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畜禽粪污厌氧消化产甲烷效能的关键影响因素 研究进展

孙广东 1,2 马俊怡 1,2 郝 书 1,2 董 $g^{1,2}$ 赵心茹 1,2 柴强龙 1,2 党 岩 1,2*

(1. 北京林业大学 水体污染源控制技术北京市重点实验室,北京 100083; 2. 北京林业大学 污染水体源控与生态修复 技术北京高校工程研究中心,北京 100083)

摘 要:在畜禽养殖方式逐渐规模化的背景下,规模化养殖所产生的粪污具有集中排放量大、处置需求强的特点,这为利用厌氧消化技术处置畜禽粪污来实现沼气产量、产能效益最大化提供了契机,完全契合我国能源结构优化、碳中和等战略的需要。然而,由于畜禽粪污组分复杂、厌氧消化调控操作条件繁多以及技术自身反应速率限制等原因影响,厌氧消化技术在处置畜禽粪污过程中存在产气效率低、过程稳定性差的缺陷,掌握其产甲烷效能关键影响因素变得尤为重要。该文分析介绍了不同畜禽粪污的特性以及影响其厌氧消化产甲烷潜力的原因,综述了近年来畜禽粪污厌氧消化的温度、pH、氨氮等主要环境因子以及进料负荷、进料方式等外部调控因素对产甲烷效能、微生物群落丰度的影响研究,探讨了投加功能菌剂、导电材料等辅助方式对系统产甲烷效能提升的作用以及系统微生物的反馈机制,对后续的重点研究方向进行了展望,以期为未来的畜禽粪污厌氧消化技术理论研究与工程应用提供参考。

关键词:畜禽粪污;厌氧消化;产甲烷古菌;产甲烷潜力;导电材料

Research progress on key influencing factors of methane production efficiency in anaerobic digestion of livestock and poultry manure

SUN Guangdong^{1,2}, MA Junyi^{1,2}, HAO Shu^{1,2}, DONG He^{1,2}, ZHAO Xinru^{1,2}, CHAI Qianglong^{1,2}, DANG Yan^{1,2*}

(1. Beijing Key Lab for Source Control Technology of Water Pollution, Beijing Forestry University, Beijing 100083, China;

2. Research Center for Source Control & Eco-remediation of Water Pollution,

Beijing Forestry University, Beijing 100083, China)

Abstract: Under the background of the gradual scale of livestock and poultry breeding methods, the generated manure has the characteristics of large centralized emissions and strong disposal demand, which provides an opportunity for the use of anaerobic digestion technology to dispose of livestock and poultry manure to achieve the production of biogas energy and maximize productivity benefits, which fully meets the strategic needs of China's energy structure optimization and carbon neutrality. However, due to the complex components of livestock and poultry manure, the various operating conditions of anaerobic digestion regulation, and the limitation of the reaction rate of the technology itself, there are defects of low gas production efficiency and poor process stability in the process of anaerobic digestion disposal of livestock and poultry manure, so it is particularly important to understand the key factors affecting its methanogenic efficiency. This paper analyzes and

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第一作者:孙广东(1993-),男,博士研究生,研究方向为有机固废厌氧生物处理和水污染控制。sunguangdong@bjfu.edu.cn

^{*}通信作者:党岩(1983-),男,教授,主要从事有机固废资源化能源化和废水低能耗处理领域研究。yandang@bjfu.edu.cn

introduces the characteristics of different livestock and poultry manures and the reasons influencing their methane-producing potential through anaerobic digestion. It reviews the research on the impacts of major environmental factors such as temperature, pH, and ammonia-nitrogen, as well as external control factors such as feeding load and feeding methods on methane-producing efficiency and microbial community abundance in the anaerobic digestion of livestock and poultry manures in recent years. It also explores the roles of auxiliary methods, such as adding functional microbial agents and conductive materials in enhancing the methane-producing efficiency of the system and the feedback mechanisms of system microorganisms. Moreover, it looks ahead to the key research directions in the future, providing a reference for the theoretical research and engineering applications of anaerobic digestion technology for livestock and poultry manures in the future.

Keywords: manure from livestock and poultry; anaerobic digestion; methanogenic archaea; biochemical methane potential; conductive material

0 引言

由于我国居民饮食结构中畜禽产品占比逐渐增 大,畜禽养殖市场呈现出稳定增长的趋势,畜禽粪污 排放量与日俱增。根据农业农村部等相关数据统 计,全国每年产生畜禽粪污总量近40亿t,约为生活 垃圾年产量的16倍[1]。畜禽粪污中有机质含量为 30%~70%,有效处理率不到50%,畜禽养殖业水污染 物排放量的化学需氧量(COD)约为1000.53万t,占 全国水污染物总排放量的46.67%[2]。畜禽粪污是具 有巨大应用潜力的碳源,其等效二氧化碳排放量约 为 1. 23 亿 t, 为生活垃圾处理碳排放量的 2. 5 倍。厌 氧消化技术通过微生物的代谢活动将废弃有机物稳 定化,同时伴有甲烷和二氧化碳产生,具有处置畜禽 粪污减量无害化、产生沼气可再生能源的双重优势, 引起各国关注[3,4]。随着我国养殖方式从分散饲养向 集约化和现代化的方式快速转变,所产畜禽粪污集 中化程度不断提高,这为厌氧消化技术推广后的设 施规模化、沼气能源产量化、产能效益最大化提供了 可能。

然而,畜禽粪污厌氧消化产甲烷过程历经水解、酸化、乙酸化和甲烷化等阶段,是一种由具有不同代谢机制的微生物共同参与的生物化学反应过程,系统中微生物之间的协同合作既微妙又复杂^[5]。厌氧消化过程涉及微生物主要包括水解酸化细菌和产甲烷古菌,至少涵盖了20个门的细菌和5个目的古菌^[6,7]。产甲烷古菌作为厌氧消化过程关键微生物,是严格的专性厌氧菌,只能利用有限的简单有机物,主要为乙酸、氢气和少数简单有机物^[8]。产乙酸菌和产甲烷古菌之间互营关系成为影响厌氧消化最重要的因素,这两类微生物的不平衡是导致厌氧消化系统不稳定的首要原因^[9]。传统的厌氧消化存在产气效率低、过程稳定性差的缺陷,常常遭遇超负荷、氨

抑制等诱发的过程失稳现象。厌氧消化系统一旦失稳,二次启动运行通常需要较长时间恢复,从而为其所服务的养殖企业或固废环保企业带来极大的环保、经济风险。因此,探明影响畜禽粪污厌氧消化过程稳定以及促进消化产甲烷效率的关键因素,对厌氧消化技术可持续性发展具有重要意义。

在化石能源逐渐枯竭的背景下,依托厌氧消化技术实现畜禽粪污的资源化高效利用,是提升碳回收和生物质能生产效率的重要举措,对助力实现我国的"双碳目标"意义重大^[10]。本文分析了以不同畜禽粪污为底物的厌氧消化产甲烷效能与潜力,总结了畜禽粪污厌氧消化产甲烷过程中不稳定的关键环境因素和干扰机制,探讨了进料负荷和不同进料方式对产甲烷效能的影响,对后期畜禽粪污厌氧消化技术的重点研究方向进行了展望,以期为该技术的基础研究和工程化应用提供参考。

1 不同底物类型厌氧消化产甲烷效能与潜力分析

牛粪、猪粪和鸡粪是我国主要的3种畜禽粪污,分别占畜禽粪污总产量的45.2%、39.2%和15.6%^[11]。由于畜禽消化特征的差异和饲料成分、饲养方式以及粪污收集方式等外部因素,各畜禽粪污的成分结构、理化性质、产甲烷潜能各不相同^[12]。各畜禽粪便的成分对比见表1。

表1 各种畜禽粪便理化性质[13]

Table 1 Physical and chemical properties of various livestock and poultry manure

畜禽粪 污种类	易降解组 分/%	结构性碳水 化合物/%	(VS/ TS)/%	C/N	pН
牛粪	39.49	60.51	75~85	30~60	7.0~8.5
猪粪	47.09	52.92	70~80	10~20	6.5~7.5
鸡粪	59.73	40.27	70~80	3~10	4.0~7.0

注:VS为挥发性固体含量,TS为总固体含量,C/N为碳氮比。

粪污底物中易降解组分含量由高到低依次为鸡粪(59.73%)、猪粪(47.08%)和牛粪(39.49%)。常见的畜禽粪污厌氧消化产甲烷工

艺为连续搅拌式反应器(CSTR)工艺、升流式厌氧污泥床反应器(UASB)工艺,具体工艺流程见图1。

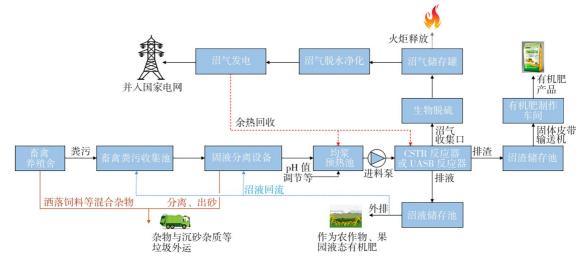


图1 畜禽粪污厌氧消化产甲烷常见的工艺流程

Figure 1 Common process flow chart of anaerobic digestion of livestock and poultry manure for methane production

1.1 牛粪污的厌氧消化产甲烷效能与潜力

牛类反刍动物以草饲料为主,其瘤胃是一个天然发酵罐,瘤胃内部微生物体系具有强大的木质纤维素水解酸化能力,将复杂的木质纤维素生物质转化为可溶性有机物和挥发性脂肪酸(VFAs),通常能够在2d内降解60%以上的纤维^[14]。由于牛饲料中易降解部分已经被牛瘤胃中微生物利用,牛粪中易降解组分与典型的鸡粪、猪粪相比含量最低,含有超过总固体含量50%的不易降解有机组分,包括纤维素、半纤维素和木质素,其复杂的结构而阻碍了牛粪在厌氧消化中的水解,使其成为限速步骤。根据牛粪化学成分分析结果,理论上牛粪的厌氧消化甲烷生产潜力值为206~219 mL/g TS,实际实验中产甲烷潜能为130~139 mL/g TS,仅为前者的60%~70%^[15,16]。

提高牛粪厌氧消化的效能,通常使用预处理、共消化、添加剂等途径来解决。通过预处理可以分解复杂的有机物,使木质纤维素化合物更容易被水解从而提高厌氧消化效能;共消化是通过将不同性质的有机废弃物混合,以提高厌氧消化效能。生物酶添加剂能够将难降解的物质,分解为更易被微生物利用的小分子有机物,从而提升厌氧消化的速率。Fernandez等[17]研究发现在对牛粪进行机械破碎和混合的预处理后,甲烷的产量可以提高12%。与其他畜

禽粪污相比,牛粪中所含的氮化合物含量较低,但结 构性碳水化合物含量最高,属于富碳基质,其较高的 C/N(30~60)也是阻碍厌氧消化的主要障碍之一[18]。 当牛粪废水与其他有机组分进行共消化时,其强大 的缓冲能力更有利于提高甲烷产量,即牛粪的存在 会促进给定底物的厌氧消化过程。Xing等[19]研究发 现当牛粪废水与餐厨垃圾以1:2.5的比例混合时,甲 烷的产率最高可达到 2.71 L/(L·d)。在牛瘤胃内部 的微生物含有产甲烷古菌,其数量占瘤胃微生物总 量的2%~4%。目前已有研究分离出的瘤胃液中产甲 烷古菌主要有4个属:甲烷八叠球菌属 Methanosarcina、甲烷微菌属 Methanomicrobium、甲烷 短杆菌 Methanobrevibacter 和甲烷杆菌属 Methanobacterium^[20,21]。当瘤胃液与牛粪以3:7的比 例混合时,牛粪中的微生物与瘤胃液中的微生物各 自发挥优势,分工协作,甲烷产率最大值达到169 mL/ g VS[22]

1.2 猪粪污的厌氧消化产甲烷效能与潜力

猪粪成分复杂,包括蛋白质、脂肪、有机酸、纤维素、半纤维素等;含氮较多,C/N约为14;脂肪含量(11.18%)显著高于牛粪、鸡粪[13]。相对于保育猪、生长期猪的猪粪,妊娠母猪的猪粪含有更多的纤维素、木质素和芳香族化合物等难降解有机物,这些物质高度腐殖化,难生物降解,保育猪粪便、生长期猪粪

便和妊娠母猪粪便的产甲烷值分别为 273,272,230 mL/g VS,且保育猪粪便和生长期猪粪便的产甲烷过程十分相近^[23]。

由于养殖模式的原因,不少猪场使用水泡粪清 粪工艺清理猪粪便,使得每头猪平均每天产生4~8 L 废水,主要包括洗涤、粪便以及尿液等成分;猪粪受 冲栏洗涤水的掺混,所形成猪粪废水具有排放水量 大,且时间相对集中的特点[24,25]。在一定范围内,随 着猪粪废水TS的增加,厌氧发酵产气效能也增加;若 猪粪废水TS含量较低时,发酵系统内酸浓度低,产甲 烷古菌群底物不足,虽然产气速率快,但容积产气效 率低;若猪粪废水TS过高,形成的水泡粪流动性差, 给猪舍的粪污管理带来麻烦。选择合适的水泡粪TS 含量对其厌氧消化尤为重要。陈欣等[26]研究猪粪废 水 TS 在 2%、6%、10% 和 14% 4个梯度下厌氧消化试 验下,发现TS对猪粪厌氧消化过程及甲烷产率有较 大影响,TS为6%和10%的甲烷产率较高,当TS进一 步提高至14%时,物料中过高VFAs以及氨氮浓度对 产甲烷古菌群活性产生抑制作用。

1.3 鸡粪污的厌氧消化产甲烷效能与潜力

鸡禽类常以谷物为饲料,饮水少,加之鸡禽类的消化道仅为其体长的7倍,比其他畜禽的消化道短,饲料在体内停留仅约4h,营养物质的消化吸收率很低,40%~70%营养物质会随粪便排出体,肥分浓厚^[27,28]。理论上每克干鸡粪全部降解可以产生421~520 mL,实际实验中产甲烷潜能为250~370 mL/g TS,比典型的牛粪、猪粪产甲烷潜能高^[29,30]。

鸡粪属于高氮物质,相对于其他畜禽粪便,含有更多的蛋白质、氨基酸及尿酸,其蛋白质组分最高,

为牛粪、猪粪的2倍[13]。在厌氧消化处理过程中,有 机氮转化为氨氮(包括NH;和游离氨NH。)的效率为 62.6%~80.3%,故高固体鸡粪厌氧消化面临着更大 的氨抑制风险,因此常控制鸡粪厌氧消化TS含量在 10%以下[31]。最初的鸡粪厌氧消化处理研究主要在 低浓度(0.5%~3%)条件下进行,将水加进鸡粪稀释 TS浓度,这也是工程普遍采用的方式,可实现反应器 的快速启动以及后期的稳定运行。由于鸡粪有着较 高的固体含量,更适合于高固体厌氧消化技术,但增 加鸡粪进料TS浓度势必增加反应器发酵液氨氮浓 度,实现高固体鸡粪厌氧处理仍然存在较大的挑 战[32]。Bi等[33,34]研究发现,在鸡粪固体浓度为15%, 水力停留时间(HRT)为20~30 d, 甲烷产率仅为 0.16~0.29 L/g,并且有较高的有机酸残余。鸡粪中 温厌氧消化系统的甲烷转化率会随着有机负荷和总 氨氮(TAN)的升高而受到抑制,在有机负荷达到8.5 kg TS/(m³·d)以上,或TAN浓度达到10000 mg/L以上 时,系统几乎不再产气直至崩溃[35]。

2 环境因子对厌氧消化产甲烷体系的影响

2.1 温度对微生物代谢活动的影响

温度是影响厌氧消化效率的关键参数,对微生物的生长速度和代谢有着重要影响,进而关系到厌氧消化体系中产甲烷性能。根据厌氧微生物适合生存的环境温度不同,可以在低温(15~25°C)、中温(35~40°C)和高温(50~60°C)条件下进行。根据表2总结的产甲烷古菌生长环境参数可知:多数产甲烷古菌的最佳生长温度为30~40°C的中温环境,低温和高温对大多数厌氧微生物而言为极端环境,不适宜生长[36.37]。

表 2 常见的产甲烷古菌属的生长环境参数

Table 2 Growth environmental parameters of common methanogenic archaea

产甲烷古菌属	拉丁学名	基本形态	通用底物	适宜温度/℃	适宜pH范围	
甲烷八叠球菌属	Methanosarcina	不规则球状	氢气/二氧化碳、甲醇、乙酸等	35~40	5.0~7.0	[38]
拟甲烷球菌属	Methanococcoides	不规则球状	甲醇	42	7.0~7.5	[39]
鬃毛甲烷菌属	Methanosa eta	杆状	乙酸	35~60	7.0~7.5	[40]
甲烷丝状菌属	Methanothrix	丝状	乙酸	35~45	7.1~7.8	[8]
甲烷杆菌属	Methanobacterium	长棒长丝	氢气/二氧化碳	35~62	6.6~8.5	[41]
甲烷热杆菌属	Methanothermobacter	长棒长丝、杆状	氢气/二氧化碳	65~70	7.0~8.0	[42]
甲烷短杆菌属	Methan obrevibacter	短棒短链	氢气/二氧化碳、甲酸	37~39	_	[43]
甲烷嗜热菌属	Methanothermus	短棒	氢气/二氧化碳、甲酸	83	<7.0	[42]
甲烷热球菌属	Methanococcus	不规则	氢气/二氧化碳、甲酸	65	_	[42]
甲烷微菌属	Methanomic robium	短棒	氢气/二氧化碳、甲酸	40	6.1~6.9	[42]
甲烷叶形菌属	Methanolacinia	不规则杆状	氢气/二氧化碳	40	7.0	[44]
甲烷螺菌属	Methan ospir illum	弯曲杆长螺旋丝	氢气/二氧化碳、甲酸	30~40	_	[45]

相比中温环境,当温度降低至10°C时,系统内氢型产甲烷古菌甲烷杆菌目(Methanobacteriales)、乙酸型产甲烷古菌甲烷八叠球菌目(Methanosarcinales)菌群丰度均降低,系统内VFAs升高,甲烷产率有所下降^[46]。王华等^[47]对比不同温度条件下厌氧发酵的试验结果发现,在10~30°C范围内,产甲烷古菌的产气能力随着温度的降低而减小,并且其日产气量的最高峰也随之往后推迟。当温度超过40°C时,微生物菌群中的物种数量会急剧下降,47°C可能是中温厌氧产甲烷古菌生长的上限^[48]。

在中温厌氧消化过程中,甲烷的生成可经由乙酸营养和氢营养型产甲烷途径实现,但高温厌氧消化过程则主要通过氢营养型产甲烷途径产甲烷^[49]。虽然高温环境下的厌氧微生物群落多样性较低,但比中温却有更高的有机物降解效率,在短时间内对寄生虫卵的杀灭率较高,有更高的产气量。然而,高温厌氧消化能耗大、能源净输出降低、运行成本较高。因此,考虑到产甲烷的效率与反应器运行能耗之间的平衡,且多样性水平高的微生物菌群具有更高的抗压性和弹性,通常建议畜禽粪污厌氧消化产

生甲烷的温度为35~37℃[50]。

2.2 pH对产酸代谢途径的影响

pH的动态变化会引起微生物群落结构的变化, 对系统稳定性起着至关重要的作用。产甲烷古菌对 pH值极为敏感, 当环境 pH值维持在6.5~7.8时, 对 大多数产甲烷古菌生存比较适宜。根据图2厌氧消 化产甲烷代谢过程可知:约70% 甲烷是产甲烷古菌 以乙酸为底物代谢的结果,目前为止,乙酸营养型产 甲烷古菌只发现了甲烷八叠球菌属 Methanosarcina 和 甲烷丝状菌属 Methanothrix, 它们的最适 pH 范围为 6.8~7.2。而水解与产酸菌对pH值有较大范围的适 应性,大多数产酸菌群在pH值为5.0~8.5内生长良 好[51]。当厌氧消化体系中pH值为5.5时,产甲烷速 率下降80%;pH值<5.0时,厌氧体系几乎不再产生 甲烷。Sun等[52]研究发现,当pH值为5.1时,几乎 90%的乙酸营养型甲烷菌和氢营养型甲烷菌在6d 的培养过程中从培养物中消失,低pH环境提高了产 甲烷微生物的死亡速度,且这种抑制是不可逆的。 因此,pH也是判断厌氧消化产甲烷反应是否正常进 行的重要指标。

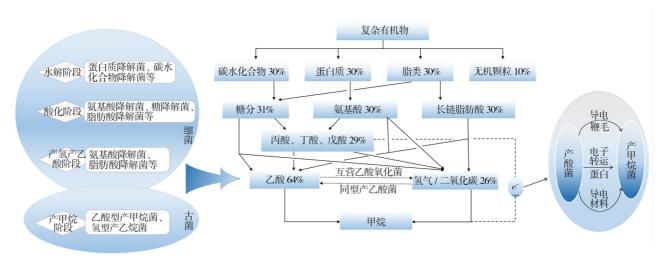


图 2 厌氧消化产甲烷代谢过程

Figure 2 Anaerobic digestion methanogenic metabolic process

pH可以改变厌氧消化过程中产酸代谢途径,导致 VFAs 的种类和含量产生很大变化^[53]。产酸代谢过程中所产生的乙酸盐、丙酸酯与丁酸酯的比例通常是 75:15:10至 40:40:20^[54]。Jiang等^[55]研究发现当 pH 值为 5.0 时,产酸消化的主要产物是乙酸;但当 pH 值为 6.0 或 7.0 时,主要产物则是丁酸。当厌氧发酵体系具有良好的缓冲能力时,局部的 VFAs 浓度变化过高不会显著降低体系的 pH,对甲烷转化效率不

会有显著影响,系统中产氢产乙酸菌群和产甲烷古 菌群代谢动态平衡在此起着关键作用。

2.3 氨氮对厌氧消化系统的影响

在厌氧消化过程中,氨氮是微生物生长所必需的营养物质,主要由发酵原料中的含氮有机化合物降解而产生,如蛋白质、氨基酸、尿素、核酸等,并以NH^{*},和NH₃(FAN)分子的形态存在,二者之和为TAN^[56]。氨氮在一定程度上可中和厌氧消化过程中

所产生的有机酸,保证厌氧消化系统具有足够的缓冲能力,维持了细胞生长所必需的中性pH条件,从而提高厌氧消化系统的稳定性。但粪污在厌氧消化过程中,具有含氮高的特点,过量的氨氮会对厌氧消化产生抑制,影响厌氧消化系统的性能,甚至导致其失稳^[57]。厌氧微生物对原料的C/N也有一定的要求,碳的消化速率比氮的速率快30~35倍,C/N的最佳范围在理论上在30~35之间,通过加碳或氮来改变体系中的碳氮比,以维持厌氧消化反应的良好运转。

乙酸营养型产甲烷古菌 Methanosaeta 和 Methanosarcina 对氨氮更敏感,而嗜氢产甲烷古菌具有较好的氨氮耐受性。当氨氮浓度为1300 mg/L时,

乙酸营养型产甲烷古菌 Methanosarcina 开始受到抑制^[58]。当氨氮浓度升高至 3500 mg/L以上时,嗜氢产甲烷古菌 Methanoculleus 也会受到不同程度的氨抑制^[59]。 孟晓山等^[61]研究发现,猪粪厌氧消化系统中氨氮超过 4000 mg/L,将促使产甲烷古菌群结构发生显著变化,乙酸营养型产甲烷古菌 Methanosaeta 逐渐被 Methanosarcina 代替,而氢利用型产甲烷古菌属中 Methanospirillum 的优势性逐渐被 Methanoculleus 和 Methanomassiliicoccus 取代,高浓度氨氮也会促使产甲烷途径由乙酸利用型为主向氢利用型为主转变。在 畜禽粪污厌氧消化过程中,氨氮抑制产甲烷效果的相关情况总结见表 3。

表 3 畜禽粪污厌氧消化氨氮抑制产甲烷效果

Table 3 Results of ammonia inhibition during anaerobic digestion of livestock and poultry manure

底物	TS/%	温度/%	TAN浓度/ (mg/L)	FAN浓度/(mg/L)	产甲烷抑制情况	参考文献
牛粪	6.4	55	>4000	>3000	较TAN1500 mg/L时,减少25%	[62]
牛粪	2.9	55	5000	3500	较抑制前减少67.3%	[63]
牛粪	4.2	40~60	6000	>8000	较TAN2500 mg/L时,减少40%	[64]
牛粪	2.0	55	6200	7000	较抑制前减少38%	[65]
			1600	-	较未添加氮源减少8%	
猪粪	2.3	37	3200	-	较未添加氮源减少62%	[66]
			4000	-	较未添加氮源减少83%	
X-1x -34:	4.5	37	6000	-	较理论值减少38%	[67]
猪粪	4.5	55	6000	-	较理论值减少78%	[67]
猪粪	15.6	25	4000	2000	较TAN1600 mg/L时,减少45%	
X±x -¥4:	2.0	35	4000	< 1000	较TAN1600 mg/L时,减少13%	[68]
猪粪	2.0	34	>5200	-	完全不产气	
鸡粪	10	55	6000		较抑制前减少50%以上	[69]
鸡粪	3~8	55	500~3500		逐渐适应OLR的提升,未减少	[70]
鸡粪	15	40	5590		较抑制前减少10%	[71]
ah 46	20	26	6960	-	较抑制前减少32%	[72]
鸡粪 30	36	9000	_	彻底酸化,不产气	[72]	

3 进料负荷和进料方式对厌氧消化产甲烷效能的 影响

3.1 水力停留时间对厌氧消化产甲烷效能的影响

有机负荷率(OLR)是运行厌氧消化系统的重要工程控制参数,通过调整 HRT可改变系统的有机负荷,从而平衡畜禽粪污厌氧消化产酸速率与产甲烷效率,实现最佳的产气性能^[73]。厌氧消化反应器产气量会随进料有机负荷的增加而加大,但当 HRT缩短到一定程度,则会因负荷过高而造成有机挥发酸的积累,尤其是丙酸,对厌氧消化过程产生抑制,从而导致产气量降低^[74]。HRT从 10 d减少到 7.5 d,促进了甲烷八叠球菌属 Methanosarcina 的生长,相对甲烷丝状菌属 Methanothrix 具有优势^[75]。单纯的粪污

厌氧消化效率相对较低,抗降解成分需要较高的HRT。通过与餐厨垃圾、粗甘油、秸秆等物料共消化可有效提高甲烷产量。在牛粪与餐厨垃圾共消化的研究中,发现缩短HRT(25,20,15,10,7,5,4d)会减少甲烷转化率,系统中拟杆菌门(Bacteroidetes)和厚壁菌门(Firmicutes)成为优势菌,醋酸合养氧化菌(主要是Pelotomaculum和Pseudothermotoga)的比例明显增加;优势产甲烷古菌由乙酸营养型产甲烷古菌转变为嗜氢型产甲烷古菌^[76,77]。在不同的HRT下,不同类型底物的厌氧消化系统中产甲烷古菌变化情况见表4。

为提高畜禽粪污的处置效率,常见的减少HRT的方法有混合搅拌和预处理。混合搅拌可使消化物

表 4 不同水力停留时间的厌氧消化系统产	甲烷古菌变化情况
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Table 4 Changes of methane archaea in a	maerahic digestian system	under different HRT

底物	ρ(TCOD)/ (g/L)	HRT	厌氧消化系统产甲烷古菌丰度变化情况	参考 文献
牛粪和食物 垃圾	74.6 ± 3.5	20 d缩短至 5 d	在 HRT 为 20 d时, Methanosarcina 丰度占产甲烷古菌总丰度的 86.7%, 同时含有Methanobrevibacter、Methanospirillum、Methanosaeta 等多类产甲烷古菌;在 HRT 为 5 d时, Methanobrevibacter 占比达 90% 以上,产甲烷古菌丰度降低明显	[76]
猪粪和食物 垃圾	40.0 ± 0.2	41 d缩短至 21 d	Methanobacterium 和 Methanobrevibacter 的丰度随着 HRT 的降低而降低, Methanosarcina 和 Methanoculleus 的丰度保持相对稳定	[78]
鸡粪	494 ± 82	60 d缩短至 40 d	缩短 HRT 使产甲烷古菌的优势菌种明显从 Methanoculleus 向 Methanobacterium 转变, Methanobacterium 在 HRT 为 40,60 d 时分别占比 48.45% 和 12.45%, Methanoculleus 在 2个系统中分别占比 1.94% 和 83.14%	[35]
葡萄糖	0.25	2次/d、1次/d、 0.5次/d	所有系统以 Methanosaeta 为主; 在较低进料频率下, Methanosaeta 的丰度略有减少	[79]

料混合和分布均匀,防止物料在发酵罐底部沉淀,增大反应物与微生物的接触面积,有利于保证良好的消化速度,从而提高消化产气量。Karim等^[80]研究发现,机械搅拌、沼气回流搅拌、出水回流搅拌和未搅拌相比,厌氧消化的产气量分别提高了22%、15%和29%。预处理方法中热水解法应用最为广泛,粪污预处理后的营养成分更容易被微生物利用,厌氧消化能力提高,停留时间也相应减少^[81]。宋晓聪等^[82]将牛粪于70℃处理3d后进行厌氧消化,发现其产气量较未处理的牛粪提高了210%,有机组分降解和木质素含量降低是产气性能提高的原因。

3.2 不同进料方式对厌氧消化产甲烷效能的影响

不同的进料方式可能造成微生物群落差异,进 而影响产气率。在较高的OLR下,低进料频率可能 在短时间内对系统造成较重的负荷冲击,高进料频 次可以有效降低消化系统的负荷冲击,使系统产生 的 VFAs 能够被及时消耗^[83]。序批式间歇流反应器 (SBR)的低进料频次会导致底物梯度随时间变化,初 始底物浓度较高。相反,连续流反应器(CFR)的长进 料时间会促使系统时刻保持稳定和低底物浓度状 态[84]。Kine 等[85]研究发现,每天进料1次时,丙酸的 浓度峰值为2500 mg/L,而每2.4 h进料1次时,丙酸 浓度始终保持在40 mg/L以下。甲烷八叠球菌属 Methanosarcina 在间歇式进料的情况下表现出一定优 势,而甲烷丝状菌属 Methanothrix 则表现出相反的行 为^[86]。在较低 OLR下,不同进料频次均未产生较重 的负荷冲击,酸化现象一般不会出现;且无论HRT或 进料方式的如何变化,甲烷丝状菌属 Methanothrix 均 为主要的产甲烷古菌属[87]。

4 外部添加材料对厌氧消化产甲烷效能的提升

4.1 投加功能菌剂对厌氧消化产甲烷效能的提升

向反应系统中投入具有一定功能的微生物功能 菌剂,能够在很大程度上提高生化效率,达到加快厌 氧系统启动时间、缩短 HRT、增加难降解原料利用 率、促进酸败系统的恢复、应对氨抑制等目标[88]。根 据目标产物的不同,可将其划分为水解产酸、产氢产 乙酸、产甲烷阶段的生物强化,其促进效果见表5。 由于畜禽粪污中含有大量的木质纤维素,有效发酵 成分不易降解,对水解产酸阶段生物强化,促进木质 纤维原料的水解,为厌氧消化可提供更多易降解的 底物。Henrik等[89]研究发现,向牛粪及其分离组中投 加乳酸杆菌 Caldicellulosiruptor lactoaceticus 可以使得 甲烷产率提高 10%~24%。然而, Costa 等[90]研究发 现,尽管在高温厌氧发酵畜禽粪便体系中投加热线 梭菌 Clostridium thermocellum 及解糖纤维素分解菌 Caldicellulosiruptor saccharolyticus, 虽然原料水解率提 高了,但甲烷的产量并没有明显增多,这是由于产甲 烷速率跟不上纤维素水解产酸菌速率,导致 VFAs 累 积,抑制了产甲烷。通过对水解产酸阶段的生物强 化,向系统中投加产氢菌,所产的沼气中氢气含量并 未增加,即甲烷与氢气组分比例无明显减小,说明投 加产氢菌所增加的氢气是以中间产物存在的。氢气 作为产甲烷古菌的底物,被进一步用于产甲烷。此 外,产氢菌的生物强化还可使甲烷菌的数量增加,即 氢营养型产甲烷古菌在数量上得到了强化[91]。

根据图 2 厌氧消化产甲烷代谢过程可知:产酸细菌和产甲烷古菌之间存在种间电子转移(IET),该方式是菌属之间互养代谢产甲烷极其重要的环节。由

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Table 5	Dramating offeat	of hiofunational he	antorioties on e	naerobic digestion
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生物强化菌	主要促进阶段	反应底物	强化效果	参考文献
Caldicellusiruptor lactoaceticus	水解产酸阶段	牛粪及其分离组分	提高甲烷产量10%~24%	[92]
Clostridium cellulolyticum	水解产酸阶段	家禽垃圾	提高累计甲烷产量15%	[90]
Fervidobacterium pennivorans	水解产酸阶段	鸡羽毛	提高水解率40%,未提高甲烷产量	[93]
${\it Caldicellulosiruptor\ saccharolyticus}$	产氢产酸阶段	猪粪	提高沼气产量60%~70%	[94]
C.saccharolyticusl Enterobacter Cloacae	产氢产酸阶段	猪粪+甜高粱	提高沼气产率30%	[95]
Clostridium sp. $WY-1$ Clostridium sp. $WW-5$	产氢产酸阶段	猪粪	提高甲烷产量3%~11%	[96]
Methanosaeta	产甲烷阶段	废水	提高沼气产量10%~12%	[97]

于较快的水解发酵产酸和较慢的产甲烷之间的不平衡,从而导致畜禽粪污厌氧消化处理负荷只能维持在相对较低效能,产甲烷效率受到了限制^[98]。IET产甲烷途径是微生物菌群降低热力学能垒、实现互养代谢快速产甲烷的核心过程。互养代谢产甲烷过程除了常见已知的种间氢转移^[99]和种间甲酸转移^[100]之外,美国马萨诸塞大学 Derek Lovley 教授团队^[101]于2010年在国际上首次报道了电活性微生物硫还原地杆菌 Geobacter sulfurreducens 和金属还原地杆菌 Geobacter metallireducens 之间存在直接种间电子传递(DIET)过程,这种代谢方式不需要电子穿梭体,可以突破传递过程的速率限制,能够以更高的电子转移效率或较低的能量消耗直接将电子转移至二氧化碳,将二氧化碳还原为甲烷,可极大提升产甲烷速率。目前已发现2类种间直接电子传递形式,一类是依靠细胞外

膜上的c型细胞色素,另一类是通过导电菌毛(pili)[102]。

通过对参与DIET产甲烷途径的互养代谢微生物群落结构进行厌氧消化调控,增加可参与DIET过程的微生物丰度,进而提高产甲烷效率。Jiang等^[92]研究发现通过添加富集的丙酸降解产甲烷培养基,培养基中所含优势菌属为甲烷丝状菌属 Methanothrix (66.15%~94.70%)和甲烷杆菌属 Methanobacterium (4.81%~33.56%),相比与非生物强化组,系统中甲烷丝状菌属 Methanothrix 的丰度超过80%,有机负荷和沼气产量分别提高8倍和12倍。目前报道的确认可以参与直接种间电子传递的菌属(表6)可知:供电子微生物多为具有胞外电子传递能力的微生物,如地杆菌属 Geobacter,而受电子的产甲烷古菌则主要是可以利用乙酸盐的鬃毛甲烷菌属 Methanosaeta 和甲烷八叠球菌属 Methanosarcina。

表 6 可参与种间直接电子传递型互营产甲烷过程的微生物

Table 6 Microorganisms participated in DIET-type methane production process

底物	供电子微生物	受电子微生物	参考文献
乙醇	Geobacter metallireducens	Methanothrix harundinacea	[103]
乙醇	Closstridium spp.	Methanosarcina mazei	[103]
乙醇	Syntrophomonadaceae	Methanos arcinaceae	[104]
乙醇	$Geobacter\ metallireducens$	Methanosarcina barkeri	[105]
乙醇	$Geobacter\ metallireducens$	$Methanosarcina\ acetivorans$	[105]
乙醇、葡萄糖	Bacteroidales, Corynebacterium	Methanosarcina spp.	[106]
乙酸盐	$Geobacter\ metallireducens$	Methanosarcina barkeri	[105]
乙酸	Proteinic lasticum, Kosmotoga	Methanosarcina spp.	[103]
1-丁醇	Geobacter	Methanosa eta	[105]
硫代硫酸盐	$Rhodopseudomonas\ palustris$	Methanosarcina barkeri	[107]
牛粪	Clostridium	Methanosarcina spp.	[108]

微生物功能菌剂投加成功的关键是能否在系统中持久稳定地发挥作用,所投加的微生物功能菌繁殖速度需高于其被土著菌吞噬及流失的速度,保证生物强化菌剂在被强化体系内的生物净持有量。为增加生物强化菌剂在被强化体系中的稳定性,可将生物强化菌剂固定化在载体上再进行投加,固定化

的载体主要有沸石、海泡石等[109,110]。对于连续流系统,在不断地进出料冲击及土著微生物的竞争下,微生物功能菌剂容易逐渐被淘汰,定期多次投加生物强化菌剂效果优于一次性投加[111]。

4.2 投加导电材料对厌氧消化产甲烷效能的提升 采用导电材料强化 DIET产甲烷途径的畜禽粪污 厌氧处理技术,是提高畜禽粪污资源利用的有效途径和研究热点。在厌氧系统中投加外源导电材料(如颗粒活性炭[112]、碳布[113]、生物炭[114]、石墨烯[115]、磁铁矿[116]、磁性颗粒活性炭[117]、聚苯胺[118]等)可强化甲烷八叠球菌属 Methanosarcina 等产甲烷功能菌参与DIET 过程,进而提高厌氧体系甲烷转化效率,其具体影响效果见表7。外源导电材料具有高导电性、化学稳定性、吸附能力强等特点,能够作为生物导电连接重要补充;同时,不需要微生物消耗能量以合成生物电连接的自身导电结构,微生物可以利用更多能量于代谢作用。Pan等[119]研究发现,添加生物炭可以提

高鸡粪、牛粪及其混合物的产气潜力,并能提高所产沼气的甲烷含量,通过吸附作用,降低氨氮和硫化氢的含量。Lee等[120.121]研究发现在系统中投加生物炭和活性炭后,其表面富集了鬃毛甲烷菌属Methanosaeta、甲烷螺菌属Methanospirillum和甲烷杆菌属Methanobacterium等产甲烷古菌。Zeng等[122]探究了碳基导电材料负载核黄素后对畜禽粪污厌氧消化的影响,通过投加核黄素改变了微生物群落结构并提高了与DIET相关的功能基因丰度,最终提高了反应器的容积产气率和有机物去除率。

表7 导电材料对畜禽粪污的厌氧消化影响

Table 7 Effect of conductive materials on anaerobic digestion of livestock and poultry manure

碳基导电材料	底物	剂量	产甲烷促进效果	参考文献
生物炭	秸秆和鸡粪	0.1 g/L	提高甲烷产量 27.8%~96.4%	[123]
纳米四氧化三铁	猪粪	_	提高甲烷产量17.37%	[124]
微米级Fe ⁰	猪粪	75 mmol/L	提高甲烷产量 23.9%	[125]
W-碳基	活性污泥+牛粪	5 g/L	缩短厌氧消化时间,甲烷产量提高58.8%	[126]
颗粒活性炭	鸡粪	100 g/L	提高甲烷产量 26.7%	[127]
生物炭	鸡粪	5%(占干物质比例)	提高甲烷产量 68.97%	[128]
磁铁矿	鸡粪	100 g/L	提高甲烷产量 2.8%	[127]

4.3 投加非导电材料对厌氧消化产甲烷效能的 提升

在厌氧系统中投加外源非导电材料(如尼龙纤 维[129]、聚乙烯[130]、碱性树脂[131]等)也可强化厌氧体系 产甲烷效率。非导电材料能够提升甲烷产率主要是 因其比表面积大,易于吸附定植相关功能微生物,如 厌氧消化细菌、产甲烷古菌等,使其能够高效地参与 到厌氧消化过程,部分载体还具有吸附能力、离子交 换能力、优化菌群结构等特点[132]。不同材质类型、粒 径及浓度对厌氧消化的影响存在较大的差异,还有 可能存在抑制厌氧消化的情况。Zhang等[133]发现在 55,65 ℃下,聚乙烯塑料的存在能使奶牛粪污厌氧消 化的甲烷产量分别提高8.4%、41.2%,COD降解率分 别提高到52.8%和52.4%。而厌氧消化系统中塑料 含量超过0.2 g/L对厌氧消化有不良影响,甲烷产量 减少了17.9%~19.3%,这是由于厌氧消化系统中存 在的微塑料物质影响了微生物种群的丰度和结构, 使得甲烷杆菌属 Methanobacterium 和鬃毛甲烷菌属 Methanosaeta 的丰度降低[134]。

5 结束语

厌氧消化技术是处置畜禽粪污有效手段,通过 对比分析牛粪、猪粪、鸡粪的粪污特性,牛粪因含有 较多不易降解有机组分进而产甲烷潜力最低,鸡粪 因含有较多营养物质进而产甲烷潜力最高。适宜的 温度、pH、氨氮等环境条件是保障畜禽粪污厌氧消化 稳定性运行的基础。基于系统产甲烷微生物的代谢 环境要求,pH、氨氮变化也成为判断厌氧消化产甲烷 反应是否正常进行的重要指标。不同的 HRT、进料 方式能够改变系统产甲烷古菌类型,也会对禽粪污 厌氧消化产甲烷效能产生影响。基于厌氧消化影响 的关键因素探究,通过功能菌剂、导电材料等材料的 投加,调控系统功能性微生物群落丰度,强化电活性 微生物之间的直接种间电子传递,促进厌氧消化高 负荷运行,进而提升产甲烷效能。未来的畜禽粪污 厌氧消化产甲烷效能提升研究,应重点关注以下 方面:

1)产甲烷古菌的 DIET 代谢路径机制研究。从细胞水平对产甲烷古菌的 DIET 代谢路径进行优化提速,是提高系统有机质转化率以及容积负荷的重点突破口。产甲烷速率低于产酸速率时,有机酸在体系中积累造成系统酸化,酸积累导致厌氧发酵系统酸化的抑制作用。随着微生物菌群种间电子传递研究的深入,产甲烷功能菌株、导电材料也逐渐成为厌氧发酵失稳调控策略的研究热点,如何构建特异性

产甲烷功能菌株、改性导电材料来提高厌氧发酵系统稳定性和处理效率,成为理论研究亟待解决的问题。

2)智能化调控的厌氧消化装备研发。为精确控制厌氧消化过程中的关键参数,如温度、pH值、搅拌强度、进料负荷等,创造最适宜微生物生长和产甲烷的环境条件,可通过先进的传感器技术和在线监测设备,实时监测厌氧消化过程中的各项参数,建立智能化的控制系统,进而实现对消化过程的精准调控。基于自动化运行控制,及时调整工艺参数,可避免因参数波动导致的系统不稳定和产甲烷效率下降。

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