危害。

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