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生物炭固定化菌复合材料在环境修复中的应用研究进展

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摘要:随着工业和农业的快速发展,环境污染日益严重,对生态安全和人体健康形成严重威胁.微生物修复是一种低价、绿色和易操作的技术,但微生物对高浓度污染物耐性低,造成修复效果差和周期长.由于生物炭的孔隙率高、官能团丰富和比表面积大,被认为是一种良好的吸附剂和固定化载体.通过微生物固定化技术将菌群定殖于生物炭上而制备生物炭固定化菌复合材料,结合了二者优势又克服菌群的缺点,展现了巨大的应用前景.通过系统论述生物炭固定化菌复合材料的制备方法和特征,评价生物炭固定化菌复合材料在环境修复中的效能,讨论环境因素对生物炭固定化菌复合材料去除水中污染物和修复污染土壤的效果影响,阐明生物炭固定化菌复合材料对废水处理与土壤修复的作用机制.

关键词:菌群;生物炭;微生物固定化技术;去除效果;修复机制

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Research Progress in Application of Biochar-immobilized Bacteria Composites in Environmental Remediation

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Abstract: With the rapid development of industry and agriculture, environmental pollution has become increasingly serious, threatening ecological security and human health. Microbial remediation is a cheap, green, and easy-operation technology, but the tolerance of microorganisms to high concentrations of pollutants is low, resulting in low remediation efficacy and a long cycle. Biochar is a good adsorbent and immobilized carrier because of its high porosity, abundant functional groups, and large surface area. Biochar immobilized bacteria composites were prepared by immobilizing bacteria on biochar, which combined their advantages and overcame the disadvantages of the bacteria, displaying a huge application prospect. In this study, the preparation methods and characteristics of biochar-immobilized bacteria composites were reviewed, their effectiveness in environmental remediation was evaluated, the effects of environmental factors on the removal of pollutants in water and the remediation of contaminated soil were discussed, and their mechanism for wastewater treatment and soil remediation was clarified.

Key words: bacteria; biochar; microbial immobilization technology; removal efficiency; remediation mechanism

随着我国经济的快速发展,工、矿、农业等行业产生的废水和废渣不合理处置导致水和土壤环境污染问题日益严重[1.2].针对重金属、有机污染物和营养盐污染,微生物可以通过新陈代谢以及吸附、迁移、转化等活动降解或去除此类污染物.微生物修复技术具有环境友好型、成本低和效果好的优点[3].然而,加入到环境中的游离微生物易受到温度、氧气和水等因素以及土著微生物竞争的影响,导致游离微生物在实际环境中的存活能力以及活性有所降低,致使其修复效果不佳[4].

微生物固定化是解决上述问题的技术之一,通过化学或物理方法将游离微生物固定在载体材料上,从而提高微生物的活性^[5,6].微生物固定化技术的关键要素是固定化方法和载体的选择.根据载体与微生物之间的作用力和结合形式的不同,固定化方法主要有吸附法、包埋法、交联法、共价结合法和复合固定法^[7].按照固定化载体特性和组成不同,载体材料可分为无机载体、有机高分子载体和复合

载体等^[8]. 近几年来,生物炭被认为是一种固定微生物的优良载体.

生物炭具有大比表面积、丰富官能团和多孔结构,可用于微生物的固定化载体[10].大比表面积和多孔结构的生物炭可以富集微生物,为微生物提供栖息地,提高微生物的活性[10].生物炭多孔结构和丰富的官能团的吸附作用,固定微生物的同时吸附污染物,使得污染物与微生物的接触几率增加,从而促进微生物对污染物的降解[11].另外,生物炭中的有机质、氮、磷和钾等可为微生物生长繁殖提供所需营养物质.生物炭表面的活性官能团对重金属的还原和有机物的氧化降解方面也表现出巨大的潜力[12].所以,生物炭不仅是理想的固定微生物的载

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体,而且是一种优良的修复剂.

本文将总结生物炭固定化菌复合材料的制备方法、表征技术及其在环境修复中的应用,以期为今后其在废水处理与污染土壤修复中的应用提供一定参考.

1 生物炭固定化菌复合材料的制备

1.1 生物炭的制备

生物炭是生物炭固定化菌复合材料的主要构成之一,其制备条件(热解温度、热解时间和氮气条件等)与原材料(秸秆、木屑和污泥等)会对生物炭的特性(比表面积、孔隙结构、官能团和稳定性等)产生影响,进而影响菌群的固定化量.Liu等[13]利用不同热解温度(200、300、400、500和600℃)下制备的生物炭固定化石油烃降解菌,其对石油烃的降解率分别为43.04%、72.79%、80.54%、83.95%和85.81%;600℃下获得的生物炭作为载体,对石油烃的降解率最大.因此,生物炭作为菌群载体,其制备条件和原材料需考虑.

1.2 菌群的优选

菌群也是生物炭固定化菌复合材料的主要组成之一,参与污染物的修复,显著影响生物炭固定化菌复合材料对污染物的降解效果.针对目标污染物,筛选和驯化特异菌群,将其定殖于生物炭上,将会极大提高生物炭固定化菌复合材料对污染物的降解效率.针对地下水环境中石油烃 $C_{10} \sim C_{40}$ 成分,沈若非等^[14]筛选出了两株高效降解石油烃的菌株(赖氨酸芽孢杆菌属和杆菌属),将其固定化于生物炭上制备复合材料,对石油烃的降解率可达 70.51%.因此,特异菌群的筛选和驯化对制备高效生物炭固定化菌复合材料尤为重要.

1.3 固定化方法

微生物固定化方法将直接影响固定到载体上的 微生物量及间接影响对污染环境的修复效果.目前, 主要采用吸附法、包埋法和吸附-包埋复合法制备生 物炭固定化菌复合材料^[15].

吸附法是依靠微生物和生物炭表面的作用力(氢键、范德华力和电荷作用力等)而吸附结合在一起.首先通过热解生物质制备生物炭,然后生物炭与菌液按照一定的比例混合,吸附固定化一定时间,最后离心而得到生物炭固定化菌复合材料(图1).陈壮^[16]将改性芦苇生物炭置于接入2%铬耐性菌的液体培养基中,在30℃下吸附固定18h,然后离心、过滤和洗涤后获得改性生物炭固定化铬耐性菌复合材料.吸附法操作简单、环保经济和传质性能好,但稳定性差且细胞容易解吸而泄漏.

包埋法是利用聚乙烯醇和海藻酸盐等高分子聚合物等将微生物和生物炭包裹在凝胶网状结构中而形成黑色微珠(图1). Teng等[17]利用纳米零价铁/生物炭复合材料作载体,与溶磷菌液同时添加到海藻酸钠/聚乙烯醇溶液中,之后将上述混合溶液滴入CaCl₂溶液中而获得了生物炭负载纳米零价铁复合材料固定化溶磷菌微珠. 包埋法是一种不可逆、低成本和易操作的方法,既能减少微生物在固定化过程中的泄漏,又能使微生物不受外界因素影响而能保持高活性. 但是,固定微生物量过高时,影响微生物的物质交换,降低传质效率.

复合固定化法是通过不同固定化方法联合负载微生物到载体上的过程.目前,最常用的复合固定法为吸附-包埋法:首先将生物炭与菌群充分混合,菌群被吸附到生物炭后,再将其加入到聚乙烯醇/海藻酸盐进行包埋固定化(图1).杨雅茜[18]以制备的矿物-生物炭为固定化载体,首先将其与筛选的耐铬菌混合进行吸附固定化,离心取出后将其加入到海藻酸钠溶液中,最后将上述混合液滴到CaCl.溶液中,即可获得生物炭固定化菌微珠.根据不同的污染情况,选择不同的固定化方法结合,使其优势互补,可得到性能优异和廉价易得的固定化微生物复合材料.

2 生物炭固定化菌复合材料的表征

为更好地了解生物炭固定化菌复合材料的理化特性以及去除污染物的机制,对其进行表征分析具有重要意义.常用表征技术有:扫描电镜(SEM)、比表面积分析测试仪(BET)、傅里叶变换红外光谱仪(FTIR)、X射线衍射仪(XRD)和磁共振仪(NMR)等.

2.1 SEM 分析

SEM可以观察生物炭固定化菌复合材料表面形貌和微观结构. Wang 等[19]利用 SEM 观察铜绿假单胞菌、铁改性生物炭以及铁改性生物炭固定化菌微球.通过 SEM 观察到铜绿假单胞菌长度约为 2 μm, 呈杆状且有鞭毛;固定化的菌群在铁改性生物炭表面繁殖良好,这是因为优选的铁改性生物炭表面的官能团和粗糙结构为菌群生长提供了良好的生存环境;微球表面存在大量孔隙,为污染物的传质和降解创造了有利条件;图中显示菌群在球体内繁殖良好,表明铜绿假单胞菌与铁改性生物炭复合成功.

2.2 FTIR 分析

FTIR可用来鉴别生物炭固定化菌复合材料表面 官能团的种类及分析官能团的变化. Huang等^[20]采用 FTIR分析了生物炭固定化蜡样芽孢杆菌复合材料吸

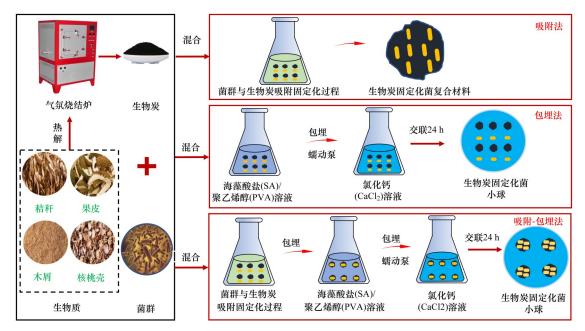


图1 吸附法、包埋法和吸附-包埋法制备生物炭固定化菌复合材料的流程

Fig. 1 Schematic diagram of biochar-immobilized bacteria composites prepared using the adsorption method and embedding method and combining the adsorption and embedding method

附镉前后其表面官能团的变化. FTIR 光谱分析表明: 3 400 cm⁻¹来自—OH的伸缩振动,吸附镉后谱峰发生显著的变化; 1 253、1 313 和 2 927 cm⁻¹代表—CH 官能团的伸缩振动,894 cm⁻¹和 823 cm⁻¹处的谱峰归因于呋喃的γ-CH和吡啶的β环,吸附镉后谱峰显著强度增加或消失; 1 632、1 082 (C=O) 和 1 434 cm⁻¹(C=C) 在吸收镉后也发生了明显的改变; 吸附镉后,—CN 吸附峰由 2 140 cm⁻¹ 处转变为 2 700 cm⁻¹,表明镉与π电子发生了配位. FTIR 光谱分析证明—OH、—CH、C=O、C=C和—CN 的络合作用在镉的吸附过程中起着重要作用.

2.3 BET分析

BET可以测定生物炭固定化菌复合材料的孔径大小、比表面积以及分析孔径分布.任静等^[21]采用BET来测定杉木、秸秆、稻壳和竹炭生物炭的孔容和比表面积,以此来优选固定化多环芳烃高效降解菌的载体.BET分析表明:稻壳生物炭的总孔孔容(0.032 cm³·g⁻¹)和比表面积(51.34 m²·g⁻¹)最大,选择稻壳生物炭作为固定化载体.生物炭的大孔隙有利于体积小且生长快的菌群进入生物炭繁殖,大比表面积可以提供更多的空间供菌群附着,为其繁殖提供栖息地,定殖菌群的效果更好.

2.4 XRD分析

XRD可以表征生物炭固定化菌复合材料的晶体结构.Yin等^[22]通过吸附法和包埋法获得了稻壳生物炭固定化菌(CFI-RHB)和稻壳生物炭固定化菌珠粒(CFI-RHB/SA)并采用XRD表征其结晶结构.XRD图

谱表明: RHB与CFI-RHB在22.1°~22.4°处的2θ衍射峰,与标准石墨碳相对应;海藻酸钠为半晶体结构,CFI-RHB/SA珠粒显示为无定形结构,可能是因为真菌以无定形状态存在于珠粒中而导致其晶体状态发生变化,另外原因是凝胶化过程使海藻酸盐的空间结构发生了改变.

2.5 NMR分析

NMR可以测定生物炭的芳香性和质子化或非质子化碳的含量,从而预测生物炭的稳定性^[23]. Chen 等^[24]利用 NMR 分析了大豆秸秆、木屑和小球藻生物炭的芳香性,其图谱分析表明,生物炭由烷基碳、芳香碳和羰基碳构成,其光谱都以 130 ppm 为中心的芳香族峰为主;生物炭芳香化程度随热解温度升高呈上升趋势,但超过一定温度,生物炭的芳香化程度有所下降.生物炭的芳香化程度越高,其稳定性越强,反之亦然.生物炭的稳定性对生物炭固定化菌复合材料在环境中的寿命至关重要.

3 生物炭固定化菌复合材料在废水处理中的应用

生物炭固定化菌复合材料因具备生物炭的吸附、还原作用又兼具微生物降解污染物的能力而备受关注,可应用于水中氨氮、磷、重金属和有机污染物的去除与降解.

3.1 生物炭固定化菌复合材料对废水中污染物的 去除性能

生物炭固定化菌复合材料已在重金属、有机

物、氨氮和磷等污染废水的处理中得到广泛应用. An等 $[^{25]}$ 通过吸附法制备了花生壳生物炭固定化假单胞菌复合材料应用于电镀废水的处理,其研究发现,复合材料对Cu(II)、Zn(II)、氨氮、Ni(II)和Cr(VI)的去除率分别为94.92%、91.46%、79.90%、

77.02%和34.40%;与生物炭相比,其去除率提高了64.12%、79.32%、73.84%、24.80%和75.49%;与菌株相比,其去除率提高了36.74%、7.98%、12.86%、17.2%和20.23%.邹宇等[26]以花生壳生物炭为载体,通过包埋法将耐镉细菌定殖在生物炭上而制备固定

表 1 生物炭固定化菌复合材料对水中污染物的去除性能

Table 1 Removal performance of biochar-immobilized bacteria composites for pollutants in water

复合材料	生物炭类型	微生物	固定化方法	污染物	处理条件 ¹⁾	去除率或去除量	文献
生物炭负载纳米零 价铁材料固定化溶 磷菌微珠	稻壳生物炭	非脱羧勒 克菌	包埋法	Pb(II)	pH=5、 t =24 h、 c_0 =200 mg·L $^{-1}$ 、 D =5 g·L $^{-1}$ 和 T =30 °C	93%	[17]
生物炭固定化活蜡 样芽孢杆菌 RC-1	稻壳、鸡粪和 污泥生物炭	活蜡样芽 孢杆菌	包埋法	$\operatorname{Cd}({\rm I\hspace{1em}I})$	$\begin{array}{c} {\rm pH=7}\;,\;t{=}30\;{\rm h}\;,\;c_{0}{=}100\;{\rm mg}\cdot{\rm L}^{-1}\;,\\ D{=}5\;{\rm g}\cdot{\rm L}^{-1}{\not \sqcap}\;T{=}30\;{^{\circ}{\rm C}} \end{array}$	158.77 mg·g ⁻¹	[20]
花生壳生物炭固定 化假单胞菌L1	花生壳生物炭	假单胞菌	吸附法	Cu(II) 、Zn(II) 、 NH ₄ ⁺ -N 、Ni(II)和 Cr(VI)	pH=7、 t =120 h、 c_0 为 1.016、 1.708、4.021 和 4.597 mg· \mathbf{L}^{-1} 、 D =1 g· \mathbf{L}^{-1} 和 T =30 °C	94.92%、91.46%、 79.90%、77.02%和 34.40%	[25]
生物炭负载微生物	花 生 売 生 物 炭	耐 Cd 菌 群	包埋法	$\operatorname{Cd}({\rm I\hspace{1em}I})$	pH=6 、 t =7 h 、 c_0 =100 mg · L ⁻¹ 、 D =3 g · L ⁻¹ ₹ Π T =30 °C	96%	[26]
生物炭基硫酸盐还 原菌	稻草、小麦和 玉米秸秆生 物炭	硫酸盐还 原菌	吸附法	Cr(VI)	pH=5、 t =8 h、 c_0 =100 mg・ L^{-1} 、 D =0.6 g・ L^{-1} 和 T =30 °C	286.54 mg·g ⁻¹	[29]
生物炭固定化紫链 霉菌 SBP1	桉 树 叶 生 物 炭	紫链霉菌	吸附法	Mn(II)	pH=7 、 t =72 h 、 c_0 =5 mg · L $^{-1}$ 、 D =2 g · L $^{-1}$ 和 T =30 °C	74.8% 或 1.15 mg·g ⁻¹	[30]
生物炭固定化硫酸 盐还原菌	稻草、小麦和 玉 米 秸 秆 生 物炭	硫酸盐还 原菌	吸附法	Cd(II)	pH=8、 t =8 h、 e_0 =40 mg·L $^{-1}$ 、 D =0.6 g·L $^{-1}$ 和 T =30 °C	77.93 mg•g ⁻¹	[31]
玉米秸秆生物炭-细 菌复合体	玉米秸秆生物 炭	代尔夫特 菌	吸附法	Cd(II)和As(V)	${ m pH}$ =5、 t =8 h、 c_0 =100 mg・ ${ m L}^{-1}$ 、 D =0.6 g・ ${ m L}^{-1}$ 和 T =30 °C	75.38 mg·g ⁻¹ 和 34.03 mg·g ⁻¹	[32]
Triton X-100-生物 炭固定化铜绿假单 胞菌	稻秆生物炭	铜绿假单 胞菌	吸附法	苊	pH 为 4.5 ~ 10.5、 t =72 h、 c_0 为 0 ~ 300 mg·L ⁻¹ 、 D =1 g 和 T 为 0 ~ 30 °C	76% ~ 78%	[27]
生物炭固定化菌	加拿大一枝黄 花茎生物炭	肠杆菌菌 株	吸附法	吡啶	pH=7 、 t =36 h 、 c_0 =200 mg·L ⁻¹ 、 D=2 g·L ⁻¹ 和 T =28 °C	91.7%	[33]
中药渣生物炭固定 化蜡样芽孢杆菌 LZ01	药渣生物炭	蜡样芽孢 杆菌	吸附法	氯四环素	pH=7.84、 t =36h、 c_0 =73.75 mg·L ⁻¹ 、 D =0.51(质量分数)和 T= 33.98 °C	83.83%	[34]
海藻酸钠-生物炭固 定化菌群 GYB1	玉米秸秆生物 炭	多氯联苯降解菌	包埋法	五氯联苯	pH=8 、 t =5 d 、 c_0 =1 mg · L ⁻¹ 、 T= 35 °C和 D (SA)+ D (GYB1)+ D(BC)=2%+2%+1.5%	50.50%	[35]
生物炭固定化生物 表面活性剂生产细 菌 LQ2	玉 米 秸 秆 生 物炭	弧菌	吸附法	柴油	t=7 d、c₀= 1% (体积比)、D=0.1 g和 T=30 ℃	94.7%	[36]
生物炭固定化荧光 假单胞菌 MC46	桉 树 枝 生 物 炭	荧光假单 胞菌	吸附法	三氯二苯脲	pH=7、 t =36 h、 c_0 =10 mg·L ⁻¹ 、 D=1 g 和 T =30 °C	83%	[37]
中药渣生物炭固定 化蜡样芽孢杆菌	中药渣生物炭	蜡样芽孢 杆菌	吸附法	氯四环素	pH=7 、 t =48 h 、 c_0 =50 mg · L ⁻¹ 、 D=1 g · L ⁻¹ 和 T =30 °C	(85.42 ± 0.82)%	[38]
磁性漂浮生物炭凝 胶珠固定化微生物 联合体	浒苔生物炭	复合降解 菌	包埋法	芘、苯并芘和茚并 [1,2,3-cd]芘	pH=8、 t =20d、 c_0 为20、10和 10 mg·L ⁻¹ 、 D =5 mL和 T =30 °C	89.8%、66.9% 和 78.2%	[39]
生物炭基异养硝化 细菌	稻壳生物炭	异养硝化 细菌	吸附法	NH ₄ +-N	t =48 h 、 c_0 =100 mg · L ⁻¹ 、 D=1 g · L ⁻¹ 和 T =30 °C	90.93%	[28]
生物炭固定化鞘氨 单胞菌	玉 米 秸 秆 生 物炭	鞘氨单胞 菌	吸附法	NH ₄ ⁺ -N、NO ₂ ⁻ -N, NO ₃ -N和TP	pH=7、 t =120 h、 c_0 =270 mg·L $^{-1}$ N+130 mg·L $^{-1}$ P、 D =4 g·L $^{-1}$ 和 T =25 °C	63%、38%、25%和 35%	[40]
生物炭/粘土复合颗粒固定化苍白杆菌	芦 苇 秸 秆 生 物炭	苍白杆菌	包埋法	NH ₄ ⁺ -N	pH=7、 t =168 h、 c_0 =50 mg·L ⁻¹ 、 D=1 g·L ⁻¹ 和 T =25 °C	79.39%	[41]

化生物炭小球(IBP)用于吸附水中Cd2+. 当Cd2+初始浓 度为100 mg·L⁻¹、IBP 投加量为3 g·L⁻¹、pH为6、温 度为30℃和吸附平衡时间为7h条件下,IBP对Cd2+ 吸附率为96%;灭活与未灭活的IBP对Cd2+吸附率 分别为95%和98%,证明微生物在Cd2+的吸附中发 挥着作用. Lu等[27]通过吸附法制备了表面活性剂-生物炭固定化铜绿假单胞菌(TFBIP)复合材料应用 于水中苊的去除,其研究表明,24 h处理后, 等[28] 采用吸附法获得了生物炭基固定异养硝化细 菌并研究其对水中氨氮的去除效果, 其研究发现, NaOH和NaOH+Mg(Ⅱ)修饰的稻壳生物炭固定化 异养硝化细菌对 NH4+N 的去除效率分别为 88.66% 和90.93%, 异养硝化细菌以NH4+N为氮源, 硝化 生成 NO₃-N 和 NO₂-N; 二者对 P的吸附量分别为 773.75 nmol·g⁻¹和941.17 nmol·g⁻¹. 表 1 显示了以前研 究报告的生物炭固定化菌复合材料对水中污染物的 去除性能.

3.2 实验参数对生物炭固定化菌复合材料去除水中污染物的影响

实验参数会影响生物炭固定化菌复合材料表面 电荷以及微生物的生长繁殖,选择最优的实验参数 将会增强其去除水中污染物的效果. 污染物初始浓 度影响生物炭的活性位点以及微生物的活性; 溶液 pH影响生物炭表面电荷和活性位点、微生物的生 长和活性以及污染物的化学形态;温度影响生物炭 的孔隙大小和微生物的活性; 投加量影响复合材料 的活性位点等. 朱晓丽等[29]探究了溶液 pH 对秸秆 生物炭固定化硫还原菌去除水中 Cr(VI)的影响. Cr(Ⅵ)的去除随着pH的增加而先增加后减小;当 pH=5时, Cr(VI)去除率达到最大值; 当pH < 7 时, OH-与阴离子Cr(VI)竞争吸附位点能力弱; 当 pH>7时, OH⁻与阴离子 Cr(Ⅵ)竞争吸附位点能力 强,导致Cr(VI)去除率下降;同时,硫还原菌的 生长繁殖受到碱性环境的抑制也会致使 Cr(VI)去 除率下降. 王梓婷[42]探讨了生物炭固定化希瓦氏菌 的添加量对水中Cr(VI)去除的影响.Cr(VI)去除率 随着添加量的增加而增加, 当添加量为4颗·mL⁻¹ 时, Cr(VI) 去除率最高(90.41%); 当添加量从1.2 颗·mL-1增加至2颗·mL-1时, Cr(VI)的去除率增加 了 43.77%; 当添加量从 2 颗·mL⁻¹增加至 4 颗·mL⁻¹ 时,去除率仅增加了10.47%.这是由于添加量低于 2颗·mL⁻¹时, Cr(Ⅵ)浓度高, 复合材料能够发挥最 大的吸附和还原作用; 当添加量大于2颗·mL-1时, 复合材料过量, Cr(VI)去除率接近平衡, 导致 Cr (VI)去除率增加不明显. Nie 等[33]研究了吡啶初始

浓度、溶液pH和反应温度对生物炭固定化吡啶降 解混合菌去除水中吡啶的影响, 其研究发现, 当吡 啶初始浓度从50 mg·L⁻¹增加到500 mg·L⁻¹时,固定 化菌对吡啶的去除呈下降趋势但保持高的效率, 主 要原因为高浓度吡啶对降解菌的毒害作用,但是生 物炭为降解菌提供了稳定环境使固定化菌对吡啶的 去除保持高的效率; 当pH=7时, 固定化菌对吡啶 的去除率最大; 当pH大于7或小于7时, 酸性或碱 性破坏了降解菌的酶分子结构,导致降解菌降解吡 啶的能力下降. 随着温度从 15 ℃升高到 30 ℃时, 固定化菌对吡啶的去除率增加,这与温度升高而降 解菌细胞的代谢加快有关;高于30℃时,吡啶的 去除率呈下降趋势, 这是因为过高温度使降解菌细 胞内的酶失活而导致降解菌代谢能力降低. Sun 等[41]以生物炭/黏土复合颗粒为载体,将苍白杆菌 固定到复合颗粒上,考察温度对氨氮去除的影响. 当温度从 15 ℃升高到 30 ℃时, 固定化菌对 NH4+N 的去除率逐渐增加; 当温度为35℃, NH4+N的去 除率急剧下降.温度过低,影响细胞的酶促反应, 限制细菌的生长;温度过高,致使细胞内蛋白质失 活,降低细菌活性.

综上所述,相比于游离菌+生物炭组合,生物炭固定化菌复合材料在去除水中污染物的性能方面有明显的提高.通过微生物固定化技术将生物炭与菌群相结合构成的生物炭固定化菌复合材料,在废水处理中展现出巨大的应用潜力.同时,水环境较为复杂,优化环境因素对生物炭固定化菌复合材料去除水中污染物的影响是必要的.

4 生物炭固定化菌复合材料在污染土壤修复中的 应用

生物炭固定化菌复合材料作为一种用于固定土壤中重金属和有机污染物的修复剂,可以有效降低土壤中重金属和有机污染物的毒性、生物利用性和迁移性而受到关注^[43].

4.1 生物炭固定化菌复合材料对土壤中污染物的 修复性能

生物炭固定化菌复合材料对修复土壤污染物的研究起步较晚,且主要集中于实验室规模,在实际场地中的应用研究较少.朱晓丽等[44]制备了小麦秸秆生物炭固定化硫酸盐还原菌应用于Cd污染土壤的修复,结果表明,生物炭(XM700)、硫酸盐还原菌(SRB)和生物炭固定化硫酸盐还原菌(IBXM700)对Cd²+的钝化效果分别为: IBXM700 > SRB > XM700;与IBXM700和 SRB 相比,IBXM700显著降低可交换态的Cd 和提高残渣态的Cd. Ji 等[45]利用玉米秸秆

生物炭为载体固定化磷酸盐溶菌应用于土壤中 Pb2+ 的钝化,研究发现,其固化率可达70%,与磷酸 盐溶菌相比约提高了1.5倍,与生物炭相比约提高 了3倍. Chen 等[46]通过吸附法将铬还原菌固定到浒 苔生物炭协同修复铬污染土壤, 其研究发现, 与 生物炭和铬还原菌相比, 生物炭固定化菌对铬的 还原率(94.22%)提高了约11%~23%. 顾玲峰[47]开 展了生物炭固定化芽孢杆菌对 Cr(VI)-芘污染土壤 的修复.结果表明,修复28d,对土壤中芘和 Cr(VI)的去除率分别为82.32%和55.64%.Qi等[48]将 镉、铀耐性混合菌负载到生物炭应用于镉-铀污染 土壤的修复,结果表明:75 d后,3% 复合材料的 施用使得DTPA可提取态的镉和铀分别降低了69% 和 56%. 张秀霞等[49]研究了污泥生物炭固定化菌修 复石油污染土壤,其研究发现,90 d的修复,污 泥生物炭固定化菌组对总石油烃降解率为58.80%, 比污泥生物炭、添加污泥生物炭+游离降解菌对总 石油烃降解率提高了 33.73% 和 13.66%. Xiong 等[50] 开展了稻壳生物炭固定化菌修复多环芳烃污染土 壤的研究,结果显示,生物炭、游离菌和生物炭 固定化菌对荧蒽的去除率分别为0、0和52.1%, 对菲的去除率分别为0、47.3%和62.6%,对芘的 去除率分别为13.5%、19.7%和62.1%,固定化菌 对荧蒽、菲和芘的降解率提高了52.1%、15.3%和 42.5%. Liu 等[51]采用壳聚糖改性生物炭固定化复合 菌剂应用于石油污染土壤的修复. 经过60 d的油污 染土壤修复, 壳聚糖改性生物炭固定化菌对原油 的去除率为45.82%, 比天然修复高了21.26%. 表2 总结了生物炭固定化菌复合材料修复污染土壤的 案例.

4.2 环境因素对生物炭固定化菌复合材料修复污染土壤的影响

土壤 pH、温度、湿度、污染物初始浓度和材料施用量等会对生物炭固定化菌复合材料修复污染土壤的效果产生影响,不仅影响微生物的生长代谢,而且影响生物炭的吸附作用. 牟珍珍[60]利用生物炭固定化镉耐性菌修复镉污染土壤并开展了修复时间、土壤 pH和材料施用量等对修复镉污染土壤的影响研究,结果证明,生物炭固定化菌对镉的去除随着 pH的升高而先增高后降低,最佳pH为7;随着修复时间,生物炭固定化菌对镉去除逐渐升高;随着施用量的增加,镉去除率先上升后趋于稳定. 姜庆宏等[61]研究了土壤湿度对玉米秸秆生物炭固定化铬还原菌修复 Cr(VI)污染土壤的影响,结果表明,当土壤湿度为 14% 时,Cr(VI)去除最大;当土壤湿度为 23% 时,Cr(VI)去除最大;当土壤湿度为 23% 时,Cr(VI)

去除最小;这是因为土壤含水率过低,抑制微生物的代谢生长,影响 Cr(VI)的去除;含水量过高,限制微生物的好氧呼吸,降低了 Cr(VI)的去除.Yin等[22]研究了柴油初始含量对稻壳生物炭和海藻酸钠固定化复合真菌修复柴油污染土壤的影响.随着柴油初始含量从 5 000 mg·kg⁻¹增加到 20 000 mg·kg⁻¹,游离的复合真菌和固定化的复合真菌对柴油降解率呈下降趋势,这是因为高浓度柴油对微生物的毒害作用以及土壤成团导致细胞窒息死亡,致使柴油的去除率下降.相比游离复合真菌,固定化的复合真菌对柴油降解率下降趋势缓慢,这与生物炭和海藻酸盐为微生物提供了必需的营养物质和稳定的环境有关.

4.3 生物炭固定化菌复合材料对植物生长和土壤 理化性质的影响

生物炭固定化菌复合材料对污染土壤修复的同 时还会影响土壤酶活性、pH、微生物活性、营养 元素、有机质、阳离子交换量和植物生长等的影响 (图 2). Song 等[53] 利用生物炭固定化芽孢杆菌 (PBM4)修复多环芳烃和重金属污染土壤,结果发 现,PBM4处理可有效提高猪毛蒿生物量,降低多 环芳烃和重金属在其内的积累, 明显提高了能去除 多环芳烃和重金属的微生物丰度. Zhang 等[54]研究 了磷酸球磨生物炭固定化耐镉溶磷菌的应用对植物 生长和镉形态变化的影响,结果表明,复合材料的 施用显著提高了小白菜生物量和磷含量,降低了镉 在小白菜叶和根的积累量,降低了25.90%~ 43.46%. Wang 等[55]发现磁性生物炭固定化微生物复 合材料的应用明显提高了土壤的pH、降低了可利 用 Cd 和提高了微生物数量(700%). 胡松伯[62]探究 了铁改性生物炭负载节杆菌(bFeMBC)降解阿特拉 津过程中对土壤微生物群落的影响,结果发现: bFeMBC施用有效去除污染物的同时增加了土壤微 生物的多样性和丰度. 李琋等[63]开展了生物炭负载 微生物对镉-石油烃污染土壤修复的研究并探讨了 其对土壤微生物、酶活性和pH的影响,结果表明, 固定化微生物组的土壤 pH 轻微下降,显著提高了 土壤多酚氧化酶活性、过氧化氢酶活性、脱氢酶活 性和微生物数量.

综上所述,生物炭固定化菌复合材料克服了游 离菌生长缓慢和对土壤变化敏感等缺点,富集污染 物的同时保持菌群的高活性,对污染土壤具有良好 的修复作用,有效改良土壤环境,促进植物生长. 因此,生物炭固定化菌复合材料是一种低成本和高 效的污染土壤的修复剂,可用于污染场地的大规模 修复.

表 2 生物炭固定化微生物复合材料对污染土壤的修复性能

Table 2 Remediation performance of biochar-immobilized bacteria composites for contaminated soil

复合材料	生物炭	微生物	固定化 方法	污染物	修复条件1)	修复效果	文献
生物炭固定 化蜡样芽孢 杆菌WHX-1	浒苔生 物炭	蜡样芽孢杆 菌	吸附法	Cr(VI)	t =28 d 、 D =20 g 、 c_0 =50 mg·kg ⁻¹ 和 T =28°C	94.22%的 Cr(Ⅵ)转为 Cr(Ⅲ)	[46]
混合细菌负载的生物炭	玉米秸 秆生物 炭	枯草芽孢杆 菌、蜡样芽 胞杆菌和柠 檬酸杆菌	吸附-包 埋法	U和 Cd	pH=6.9、t=75 d、D=3%、c ₀ 为 28.9 mg·kg ⁻¹ 、2.5 mg·kg ⁻¹ 和 T=25℃	U和Cd的固化率分别69%和56%	[48]
生物炭固定化菌	菠萝皮 生物炭	芽孢杆菌 W1、W2和 Y2	吸附-包 埋法	芘和 Cr(VI)	t=28 d √D=2.5 g · kg ⁻¹ √c ₀ 为 42.33 mg · kg ⁻¹ √6.95 mg · kg ⁻¹ 和 T=30 °C	芘和 Cr(VI)浓度从 42.33 mg·kg ⁻¹ 和 6.95 mg·kg ⁻¹ 减为 7.48 mg·kg ⁻¹ 和 2.58 mg·kg ⁻¹	[52]
生物炭固定 化芽孢杆菌 KSB7	花生壳 生物炭	芽孢杆菌	吸附法	多环芳香烃、 Zn(Ⅱ)、Pb (Ⅱ)、Cr(Ⅵ)和 Cu(Ⅱ)	t=90 d、D=200 mL(悬浮液)、c ₀ 为50.3、241、217、106和94 mg·kg ⁻¹ 和 T=28℃	去除率94.17%、58.46%、53.42%、84.94%和83.15%	[53]
磷酸球磨改性生物炭固定化抗镉溶磷菌(MPBC)	玉米秸 秆生物 炭	抗镉溶磷菌	吸附法	Cd(II)	<i>t</i> =30 d 、 <i>D</i> =1%(质量比)、 <i>c</i> ₀ = 43.2 mg·kg ⁻¹ 和 <i>T</i> =26 ℃或 20 ℃	降低土壤中 20.12% Cd含量和叶 片中 25.90% Cd 积累量	[54]
磁性生物炭- 微生物复合 材料	稻壳生 物炭	芽 孢 杆 菌 K1	包埋法	Cd(II)	<i>t</i> =30 d 、 <i>D</i> =1%(质量比)、 <i>c</i> ₀ = 43.2 mg·kg ⁻¹ 和 <i>T</i> =26 ℃或 20 ℃	提高了土壤的 pH、降低了可利用 Cd和提高了微生物的数量	[55]
茶叶生物炭 固定化雷尔 氏菌		雷尔氏菌	吸附法	Cr(VI)和 Cd(Ⅱ)	pH= 4.96 、t=30 d 、D=40 g · kg ⁻¹ 、c ₀ 为 16.93 mg · kg ⁻¹ 、0.18 mg · kg ⁻¹ 、 T=25 °C和 M 为 60% ~ 80%	促进了菌群生长、改变无机氮形态、增加 NO ₃ -N供应、降低 Cd 和 Cr生物有效性和降低植物组织 Cd 和 Cr含量	[56]
生物 炭 固 定 化混合菌	玉米秸 秆生物 炭	猪粪肠杆菌 G3、烟粉肠 杆菌 I12 和 水痘克雷伯 菌 J2	吸附-包 埋法	₽b(Ⅱ) 和 Cd(Ⅱ)	t=45 d、D=3 %、c ₀ 为492 mg·kg ⁻¹ 、 2.1 mg·kg ⁻¹ 、T=25 °C和 M=70%	DTPA可提取 Cd 和 Pb 含量降低了 22.05% ~ 55.84% 和 31.64% ~ 48,13%、小白菜茎部 Cd 和 Pb 降低了 28.68% ~ 51.01% 和 24.18% ~ 52.87%	[57]
生物炭固定 化黄分枝杆 菌	稻壳生 物炭	分枝杆菌	吸附法	菲、荧蒽和芘	t =18 d、 D =0.25 g、 c_0 = 677 mg·kg $^{-1}$ 和 T =30 °C	去除率分别为(62.6 ± 3.2)%、(52.1 ± 2.3)%和 (62.1 ± 0.9)%	[50]
壳聚糖-生物 炭固定化微 生物	甘蔗渣 生物炭	铜绿假单胞 菌和地衣芽 孢杆菌	吸附法	原油	pH=7、 t =60 d、 D =2 g、 c_0 = 8 000 mg·kg ⁻¹ 、 T =35 °C和 M = 20%	去除率为45.82%	[51]
生物炭固定 化适度嗜盐 菌	棉秆生 物炭	地中海马特尔氏菌 AD-3	吸附法	菲	pH=8.35 、 t =40 d 、 D =1%、 c_0 = 52.29 mg·kg ⁻¹ 、 T =35 °C和 M = 20%	去除率为91.67%	[58]
生物炭固定 化鞘氨单胞 菌PJ2	松针生 物炭	鞘氨单胞菌 PJ2	吸附法	PAHs	pH=7.59 、 t =60 d 、 D =50 mL、 c_0 = 1 939.9 mg · kg $^{-1}$ 、 T =28 °C和 M = 50%	去除率为 58.64%	[59]
生物炭固定化菌微球	铁改性 稻壳生 物炭	环芳香族碳 氢化合物	包埋法	華	$t=6 \text{ d}_{\circ}D=2 \text{ g}_{\circ} c_0=41.2 \text{ mg} \cdot \text{kg}^{-1}_{\circ}$ 、 $T=25 ^{\circ}$ C和 $M=60\%$	去除率为30%	[19]
稻壳生物炭 固定化真菌 复合材料	稻壳生 物炭	柴油降解微 生物	吸附-包 埋法	柴油	t =60 d、 D =4%(质量比)、 c_0 = 10 000 mg・kg $^{-1}$ 、 T =25 $^{\circ}$ C和 M = 25%	去除率为64.10%	[22]

1)t: 修复时间; T: 反应温度; D: 复合材料投加量; c_0 : 污染物初始浓度; M: 土壤湿度

5 生物炭固定化菌复合材料对环境中污染物的去除机制

生物炭固定化菌复合材料对环境中污染物的去

除机制主要涉及生物炭的吸附、还原以及菌群的生物降解(图3)^[64].

生物炭固定化菌复合材料对环境中重金属、氨氮和磷等无机物的去除机制主要涉及^[65]: ①生物炭的孔

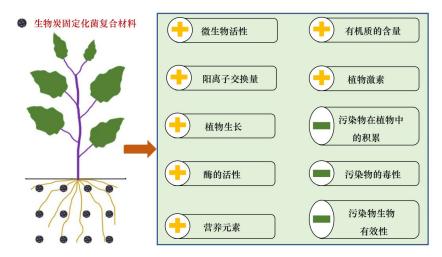


图 2 生物炭固定化菌复合材料对植物生长和土壤理化性质的有利影响

Fig. 2 Beneficial effects of biochar-immobilized bacillus on plant growth and physicochemical properties of soil

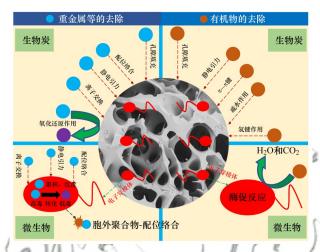


图 3 生物炭固定化菌复合材料对环境中污染物的去除机制 Fig. 3 Mechanism of biochar-immobilized bacteria composites for the removal of pollutants in environment

隙填充、静电吸引、离子交换和配位络合等作用来吸附重金属等无机物;②菌群细胞表面官能团的静电吸引、配位络合和离子交换等吸附重金属等无机物;生物炭介导下菌群对重金属的氧化还原转化、胞内积累、沉淀去除以及胞外聚合物的络合固定.

生物炭固定化菌复合材料对环境中有机物的去除机制主要包括^[66]:①生物炭的静电引力、π一π键、疏水作用、氢键和孔隙填充等吸附有机污染物;②生物炭介导下菌群将有机污染物作为自身生长代谢的碳源,通过酶促反应将有机污染物降解为CO₂和H₂O等.

6 展望

目前,生物炭固定化菌复合材料在环境修复中 大规模应用仍存在局限性,许多问题需要进一步 研究:

(1)针对固定化方法是微生物固定化技术的核

- 心,开发新型固定化法或采用联合固定化法是提高 微生物固定化量的有效方法之一.
- (2)针对复合污染物和新兴污染物污染土壤, 为提高生物炭固定化菌复合材料修复复合污染物和 新兴污染物污染土壤效果,优选高活性和易培养的 混合菌和特异菌是必要的.
- (3)研发生物炭的改性技术来增加生物炭官能 团数量、改变生物炭表面电荷以及生物炭比表面 积,从而提高生物炭对微生物负载量.

7 结论

通过微生物固定化技术制备的生物炭固定化菌复合材料具有生物炭与微生物的双重修复作用,在处理废水及修复污染土壤领域展现出广阔应用前景.本文详细论述了生物炭固定化菌复合材料的制备方法和表征技术;评价了其治理污水和修复污染土壤的效能、影响其修复效果的因素及其应用对植物生长和土壤理化性质的影响;阐明了其修复污染物的机制.以期为生物炭固定化菌复合材料在环境污染治理中的广泛应用提供理论和实践参考.

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