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## 内环境调节层对厌氧生物反应器填埋场中氮转化的影响

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摘要: 为探究内环境调节层对厌氧生物反应器填埋场内环境和氦组分的长效影响,特设矿化垃圾+重质碳酸钙、矿化垃圾+天然沸石这2种内环境调节层分别置入模拟反应器 R2和R3中,同时设 R1(不含内环境调节层)作为对照,监测分析 390 d内固相垃圾和渗滤液中氦组分的变化. 结果表明,R1、R2和R3中pH、碱度、氧化还原电位(Eh)、含水率(MS)的大小关系分别为 pH(R2) > pH(R3) > pH(R1)、碱度(R2) > 碱度(R3) > 碱度(R1)、Eh(R2) < Eh(R3) < Eh(R1)、MS(R3) > MS(R2) > MS(R1). R1、R2和R3中垃圾全氮降解转化率为 79.2%、82.3%和88.5%,氨氮为48.3%、60.1%和67.7%,硝态氮为38.5%、44.2%和53.4%;渗滤液中总氮、氨氮和硝态氮的浓度对比分别为 TN(R3) < TN(R2) < TN(R1)、NH<sub>4</sub><sup>+</sup>-N(R3) < NH<sub>4</sub><sup>+</sup>-N(R1) < NH<sub>4</sub><sup>+</sup>-N(R2)和NO<sub>3</sub><sup>-</sup>-N(R3) < NO<sub>3</sub><sup>-</sup>-N(R2) < NO<sub>3</sub><sup>-</sup>-N(R1). 总体看来,矿化垃圾+ 更质碳酸钙、矿化垃圾+ 天然沸石组成的内环境调节层均能长期优化内环境,为氮的降解转化提供有利条件,而矿化垃圾+ 天然沸石不仅能促进垃圾和渗滤液中氮组分的降解转化,还能一定程度上控制渗滤液循环造成的氨氮累积.

关键词:内环境;调节层;氮;厌氧生物反应器填埋场;转化

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# Effect of Environment Adjustment Layers on Nitrogen Transformation in Anaerobic Bioreactor Landfills

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Abstract: To investigate the perennial effect of environment adjustment layers on the interior environment and nitrogen transformation in anaerobic bioreactor landfills, three sets of simulated anaerobic bioreactor landfills and two kinds of environment adjustment layers of mineralized refuse with heavy calcium carbonate (R2) and mineralized refuse with natural zeolites (R3) were designed and established. The degradation and transformation of nitrogen in waste and leachate had been monitored for 390 days. The results showed that, the value orders of pH, alkalinity, oxidation reduction potential and moisture content (MS) were pH(R2) > pH(R3) > pH(R1), alkalinity (R2) > alkalinity (R3) > alkalinity (R1), Eh(R2) < Eh(R3) < Eh(R1) and MS(R3) > MS(R2) > MS(R1). In R1, R2 and R3, the degradation rates of total nitrogen, ammonia nitrogen in waste were 79.2%, 82.3% and 88.5%, 48.3%, 60.1% and 67.7%, 38.5%, 44.2% and 53.4%, respectively. Concentration comparison results of total nitrogen, ammonia nitrogen and nitrate nitrogen in leachate were TN(R3) < TN(R2) < TN(R1), NH<sub>4</sub><sup>+</sup>-N(R3) < NH<sub>4</sub><sup>+</sup>-N(R1) < NH<sub>4</sub><sup>+</sup>-N(R2) #INO<sub>3</sub><sup>-</sup>-N(R3) < NO<sub>3</sub><sup>-</sup>-N(R2) < NO<sub>3</sub><sup>-</sup>-N(R1). Additionally, both of mineralized refuse with heavy calcium carbonate and mineralized refuse with natural zeolites could long-term adjust and optimize the interior environment of anaerobic bioreactor landfills for the degradation and conversion of nitrogen. Mineralized waste with natural zeolite could not only promote the degradation and transformation of nitrogen components in waste and leachate, but also control the accumulation of ammonia nitrogen through leachate recirculation.

Key words; interior environment; environment adjustment layers; nitrogen; anaerobic bioreactor landfill; transformation

目前大量的城市生活垃圾主要通过填埋处理<sup>[1]</sup>,厌氧生物反应器填埋场能促进垃圾降解<sup>[2]</sup>,去除渗滤液中污染物<sup>[3]</sup>,加速填埋场的稳定化<sup>[4]</sup>,因此得到快速的发展和运用<sup>[5]</sup>. 氮是垃圾的重要组分<sup>[6]</sup>,填埋场中垃圾所含的有机氮经水解氨化等作用转化为氨氮<sup>[7]</sup>. 在厌氧生物反应器中,仅有小部分氨氮通过挥发为 NH<sub>3</sub> 或经厌氧氨氧化作用生成 N<sub>2</sub> 等方式转化去除<sup>[8]</sup>,其余大部分仍留在垃圾体和

渗滤液中<sup>[9]</sup>,因此,在渗滤液持续回灌或循环过程中易造成氨氮累积<sup>[10]</sup>.大量的氨氮将会和积累的有机酸一起破坏填埋场中的内环境<sup>[11]</sup>,抑制微生物

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对垃圾的降解<sup>[12]</sup>.一些研究通过添加碱性物质<sup>[13]</sup>、提供微氧<sup>[14]</sup>和对渗滤液进行异位硝化处理后回灌<sup>[15]</sup>等措施来缓解有机酸和氨氮的累积,改善厌氧填埋场的内环境,促进氮的转化和去除,取得了一定的效果.然而这些措施操作较复杂,在持续性和经济实用性等方面也并不理想.为长效、简单地对内环境调节和优化,本研究在固相垃圾体中构建内环境调节层,重点探讨不同内环境调节层对厌氧生物

反应器填埋场中氮转化去除的影响.

#### 1 材料与方法

#### 1.1 实验材料

#### **1.1.1** 垃圾来源及性质

垃圾采自长春市垃圾中转站,去除金属、橡胶、玻璃和陶瓷等不可降解物质,破碎至2 cm. 预处理后垃圾具体组成见表1.

表 1 垃圾初始物理组成

Table 1 Initial physical composition of refuse

组成	餐厨	果皮	纸	木材	纺织	沙土	其它混合类
湿重质量分数/%	25. 82	35. 60	26. 70	1. 10	2. 53	2. 31	5. 93

垃圾含水率 (moisture content, MS) 为 (34.97%  $\pm 1.33\%$ ),全氮 (total nitrogen of solid waste, TNs) 质量分数为 (19 200.0  $\pm$  300.0)  $\mathrm{mg}\cdot\mathrm{kg}^{-1}$ ,氨氮 (NH<sub>4</sub><sup>+</sup>-N)<sub>s</sub> 为 (89.7  $\pm$  2.8)  $\mathrm{mg}\cdot\mathrm{kg}^{-1}$ ,硝态氮 (NO<sub>3</sub><sup>-</sup>-N)<sub>s</sub> 为 (51.3  $\pm$ 0.8)  $\mathrm{mg}\cdot\mathrm{kg}^{-1}$ .

#### 1.1.2 垃圾渗滤液来源及性质

垃圾渗滤液取自长春市蘑菇沟垃圾填埋场,垃圾填埋龄在2 a 以下,具体性质见表2.

#### 1.1.3 内环境调节层材料

采用矿化垃圾、重质碳酸钙和天然沸石作为内

环境调节层材料,具体性质见表3.

表 2 垃圾渗滤液初始理化性质1)

Table 2 Initial physical and chemical properties of leachate

组分	值	组分	值
pH	5. 17	TN/mg·L <sup>-1</sup>	812. 0
Eh/mV	- 189. 7	$NH_4^+$ -N/mg·L <sup>-1</sup>	381.7
碱度/mg·L-1	7 131. 8	TN/mg·L <sup>-1</sup> NH <sub>4</sub> <sup>+</sup> -N/mg·L <sup>-1</sup> NO <sub>3</sub> <sup>-</sup> -N/mg·L <sup>-1</sup> NO <sub>2</sub> <sup>-</sup> -N/mg·L <sup>-1</sup>	1.0
TOC/mg·L <sup>-1</sup>	5 985. 0	$NO_2^-$ -N/mg · L $^{-1}$	0. 2

1) 渗滤液中总氮、氨氮、硝态氮和亚硝态氮分别表示为 TN、 $NH_4^+-N$ 、 $NO_3^--N和NO_2^--N$ ;固相垃圾中全氮氨氮、硝态氮和亚硝态氮分别表示为  $TN_s$ 、 $(NH_4^+-N)_s$ 、 $(NO_3^--N)_s$  和 $(NO_2^--N)_s$ 

表 3 内环境调节层材料的性质

Table 3 Characteristics of interior environment adjustment layer materials

材料	粒径	含水率	$\mathrm{TN}_{\mathrm{s}}$	$(NH_4^+-N)_s$	$(NO_3^N)_s$	На
17) 144	/mm	/%	/mg·kg <sup>-1</sup>	/mg·kg <sup>-1</sup>	/mg•kg <sup>-1</sup>	рп
矿化垃圾	0.50 ~ 2.00	22. 3	1 300. 0	17. 6	27. 1	8. 35
重质碳酸钙	$0.50 \sim 2.00$	0.8	0.0	0. 0	0.0	10. 62
天然沸石	$0.50 \sim 2.00$	1. 2	0.0	0. 0	0.0	7. 23

#### 1.2 实验设计

#### 1.2.1 实验装置

实验装置如图 1 所示,反应器为有机玻璃槽 (40 cm×30 cm×40 cm). 垃圾分两层装填到反应器中,单层垃圾质量为 9 kg,高度为 14 cm,密度约为 550 kg·m<sup>-3</sup>. 两层垃圾之间为内环境调节层,厚度为 2 cm. R2 内环境控制层为 1 kg 矿化垃圾 +1 kg 重质碳酸钙,R3 为 1 kg 矿化垃圾 +1 kg 天然沸石,R1 无.

#### 1.2.2 实验运行

初始运行时,向3组反应器各加入5L新鲜垃圾渗滤液,并置于室温下运行390d.渗滤液每隔3d循环一次,每10d补充50mL去离子水模拟降水.

#### 1.3 指标分析

垃圾通过前后4个采样孔(如图1)采集,立即

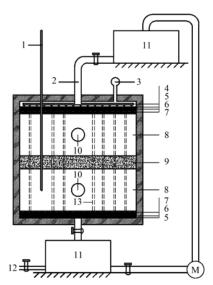
采用烘干法(CJ/T 313-2009)测定含水率,其后粉碎并混合均匀,采用四分法取样并用半微量开氏法(CJ T 103-1999) 测定  $TN_s$ 、( $NH_4^+$ -N)。和( $NO_3^-$ -N)。等.

渗滤液通过下方渗滤液取样口采集,并按照文献[16]规定的方法测定 pH、碱度、氧化还原电位(Eh)、总氮 (TN)、氨氮 (NH $_4^+$ -N)、硝态氮 (NO $_3^-$ -N) 和亚硝态氮 (NO $_2^-$ -N) 等指标.

#### 2 结果与分析

#### 2.1 内环境参数

由图 2 可见, R1、R2 和 R3 中 pH 和碱度在初始 10 d 迅速下降到最小值, 分别为 3.77、3.20、3.27 和1 332.2、1 276.4、1 122.7  $\text{mg} \cdot \text{L}^{-1}$ , 这主要是由于垃圾中大量有机酸溶入渗滤液中造成[17].

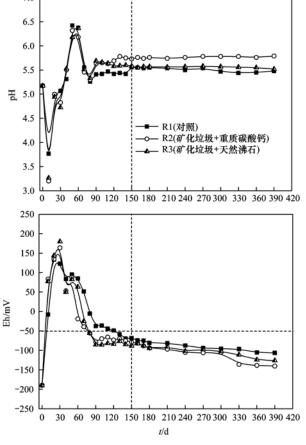


温度计; 2. 渗滤液循环孔; 3. 真空集气袋; 4. 布水管;
 钢丝网; 6. 石英砂; 7. 细碎石; 8. 垃圾层; 9. 内环境调控层; 10. 垃圾取样孔; 11. 集水槽; 12. 渗滤液取样口; 13. 液/气流动管

#### 图 1 厌氧生物反应器装置示意

Fig. 1 Schematic diagram of anaerobic bioreactor

10~60 d,由于微生物对有机酸的降解,pH 和碱度升高到最大值,分别为 6.42、6.32、6.36 和



7 784. 6、8 445. 7、7 465. 7  $mg \cdot L^{-1}$ . 此后,反应器内氧气耗尽,厌氧微生物开始水解酸化垃圾中有机物,pH 和碱度逐渐下降,150 d 后趋于稳定. 实验结束时,R1、R2 和 R3 中 pH 和碱度分别为 5. 48、5. 79、5. 59 和4 267. 5、5 198. 8、4 689. 3  $mg \cdot L^{-1}$ .

Eh 初始阶段,R1、R2 和R3 中Eh 均迅速升高,均在第30 d 分别达到最大值122.9、163.7 和179.7 mV,这是由在垃圾的装填过程中携带进入的氧气造成.30 d后,Eh 不断下降,3 组反应器逐渐进入厌氧状态.150 d后,R1、R2 和R3 中Eh 均在-50 mV以下且持续下降,实验结束时分别为-106.2、-140.3和-126.1 mV.

渗滤液开始循环后,垃圾体 MS 逐渐升高,分别在 120 d、120 d 和 60 d 达到最大值 58.0%、67.4%和 62.3%. 此后由于蒸发和微生物利用等因素<sup>[18]</sup>,3组反应器中的 MS 均有不同程度下降,150 d 后基本处于稳定.实验结束时,R1、R2 和 R3 中的 MS 分别为 46.5%、51.9%和 61.8%.

#### 2.2 固相垃圾中氮组分的变化

图 3 显示了 R1、R2 和 R3 固相垃圾中 TN。的

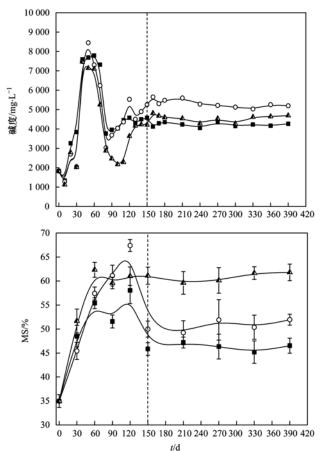


图 2 反应器中内环境指标随时间的变化

Fig. 2 Variations of interior environment indicators over time in reactors

含量在整个 390 d 内不断下降,分别由初始值 19 200.0  $mg \cdot kg^{-1}$ 下降到4 000.0、3 400.0和2 200.0  $mg \cdot kg^{-1}$ . ( $NH_4^+ \cdot N$ )。的含量也分别由 89.7  $mg \cdot kg^{-1}$ . 持续下降到和 46.3、35.8 和 28.9  $mg \cdot kg^{-1}$ . 由于垃圾装填过程中携带了一定量氧气进入反应器中,因此可以发现 60 d 前 R1、R2 和 R3中( $NO_3^- \cdot N$ )。的含量分别从 51.3  $mg \cdot kg^{-1}$ 升高到 82.6、87.4 和 85.2  $mg \cdot kg^{-1}$ ,此后厌氧环境开始形成,( $NO_3^- \cdot N$ )。经反硝化作用转化,含量不断降低,实验结束时分别为 31.6、28.6 和 23.9  $mg \cdot kg^{-1}$ .

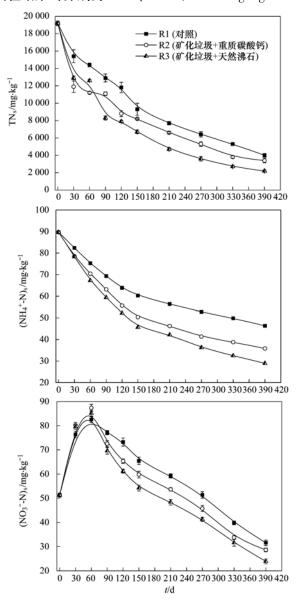


图 3 固相垃圾中氮组分随时间的变化

Fig. 3 Variations of nitrogen compounds over time in solid waste

#### 2.3 渗滤液中氮组分的变化

3 组模拟反应器渗滤液中氮组分的变化趋势如图 4 所示. 在 1~60 d 内, 垃圾中易降解含氮有机物

被好氧微生物快速降解转化进入渗滤液,使得 R1、R2 和 R3 渗滤液中 TN 的浓度分别从初始 812.0 mg·L<sup>-1</sup> 上升到最大值5 331.0、4 497.0 和3 957.0 mg·L<sup>-1</sup>. 60~120 d,由于反硝化等作用,渗滤液中 TN 持续下降,120 d 后 TN 浓度变化趋于稳定,实验结束时,分别为3 261.0、2 745.0和2 465.0 mg·L<sup>-1</sup>.

R1、R2 和 R3 渗滤液中NH4-N浓度均呈现波 动上升趋势,实验结束时,分别从初始的381.7 mg·L<sup>-1</sup> 上升到1 914.6、2 152.4和1 541.6 mg·L<sup>-1</sup>. 在 1~40 d 内, R1、R2 和 R3 渗滤液中NO<sub>3</sub>-N浓度 分别从初始值 1.0 mg·L<sup>-1</sup> 上升到 252.9、214.0 和 180.6 mg·L<sup>-1</sup>,这主要由硝化细菌利用反应器中残 留氧气将NH<sub>4</sub> -N转化而来<sup>[19]</sup>. 40 d 后,NO<sub>3</sub> -N波动 下降,实验结束时,R1、R2和R3中渗滤液的NO,-N 浓度分别为 32. 2、18. 6 和 10. 4 mg·L<sup>-1</sup>. NO<sub>2</sub>-N的 稳定性不如NO; -N,能优先被反硝化菌利用,因此其 浓度远低于NO; -N. 1~50 d, NO; -N在3组反应器 中的浓度分别从 0.16 mg·L<sup>-1</sup> 上升到 4.72、5.78 和 5.90 mg·L<sup>-1</sup>;50~80 d,NO<sub>2</sub>-N被反硝化细菌快 速利用,其浓度又迅速下降到 0.21、0.20 和 0.18 mg·L-1;80 d 后,3 个反应器渗滤液中NO2-N趋于 稳定,且浓度极小.

#### 3 讨论

#### 3.1 内环境调节层对内环境的调节作用

150 d 后, R1、R2 和 R3 内环境参数稳定, 反应 器进入稳定的厌氧状态,对比发现 pH(R2) > pH (R3) > pH(R1)、碱度(R2) > 碱度(R3) > 碱度 (R1), Eh(R2) < Eh(R3) < Eh(R1), MS(R3) >MS(R2) > MS(R1). pH 和碱度是厌氧生物反应器 填埋场内环境的重要参数,碱度还是微生物生长所 必须的物质[20],其影响填埋场内微生物的生长状 态,进而影响氮的降解、迁移和转化[21]. 研究表 明,矿化垃圾具有较好的酸性有机物缓冲能力[22], 能提供一定的碱度<sup>[23]</sup>,因此 R2 和 R3 中 pH、碱度 均高于 R1. 另外重质碳酸钙本来就是一种碱性物 质,其缓慢溶解后能直接中和有机酸,提高反应器 的 pH 和碱度,因此 R2 中 pH 和碱度最高,最适于 微生物的生长. 厌氧微生物对 Eh 较敏感,特别是 产甲烷菌是严格厌氧微生物<sup>[24]</sup>,因此 Eh 的高低, 决定着微生物的生长状态和垃圾降解的速度[25]. 在实验运行初期,矿化垃圾和重质碳酸钙能接种 微生物,优化反应器 pH, 从而使为好氧微生物能

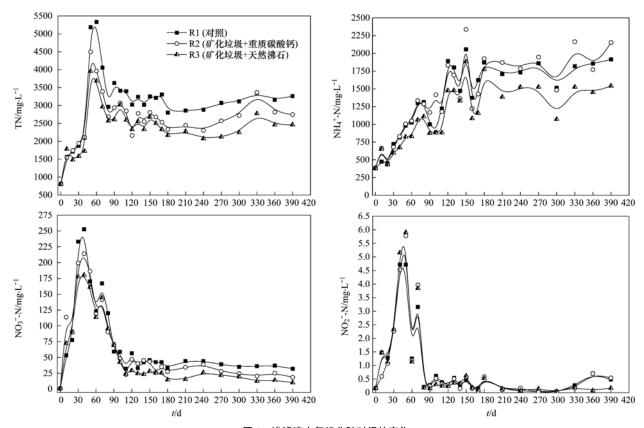


图 4 渗滤液中氮组分随时间的变化

Fig. 4 Variations of nitrogen compounds over time in leachate

迅速利用垃圾装填过程中残留的氧气,因此 R2 最快进入厌氧环境,并持续保持 Eh 最低. 此外,厌氧反应器填埋场的核心就是利用渗滤液循环技术[26],增加垃圾体的含水率,从而为微生物活动提供良好的环境[27]. R2 和 R3 中 MS 能长期保持在50%~60%的有利范围[28],这可能与内环境调节层的添加使布水更加均匀和垃圾随着降解过程的持水能力变化有关系. 综合考虑 pH、碱度、Eh 和MS 的变化,可以认为矿化垃圾+重质碳酸钙、矿化垃圾+天然沸石这两种内环境调节层均能长期优化反应器的内环境,为微生物的生长提供较有利的条件,从而促进垃圾和渗滤液中氮组分的降解转化.

#### 3.2 内环境调节层对氮转化去除的作用

R1、R2和R3中固相垃圾TN<sub>s</sub>、(NH<sub>4</sub>+-N)<sub>s</sub>和(NO<sub>3</sub>-N)<sub>s</sub>均持续下降,其降解转化率分别为79.2%、82.3%、88.5%,48.3%、60.1%、67.7%和38.5%、44.2%、53.4%。固相垃圾氮降解转化率R3>R2>R1,说明矿化垃圾+天然沸石、矿化垃圾+重质碳酸钙组成的内环境调节层有利于促进固相垃圾中氮的迁移转化。矿化垃圾含有丰富的微生物 $^{[29]}$ 和有机酸缓冲物质 $^{[30]}$ ,其能促进微生物对垃

圾中含氮有机物的降解. 同时,天然沸石能吸附氨 氮[31~33],促进氨氮的转化[34,35],因此,矿化垃圾+ 天然沸石组成的内环境调节层能更好地促进固相垃 圾中氮的降解转化. 渗滤液中 TN(R3) < TN(R2)  $< TN(R1), NO_3^--N(R3) < NO_3^--N(R2) < NO_3^--N$ (R1),可见添加矿化垃圾+重质碳酸钙、矿化垃圾 +天然沸石这2种内环境调节层后,确实能通过优 化填埋场内 pH、碱度、Eh 和 MS 等内环境参数,接 种大量微生物来促进微生物对渗滤液中TN、 NO<sub>3</sub>-N的降解转化. 另一方面,渗滤液中NH₄-N (R3) < NH<sub>4</sub><sup>+</sup>-N(R1) < NH<sub>4</sub><sup>+</sup>-N(R2),可以看出矿 化垃圾+天然沸石组成的调节层能较好地控制渗 滤液中氨氮的累积,促进渗滤液中氨氮的降解转 化,而矿化垃圾+重质碳酸钙则相反,这一差别主 要是因为天然沸石具有吸附、固定和促进氨氮转 化的能力[36~38],而重质碳酸钙虽然能提供碱度, 缓解有机酸的累积,一定程度上促进垃圾的降 解[39],但是不能促进渗滤液中氨氮的转化. 综合 固相垃圾和渗滤液中氮组分的变化可以发现在矿 化垃圾和天然沸石的双重作用下,反应器内环境 得到优化,氨氮累积得到缓解,从而有利于微生物 对含氮物质的降解转化.

#### 4 结论

- (1) 矿化垃圾+重质碳酸钙、矿化垃圾+天然 沸石这2种内环境调节层均能在一定程度上优化反 应器的内环境,为微生物降解转化氮提供有利条件.
- (2) 矿化垃圾 + 天然沸石组成的内环境调节层能促进垃圾和渗滤液中氮组分的降解转化,在一定程度上控制渗滤液循环造成的氨氮累积效应.

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