

◆特邀专稿◆

市政污泥好氧发酵过程难溶性磷酸盐转化研究进展^{*}

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摘要:从污水厂的污泥中回收磷是未来国内外市政污泥资源化的必然趋势,也是解决磷矿资源短缺的一个重要措施,已成为有机固废资源化领域的热点问题。市政污水处理厂污泥中的磷含量为7.10~27.60 g/kg,我国市政污泥产量巨大,预计2025年污泥年产量将突破9 000万t(以80%含水率计),是极具潜力的磷源。现有两种从市政污泥中回收磷的途径:从市政污泥中直接回收磷和在市政污泥处理处置过程中利用磷,而好氧发酵-土地利用是市政污泥处理处置的主流工艺之一。该工艺可将市政污泥中难溶性磷酸盐转化为可被植物吸收利用的有效磷,是实现市政污泥磷资源回收利用的有效途径。本文综述了国内外市政污泥好氧发酵过程难溶性磷酸盐转化为有效磷的研究现状,重点介绍了有机质和微生物对难溶性磷酸盐转化影响的研究进展,总结了市政污泥好氧发酵过程中难溶性磷酸盐转化所存在的问题并提出解决思路,同时对以市政污泥好氧发酵为基础的难溶性磷酸盐转化研究进行了展望,以期为市政污泥中的磷资源利用研究提供一些思考。

关键词:市政污泥 好氧发酵 难溶性磷酸盐 磷回收 调理剂

中图分类号:X705 文献标识码:A 文章编号:1002-7378(2022)03-0213-09

DOI:10.13657/j.cnki.gxkxyxb.20221019.001

全球正面临磷资源短缺危机,国际上愈来愈重视对磷资源的保护与回收利用。由于市政污水处理的除磷工艺可将污水中90%以上的磷转移到剩余污泥中^[1],我国市政污泥中的磷含量可达7.10~27.60 g/kg^[2]。截至2019年底,我国每年由污水处理产生的污泥(以含水率80%计)高达6 000万t,预计到2025年其年产量将突破9 000万t^[3]。市政污泥中的磷主要以难溶性磷酸盐的形式存在,若将其转化为能被直

接利用的磷形态,则可通过作物吸收进入磷循环。这个过程可以替代一部分农业磷肥,将极大缓解全球磷资源危机^[4]。因此,从市政污泥中回收磷已成为研究热点并在一些国家上升为国家战略^[5]。

目前已有不少国家颁布法律法规要求从市政污泥中回收磷。瑞士是欧洲第一个强制从污泥中回收磷的国家,并于2016年开始实施政策,现已建立起磷元素封闭循环的管理体系,所回收的市政污泥中磷资

收稿日期:2022-07-21 修回日期:2022-08-15

* 广西重点研发计划(桂科 AB19110001),南宁市优秀青年科技创新创业人才培育项目(RC20200108)和南宁市科学研究与技术开发计划(20183045-2)资助。

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胡湛波,苏钰婷,陈秋羽.市政污泥好氧发酵过程难溶性磷酸盐转化研究进展[J].广西科学院学报,2022,38(3):213-221.

HU Z B, SU Y T, CHEN Q Y. Research Progress on the Transformation of Insoluble Phosphate During Aerobic Fermentation of Municipal Sludge [J]. Journal of Guangxi Academy of Sciences, 2022, 38(3): 213-221.

源可以完全替代原来进口的磷矿石^[5]。德国于2017年通过了对《污水污泥条例》的修订,新条例明确要求对市政污泥需进行磷回收处理^[6]。欧盟2019年最新出台的《肥料产品法规》更是为磷回收产品自由进入市场流通打破了贸易壁垒^[7,8]。我国对污水厂的磷回收还没有出台相关政策与法律法规,但国内已有市政污泥磷回收的应用实例^[9]。国际上普遍希望通过市政污泥中的磷利用缓解磷资源危机,从市政污水厂的污泥中回收磷展现出良好的发展前景。

国内外从市政污泥中回收磷的途径主要分为两种^[1,6]:一是从市政污泥中直接回收磷,主要技术包括湿式化学法、热化学法;二是在市政污泥处理处置的过程中实现磷资源利用,主要为土地利用法。表1简要地对比了主要的市政污泥磷回收技术,其中湿式化学法的代表工艺是德国的AirPrex工艺,该工艺使含水率高的市政污泥形成鸟粪石结晶从而实现磷回收,但该方法需要外加化学试剂从而导致较高的投入成本^[10,11]。热化学法回收磷则以奥地利的AshDec

工艺为代表,即市政污泥经过焚烧、共热解等工艺形成含磷灰烬,再进一步从中回收磷^[12]。上海市3座大型污水处理厂从污泥(焚烧)飞灰中回收磷,预估全年可回收4万t的P₂O₅,但由于经济成本高,目前该技术仍处于实验阶段^[9]。尽管湿式化学法和热化学法具有磷回收率高的优势^[13],但是这两种技术存在腐蚀性强、成本高、能耗大等缺陷^[14],短期内难以在我国大力推行。与前述两种技术相比,基于好氧发酵-土地利用的磷回收途径具有操作简便、应用范围广和成本低等优势^[15,16]。好氧发酵-土地利用是我国未来市政污泥处理处置的主流路线之一^[3],市政污泥经过好氧发酵后施用在土壤中,污泥中丰富的氮、磷、钾等营养物质可以被植物直接利用^[17]。根据E20数据统计显示,好氧发酵技术在国内污泥处理领域中的占比为18%^[18],结合我国国情,好氧发酵-土地利用途径将是实现市政污泥磷资源利用的重要手段之一。

尽管通过好氧发酵技术可实现市政污泥磷资源化利用,但是现阶段在应用过程中仍存在难溶性磷酸

表1 市政污泥磷回收技术对比

Table 1 Comparison of municipal sludge phosphorus recovery technologies

磷回收利用方法 Method of phosphorus recovery and utilization	主要技术 Primary technology	技术特点 Characteristics of technology	文献来源 Resources of literature
Wet chemical method	Struvite crystallization	Municipal sludge is pretreated with acid/alkali solution and recovered by struvite crystallization, but it is highly corrosive and phosphorus can not be fully recovered	[19,20]
	Vivianite crystallization	In the anaerobic environment, iron salt is used as the coagulation agent, and it is recycled by vivianite crystallization. The recovery rate is high, but the operation is complicated	[21]
Thermochemical method	Incineration process	Almost all phosphorus can be enriched into ash by municipal sludge incineration. The recovery of phosphorus from incineration ash is relatively simple and efficient, but the energy consumption is high	[22]
	Pyrolytic process	It can reduce sludge volume and convert municipal sludge into biochar, and mainly retain phosphorus in solid products, but it has high energy consumption and low yield	[23]
	Hydrothermal carbonization	The phosphate in municipal sludge is released and enriched in hydrothermal char, but the product has low stability	[24]
Land use method	Aerobic fermentation technology	It can treat and disposal municipal sludge, and realize phosphorus resource utilization. The nutrient content is high and the energy consumption is low, but the availability of phosphorus is low	[25]
	Anaerobic fermentation technology	The organic phosphorus in municipal sludge is released by hydrolysis of anaerobic fermentation, which has the advantages of low cost and low energy consumption. However, hazards such as pollutants and heavy metals in sludge limit its application	[26]

盐转化效率低、磷利用率低等问题^[27]。目前,关于好氧发酵过程中磷资源的研究仍以总磷含量变化为主,对磷形态转化及其环境效应关注较少^[28,29]。因此,本文对市政污泥好氧发酵过程中难溶性磷酸盐转化的研究现状进行综述,总结归纳国内外市政污泥好氧发酵过程难溶性磷酸盐转化的关键问题,阐述了目前市政污泥好氧发酵中难溶性磷酸盐转化的技术策略,以期为实现市政污泥好氧发酵产品中磷资源的高效利用提供有效指导,并为我国市政污泥中磷资源利用提出建设性建议。

1 难溶性磷酸盐转化为有效磷的重要性

在市政污水中磷以溶解态或颗粒的形式存在。针对不同形态的磷所采用的处理方式不同,目前污水除磷的工艺主要为化学沉淀除磷和生物除磷^[30,31],最终将污水中90%以上的磷以难溶性磷酸盐的形式转移到剩余污泥中^[1]。市政污泥中的难溶性磷酸盐主要包括 $\text{Ca}_5(\text{PO}_4)_3\text{OH}$ 、 $\text{Ca}_3(\text{PO}_4)_2$ 、 AlPO_4 、 FePO_4 等短期内不能被作物直接利用的含磷化合物,因此富含磷资源的市政污泥中可被植物利用的磷还没有完全被开发^[32],这些物质经过处理可转化为能被植物利用的有效磷从而进入磷循环^[33]。例如溶解性有机质与难溶性磷酸盐组分中的金属离子(Ca^{2+} 、 Al^{3+} 、 Fe^{3+} 等)的螯合或络合作用使难溶性磷酸盐溶解,或难溶性磷酸盐在酸性条件下溶解释放出有效磷(H_2PO_4^- 或 HPO_4^{2-})^[34]。为提高市政污泥中的磷转化效率和利用率,国内外学者进行了相关研究。这些研究结果显示,改变好氧发酵过程的环境因素会对磷转化效率产生影响,例如温度的升高会促进磷的转化,因此越来越多的研究趋于采用更高堆体温度的发酵工艺如超高温好氧发酵工艺等来提高磷的利用率^[35];C/N能显著改变好氧发酵体系中磷转化相关微生物的潜力,当好氧发酵体系中C/N/P变化为N/P<15和C/P<200时,可提高磷的转化效率和增加磷的有效性^[36];而通过调节好氧发酵过程的曝气量发现,曝气量大的实验组污泥磷含量增加了26%^[37]。好氧发酵技术在能实现市政污泥减量化、无害化和稳定化的同时,发酵过程中的有机质矿化可以使物料中的有机磷降解从而释放出大量无机有效磷^[38-40],促进难溶性磷酸盐向有效磷转化^[10]。如Wei等^[41]发现好氧发酵时会导致非速效磷(NAP)的减少,微生物能够产生更多的不稳定态速效磷(LAP)。Wan等^[42]证明在发酵过程中,有机磷(Po)

可水解为无机磷(Pi)和微生物生物量磷(MBP)。因此,难溶性磷酸盐在好氧发酵过程中转化为有效磷,是实现市政污泥处理处置过程磷资源化利用的关键步骤。但市政污泥好氧发酵过程中难溶性磷酸盐转化机制尚不明确,缺乏相关研究,目前国内外众多学者主要围绕其转化途径进行研究。

2 市政污泥好氧发酵过程中难溶性磷酸盐转化的路径

在好氧发酵过程中难溶性磷酸盐向有效磷转化的路径分为两种:一是在好氧发酵过程中有机质分解产生的多元酸类物质与腐熟过程中形成的大分子腐殖质类物质,这两类物质对难溶性磷酸盐具有较强的溶解能力与络合能力^[43];二是利用好氧发酵过程中微生物的溶磷作用,促使市政污泥中的难溶性磷转化为有效磷^[44]。

2.1 有机质是难溶性磷酸盐转化的主要动力

市政污泥好氧发酵过程中,有机质会转化生成小分子有机酸并在腐熟过程中形成腐殖质类物质^[45],这两类物质具有较强的络合能力和活化作用,是促进难溶性磷酸盐转化为可被植物直接利用的有效磷的主要动力^[46]。

有机质转化分解出来的低分子有机酸主要有草酸、柠檬酸、酒石酸、苹果酸和乙酸等^[47],不同种类小分子有机酸的释磷效果因酸浓度的高低存在较大差异^[48]。陆文龙等^[49]研究发现低分子量有机酸如柠檬酸、草酸、酒石酸和苹果酸能通过溶解、螯合等作用明显促进Ca-P、Fe-P和Al-P中磷的释放,同时还发现磷酸盐的活化与有机酸种类、浓度和类型密切相关,其中对难溶性磷酸盐活化能力为柠檬酸>草酸>酒石酸>苹果酸。周芸等^[39]研究表明有机酸类物质对难溶性磷酸盐具有较强的溶解能力,提高发酵体系总酸度可以增强好氧发酵过程中难溶性磷酸盐的溶解效果。另外,腐殖化过程产生了大量能够与难溶性磷酸盐螯合的腐殖质,从而促进磷素从难溶性磷酸盐中释放。Bangar等^[50]发现有机质分解而成的腐殖质物质可通过螯合作用增强难溶性磷酸盐的溶磷效果,表现出腐殖质对难溶性磷酸盐转化的重要影响。Satisha等^[47]在好氧发酵实验中发现发酵体系中产生的腐殖酸不仅可以降低体系pH值,而且还可通过络合作用与金属离子结合,促进难溶性磷酸盐的溶解,进而促进难溶性磷酸盐向有效磷转化。有研究发现,腐殖酸与难溶性磷酸盐通过金属离子(Fe^{3+} 、

Al^{3+})搭桥形成三元复合体来促进磷的释放^[51,52]。

根据上述报道,研究者普遍认为有机质对促进难溶性磷酸盐转化为有效磷起重要作用,而有机质类物质繁多^[53],深入探究促进难溶性磷酸盐转化的关键有机质类物质,对提高难溶性磷酸盐转化效率更具可行性。

2.2 微生物是难溶性磷酸盐转化的主要贡献者

在市政污泥好氧发酵过程中,以解磷菌为代表的微生物可将难溶性磷酸盐转化为能被植物直接利用的有效磷^[54]。难溶性磷酸盐转化的过程相当复杂,而且解磷菌种类繁多,但总体而言,从解磷路径的角度来揭示市政污泥难溶性磷酸盐的转化机理更为准确。因此,解磷微生物的解磷机理研究可分为无机解磷机理和有机解磷机理。

无机解磷菌的主要作用是通过产生酸性物质溶解无机磷化合物^[55],如变形杆菌门(Proteobacteria)细菌在丙酸盐、丁酸盐、葡萄糖和其他小分子的分解以及磷的溶解中起着关键作用^[56]。厚壁菌门(Firmicutes)微生物在堆肥过程中会水解多糖或降解纤维素,能够同时消耗营养物质和增加难溶性磷的溶解^[57]。而细菌属中的假单胞菌(*Pseudomonas*)、芽孢杆菌(*Bacillus*)、根瘤菌(*Rhizobium*)、肠杆菌(*Enterobacter*)等菌株具有强大的磷溶解能力^[58-61]。现有研究报道,市政污泥好氧发酵过程中无机解磷菌的解磷作用主要按以下4种方式进行:(1)解磷菌利用市政污泥中的有机质进行生命代谢活动,分泌出的低分子量有机酸会使市政污泥中的难溶性无机磷溶解^[62-64];(2)通过呼吸作用释放 CO_2 ,降低市政污泥好氧发酵体系的pH值,在酸性条件下市政污泥中多数难溶性磷酸盐都能被溶解^[65];(3)通过 NH_4^+ 的同化作用释放 H^+ ,从而使市政污泥好氧发酵体系形成有利于难溶性磷酸盐转化的酸性环境^[66,67];(4)部分解磷菌能够释放 H_2S , H_2S 通过与磷酸铁作用来促进磷的释放^[68]。

目前已分离出多种具有降解有机磷化合物能力的微生物,如黄杆菌属(*Flavobacterium*)、芽孢杆菌属(*Bacillus*)、产碱杆菌属(*Alcaligene*)、假单胞菌属(*Pseudomonas*)和巨大芽孢杆菌(*Bacillus megaterium*)等,这些菌属的细菌可通过协同代谢和生物矿化的方式参与有机磷化合物的降解^[69]。微生物对有机磷化合物的降解主要是通过有机磷水解酶或磷酸三酯酶等对难溶性有机磷化合物进行水解^[70]。微生物在生长过程中会产生一些可以分泌到细胞外的酶

类,如磷酸酶、植酸酶、核酸酶以及脱氢酶^[71,72]。现有研究发现,与解磷微生物相关的酶在难溶性磷酸盐转化过程中起着特定作用:磷酸酶通过断开磷酸酯键释放磷酸根离子^[73],植酸酶则是将磷从植酸中释放出来^[74],等等。潘贺贺^[75]发现好氧发酵过程中磷酸酶与总磷、有机磷间存在显著的相关性;另外,Wan等^[42]还发现碱性磷酸酶等相关酶有助于好氧发酵过程中难溶性磷酸盐转化,市政污泥经发酵后可被植物利用的磷含量增加121%。

目前研究显示,解磷菌对难溶性磷酸盐转化具有直接性和专一性^[76-78],是市政污泥好氧发酵过程中推动难溶性磷酸盐转化的主要贡献者。但解磷菌的作用机制复杂且易受环境影响,其在好氧发酵过程中的活性难以控制,因此提高难溶性磷酸盐转化效率的有效手段仍然缺乏。

3 市政污泥好氧发酵过程中难溶性磷酸盐转化存在的问题及解决思路

3.1 市政污泥好氧发酵过程中难溶性磷酸盐转化存在的问题

虽然好氧发酵技术是市政污泥处理处置的主流工艺之一,但是好氧发酵过程中对磷资源的实际利用仍未开展。目前的研究主要存在以下3个问题:(1)我国市政污泥中有机质含量相对较低^[79],导致难溶性磷酸盐转化率不高——我国市政污泥中有机质含量较低且多为难降解的大分子有机物、菌体及有毒物质等^[80-82],而有机质作为难溶性磷酸盐转化的主要驱动力,在很大程度上限制了市政污泥好氧发酵中难溶性磷酸盐的转化;(2)市政污泥好氧发酵过程中难溶性磷酸盐的主要转化机制尚不明确——难溶性磷酸盐转化过程复杂,明晰主要转化机制和关键因素是提高难溶性磷酸盐转化效率的前提;(3)对促进好氧发酵过程中难溶性磷酸盐转化的专有微生物菌群的相关研究不足,其适宜菌群配伍和最佳含量等目前仍不清楚^[83,84]。

3.2 促进市政污泥好氧发酵过程中难溶性磷酸盐转化的对策

市政污泥好氧发酵是一个复杂的生化过程^[85],难溶性磷酸盐转化在该过程中实现,转化效率与发酵环境密不可分。发酵过程中工艺参数的变化会对有机质转化产生不同效果进而影响磷转化^[86,87],因此探索最佳工艺条件成为提高难溶性磷酸盐转化效率的对策之一。未来应围绕市政污泥好氧发酵过程的

工艺参数与难溶性磷酸盐转化之间的关系开展更全面的研究,建立好氧发酵过程中难溶性磷酸盐转化的数学模型,并开发相应的数值模拟工具,用于预测最佳工艺条件,进一步提高转化效率。此外,在市政污泥好氧发酵体系中难溶性磷酸盐转化的过程错综复杂,通过研发智能化发酵装置,并根据实际运行情况对工艺参数进行实时调整,有利于维持难溶性磷酸盐高效转化的最佳工艺条件。

添加调理剂是解决难溶性磷酸盐转化问题的另一种思路。调理剂可以从以下两种功效进行选择:直接作用于难溶性磷酸盐和改善发酵条件间接促进难溶性磷酸盐转化。由于生物调理剂直接影响难溶性磷酸盐的转化,因此在市政污泥好氧发酵中添加富含解磷微生物的菌剂,可突破市政污泥中相关解磷微生物丰度低的局限性^[88]。在好氧发酵期间,丰富的解磷微生物直接将更多难溶性磷酸盐转化为有效磷,提高了转化效果。据报道,接种解磷菌剂后污泥中磷的转化效率是未接种菌剂处理的3.75倍^[27];Zhang等^[89]对生物调理剂进行优化,从好氧发酵产物中筛选出耐高温解磷菌并接种至初始发酵物料,发现添加菌剂的发酵体系中可被植物利用的磷含量增加了14%。因此,未来可通过筛选获得高效菌株,进一步制备为高效解磷菌剂,从而提高好氧发酵中难溶性磷酸盐转化效率。另一方面,有机质是影响难溶性磷酸盐转化的主要动力,但我国市政污泥有机质含量相对较低,通过添加富含碳素的调理剂是一条可行的思路。加入生物炭可改善堆体环境,且不同的生物炭对难溶性磷酸盐的转化效果存在差异^[41]。康军等^[90]将市政污泥与玉米秸秆以2:1的比例混合进行好氧发酵,产物中的速效磷含量增加了22.2%。因此,选择可增加好氧发酵体系中有机质含量的调理剂,也是弥补市政污泥有机质含量不足、促进难溶性磷酸盐转化的有效策略之一。

4 展望

目前国内外对市政污泥好氧发酵过程中磷的资源化利用研究仍处于起步阶段,针对该过程中难溶性磷酸盐转化效率不高、磷利用率低等问题,结合未来产业化需求,笔者认为下一步的研究方向如下。

(1)建议国家有关部门开展市政污泥磷回收的相关标准及政策研究,构建市政污泥磷回收体系。目前欧盟一些国家率先出台了污泥磷回收的相关政策与法律法规,磷回收产品能够自由进入市场流通,我国

应积极探索市政污泥好氧发酵磷回收的应用模式,加强宣传导向,鼓励科研机构和企业开展市政污泥好氧发酵磷资源利用的相应研究和应用示范。

(2)优化市政污泥好氧发酵工艺及设备,在市政污泥稳定化、减量化和无害化处理的同时,实现难溶性磷酸盐高效转化,提升好氧发酵产物磷资源利用率。

(3)研发促进难溶性磷酸盐转化的高性能调理剂。比如筛选耐高温的解磷微生物制成高效解磷微生物调理剂,实现好氧发酵过程中难溶性磷酸盐的高效转化;选择有机质含量丰富的易生物降解碳源作为调理剂,提高发酵体系初始有机质含量,促进市政污泥中难溶性磷酸盐的有效转化。

致谢:

感谢林幻云、周倩、张许波、李佩仪在调研过程给予协助。

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Research Progress on the Transformation of Insoluble Phosphate During Aerobic Fermentation of Municipal Sludge

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Abstract: The recovery of phosphorus from sewage sludge is an inevitable trend of municipal sludge resource utilization at home and abroad in the future. It also an important measure to solve the shortage of phosphate rock resources, which has become a hot issue in the field of organic solid waste recycling. The phosphorus content in the sludge of municipal sewage treatment plants is 7.10–27.60 g/kg. The output of municipal sludge in China is huge. It is estimated that the annual output of sludge will exceed 90 million tons (calculated by 80% moisture content) in 2025, which is a potential phosphorus source. There are two ways to recover phosphorus from municipal sludge: Directly recovering phosphorus from municipal sludge and utilizing phosphorus in municipal sludge treatment and disposal. At present, aerobic fermentation-land application use is one of the mainstream technologies for municipal sludge treatment and disposal. This technology can convert insoluble phosphate in municipal sludge into available phosphorus, which is an effective way to realize the recycling of phosphorus resources in municipal sludge. In this article, the research status of the conversion of insoluble phosphates into available phosphorus in the aerobic fermentation process of municipal sludge at home and abroad is reviewed. The research progress of the effects of organic matter and microorganisms on the conversion of insoluble phosphates is mainly introduced. The problems existing in the conversion of insoluble phosphates in the aerobic fermentation process of municipal sludge are summarized and the solutions are proposed. At the same time, the research on the conversion of insoluble phosphates based on aerobic fermentation of municipal sludge is prospected, in order to provide some thoughts for the utilization of phosphorus resources in municipal sludge.

Key words: municipal sludge;aerobic fermentation;insoluble phosphate;phosphorus recovery;conditioner

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