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## 大块纳米气泡在废水处理中去除有机污染物的作用

安博快递 体积 11、文章编号： 96 (2021) 引用本文

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### 抽象的

多种有机污染物的产生，使废水处理变得复杂；因此，寻找可持续、有效的处理技术引起了人们的广泛关注。近年来，块状纳米气泡具有不同于微气泡的非凡特性，包括稳定性高、在水中停留时间长、比表面积大、气体传输效率和界面电位高、以及产生自由基的能力等。废水处理具有诱人的技术优势和广阔的应用前景。本文详细总结了块状纳米气泡的基本特征，系统地讨论了其在有机废水处理中的实施途径和机制的最新研究成果，包括改进气浮工艺、增加水体曝气以推广好氧生物技术，包括生物活性炭、活性污泥和膜生物反应器，并产生氧化有机化合物的活性自由基。最后，分析了目前体纳米气泡的技术难点，并提出了未来体纳米气泡技术研究的重点领域。

### 关键点

- 散装纳米气泡可以提高浮选效率，去除油脂等有机物。
- 微生物生物降解过程的氧气传质速率加快。
- 大块纳米气泡的崩溃形成自由基，促进氧化过程。

### 介绍

纳米气泡被认为是存在于固液界面或分散在液体介质中的直径小于1 $\mu$ m的球形包裹 (Alheshibri等人, 2016 )。根据形态和位置可分为固液界面纳米气泡 (表面纳米气泡) 和块状纳米气泡。表面纳米气泡是吸附在固液界面上的薄饼状气泡，其高度为20-30 nm，曲率半径为100 nm量级 (Tyrrell and Attard 2001)，而块状纳米气泡的形态显示为一个完整的球体，其纳米级尺寸小于 1  $\mu$ m。这两类纳米气泡的研究历史和研究方法都是相对独立的系统。形态的差异也导致了两类纳米气泡的制备方法和应用存在很大差异。目前，随着研究的进展，

包括直接浸泡、温度变化、电化学反应、乙醇-水交换等多种方法已被应用于生成表面纳米气泡(Qiu et al. 2017)，用于表面清洁、和微型管道设计；然而这些技术还没有得到很好的发展。通过电解法 (Hao et al. 2020)、多孔膜法 (Ulatowski et al. 2019)、超声空化法 (Ulatowski et al. 2019) 和水动力空化法 (Oliveira et al. 2018) 产生块状纳米气泡。另一方面，在水处理中发挥着主导作用。

与普通微气泡（直径小于50 $\mu\text{m}$ ）或传统大气泡（直径大于1mm）不同，块状纳米气泡表现出独特的物理性质，在生物修复、农业和生物医学领域的应用受到广泛关注，尤其显示出巨大的技术优势和优势。废水处理领域的前景。由于水资源短缺、水污染严重，废水处理已成为社会发展的重点。近年来，大块纳米气泡已广泛应用于含油废水、焦炭废水、印染废水 (Bui和 Han, 2020)、腈纶废水 (Zheng等, 2015) 和重金属离子废水 (Kyzas等, 2015) 的处理。2019) 等。随着散装纳米气泡的巨大应用，预计散装纳米气泡可以成为增强水处理技术的潜在驱动力，从而在相对于表面纳米气泡的工业应用中发挥重要作用。有机物，如味道和气味化合物，极其复杂，被认为具有泥土霉味(Zhou et al. 2021)，在很大程度上影响着人们的生活(Cen et al. 2020)，并增加了处理过程的难度和成本。由于大体积纳米气泡技术环境友好、无二次污染，处理废水中的有机污染物，人们进行了大量的研究探索。然而，可以发现，虽然浮选过程是工厂水处理的常见步骤，但以往关于基于块体纳米气泡的浮选方法的大量研究集中在金属离子的去除上，很少有研究关注有机物。，值得深入关注。另一方面，好氧生物方法广泛应用于水处理，但很少有综述特别强调通过大体积纳米气泡与好氧生物技术相结合来提高有机物的生物降解效率，同时阐明其机理，以促进更深入的研究和研究。应用。

本研究的主要目的是全面表征大块纳米气泡的基本性质，并深入了解该技术在处理含有机污染物的废水方面的应用现状和机理，重点是与大块纳米气泡相结合的好氧生物方法。最后，同时探讨了废水处理中有机污染物尚待解决的未知问题以及该领域的未来展望。

## 基本特性

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### 高稳定性

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对块状纳米气泡的最早研究可以在 Johnson 和 Cooke 发表的一篇文章中找到 (1981)。他们报告说，海水中的剪切力可以产生充满气体的纳米尺寸气泡，由于天然存在的表面活性剂形成的表面膜的存在，这些气泡可以稳定长达 24 小时。然而，根据经典热力学，液体中球形气泡中存在的附加压力可以用杨-拉普拉斯公式方程[方程1]来表征。( 1 ); 刘和曹2016 ]：

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$$\Delta P = 2\sigma/r$$

(1)

式中： $\Delta P$ 为气泡内外压强差，也称拉普拉斯压强； $\sigma$ 是气泡/液体界面的表面张力； $r$ 为气泡半径。当气泡收缩时，内部压力会相应增加，并且由于气体倾向于溶解到液体中，这会导致气泡破裂。例如，半径为100 nm的气泡内压高达 $1.5 \times 10^6$  Pa；当表面张力为72 m N/m、液体压

力为 $10^5$  Pa时(Gurung et al. 2016 ), 计算结果表明直径为100 nm的纳米气泡只能在水中存在10  $\mu$ s(Ljunggren and Eriksson 1997); Nirmalkar 等人, (2018) )。然而, 在近十年来, 块状纳米气泡得到了广泛的研究(Alheshibri et al. 2016 ), 随着激光衍射技术等纳米气泡跟踪技术和其他检测方法的进步, 更多的证据证实了纳米气泡可以稳定存在。长时间在液体介质中。这可以归因于它们的小尺寸和高界面电势, 如下所述。

普通大气泡产生后, 会借助浮力快速向上移动并消失在水面上 (Demangeat 2015), 而大块纳米气泡可以在水中停留数天甚至数月 (Azevedo et al. 2016; Liu et al. 2013) )。气泡的上升速度与其尺寸密切相关, 随着气泡半径的减小, 上升速度和停留时间会分别变慢和变长。一项计算表明, 半径为 50 nm 的纳米气泡的上升速度可低至 2.7 nm/s (Alheshibri et al. 2016 )。浮力对纳米气泡的影响可以忽略不计; 因此, 大块纳米气泡在水中的寿命比大气泡更长。尼尔马尔卡等人。( 2018 ) 报道称, 大块纳米气泡悬浮液在几个月内保持稳定, 在此期间平均直径保持恒定, 表明不存在显着的气泡聚结、气泡破裂或奥斯特瓦尔德熟化效应 (Nirmalkar et al. 2018 )。顺便说一句, Weijs 等人。( 2012 ) 利用分子动力学深入了解了稳定性的原因, 表明大块纳米气泡簇中的纳米气泡通过屏蔽效应相互保护免受扩散 (Weijs 等人, 2012 )。

此外, 块状纳米气泡具有高界面电势。由气泡表面电荷引起的电势差通常用 zeta 电位来表征, 这是悬浮颗粒表现出的物理特性, 用于测量颗粒和气泡之间的静电排斥或吸引力

(Parmar 和 Majumder 2013 )。Zeta 电位可有效预测胶体系统的长期稳定性。如果悬浮液中的所有颗粒均表现出较高的 zeta 电位 (负或正), 则它们往往会长期稳定; 否则, 它们很容易聚集和合并 (Gurung et al. 2016) )。吸附有带负电的 OH<sup>-</sup>离子的纳米气泡表面称为表面带电离子层, 它会吸引带正电的 H<sub>3</sub>O<sup>+</sup>离子, 从而形成双电层 (Boshenyatov et al. 2019) )。相邻气泡之间双电层重叠产生的静电排斥力提供了对气泡合并的抵抗力; 因此, 纳米气泡表现出意想不到的稳定性和耐久性 (Calgaroto et al. 2014) )。根据对传统杨-拉普拉斯方程进行修改的理论模型, 如方程[Eq. ( 2 )——方程。 ( 5 ) ; Satpute 和 Earthman 2021 ] :

$$p_{\text{我nt}} + p_{\text{rep}} = p_{\text{eXt}} + p_{\text{st}}$$

(2)

$$\frac{1}{r}$$

$$p_{\text{rep}} = 3\epsilon_0 \epsilon \sigma^2$$

(3)

$$\frac{1}{r}$$

$$\sigma = \epsilon_0 \epsilon \Psi \lambda$$

(4)

$$\lambda = \epsilon_0 \epsilon k_B T / (2 N_A q^2)$$

(5)

其中  $p_{\text{int}}$  为内部压力,  $p_{\text{rep}}$  为气泡表面吸附的负离子之间的斥力产生的麦克斯韦压力,  $p_{\text{ext}}$  为外部压力,  $p_{\text{st}}$  为表面张力产生的压力,  $k_e$  为库仑压力流体常数 ( $1.1 \times 10^8 \text{ N m}^2 \text{ C}^{-2}$ ),  $\sigma$  是气泡的表面电荷密度, 使用来自双电层 Debye-Hückel 理论的方程根据 zeta 电位计算,  $\epsilon_0$  是介电常数自由空间的介电常数,  $\epsilon$  是流体的介电常数,  $\lambda$  是德拜长度,  $\xi$  是 zeta 电位,  $I$  是稳定离子的体积浓度, 即  $\text{OH}^-$  在 pH 下为  $10^{-4} \text{ mol m}^{-3}$ ,  $T$  为温度,  $k_B$  为玻尔兹曼常数,  $N_A$  为阿伏伽德罗数。

该模型成功地阐明了表面上的  $\text{OH}^-$  之间的排斥力足以最终平衡纳米气泡尺寸下的表面张力, 从而使纳米气泡表现出极大的稳定性。如以往文献所示, zeta 电位值随介质 pH 值的变化而变化, 表现出随着溶液 pH 值的增加, zeta 电位的负性增大的趋势, 导致气泡表面电荷增加, 由于静电斥力使气泡更加稳定 (Meegoda 等人, 2018)。在较宽的溶液 pH 范围内, 纳米气泡通常表现出较高的负 Zeta 电位, 这是由于气泡的气水界面上的氢氧根离子 ( $\text{OH}^-$ ) 相对于氢离子 ( $\text{H}^+$ ) 过量, 但在酸性环境中形成的纳米气泡总是表现出较低的 Zeta 电位。与中性或碱性条件相比, 由于需要缺乏  $\text{OH}^-$  来稳定双电层 (Nirmalkar et al. 2018), 因此在 pH 3 以下的强酸性介质中产生的纳米气泡的 zeta 电位往往为正且较低, 导致气泡合并的可能性更高 (Zhang et al. 2020)。30 mV 的 zeta 电位大小被认为是能够产生巨大斥力并有助于纳米气泡持久存在的临界值 (Ushikubo et al. 2010)。

截至目前, 已经提出了各种理论模型来解释块状纳米气泡的稳定机制, 包括静电斥力模型 (Yasui et al. 2018)、屏蔽效应理论 (Weijs et al. 2012)、表皮模型 (Yount 1979)、粒子缝隙模型 (Atchley and Prosperetti 1989)、动态平衡模型 (Yasui et al. 2016)。然而, 块状纳米气泡的意外稳定性需要进一步研究才能在未来获得统一的结论。

## 气液传质效率高

水中的块状纳米气泡周围存在气液界面, 导致气泡压缩, 从而在气泡上升到水面的过程中不断收缩并表现出自加压效应。气体从高压区向低压区扩散的速率与压力梯度成正比 (Li et al., 2014), 从气泡内部向周围液体的传质速率随着压力梯度的增大而加快。气泡缩小。同样, 块状纳米气泡显示出较大的比表面积, 因为气泡的表面积与气泡的半径成反比 (Kim et al. 2018)。例如, 1000 个直径为 5 mm 的气泡体积的总表面积为  $0.0785 \text{ m}^2$  (总体积 =  $6.54 \times 10^{-5} \text{ m}^3$ ), 而相同体积的直径为 100 nm 的气泡的总表面积为  $0.0785 \text{ m}^2$  (总体积 =  $6.54 \times 10^{-5} \text{ m}^3$ ) (体积/气泡 =  $5.24 \times 10^{-22} \text{ m}^3$ ), 当气泡数量等于  $1.25 \times 10^{17}$  时, 总表面积更高, 达到  $3925 \text{ m}^2$  (Batagoda et al. 2019)。如前所述, 更多的气体可以通过气泡界面溶解到水中, 并且由于大体积纳米气泡的自增压效应、长的停留时间和大的比表面积, 有效地提高了气体转移到液相的效率。直到气泡的内部压力达到一定的极限值时, 气泡才会破裂并消失。

## 产生自由基的能力

多项研究表明，强超声波和水动力空化作用会导致纳米气泡破裂并产生自由基 (Masuda et al. 2015)。在纳米气泡收缩的过程中，双电层中积累的电荷密度迅速增加。由于气泡破裂瞬间气液界面发生剧烈变化，聚集在界面上的高浓度离子瞬间释放出积聚的化学能。同时，当被动态刺激强力压缩时，被捕获的水分子会受到极高的温度和压力 (Krishnan et al. 2006)，从而导致裂解，产生各种活性氧 (ROS)，包括羟基自由基 ( $\cdot\text{OH}$ )、超氧阴离子自由基 ( $\text{O}_2\cdot^-$ ) 和单线态氧 ( $^1\text{O}_2$ ) (Atkinson 等人, 2019 年; Liu 等人, 2015 年; Lyu 等人, 2019 年)。一些研究已经应用电子自旋共振或自由基抑制剂的作用来验证它们的存在。米凯利迪等人。(2020)利用基于5,5-二甲基-1-吡咯啉N-氧化物(DMPO)自旋捕获的电子顺磁共振定量分析空气-纳米泡和氧气-纳米泡系统中的自由基。在 DMPO 浓度为 100 mM 时，根据校准曲线，推断空气-纳米气泡系统中的总自旋浓度约为 1.5  $\mu\text{M}$  (Michailidi 等人, 2020)。王等人。(2020)使用 2-丙醇、苯醌 (BQ) 和  $\text{NaN}_3$  作为清除剂分别捕获  $\cdot\text{OH}$ 、 $\text{O}_2\cdot^-$  和  $^1\text{O}_2$ ，以确定诱导土霉素光降解的可能反应物种，并得出结论： $\cdot\text{OH}$ 、 $\text{O}_2\cdot^-$  和  $^1\text{O}_2$  可以共存于纳米气泡溶液中，而  $\cdot\text{OH}$  在氧化反应中起主导作用 (Wang et al. 2020)。刘等人。(2016)使用灵敏的荧光探针 APF 证实了氧纳米气泡水中  $\cdot\text{OH}$  的存在，并以亚微摩尔数量级量化了其含量 (Liu et al. 2016)。这些活性氧的强大氧化功能逐渐被考虑应用于高级氧化工艺中，以处理废水处理中的持久性有机污染物（如多氯联苯和酚类卤代化合物），也可用于病原体的消毒。

正是由于上述独特的优点，块状纳米气泡在废水处理领域受到越来越多的关注，并在改进去除有机物的常规工艺方面显示出广阔的潜力。根据以往的研究，可以得出结论，大块纳米气泡可以通过多种途径去除水中的有机污染物，大致包括：(1) 改进气浮工艺，去除脂肪、油类、低密度有机悬浮固体和胶体；(2) 促进水中曝气，以加强由生物活性炭过滤器、活性污泥和膜生物反应器组成的常规生物降解过程；(3) 产生自由基来氧化和降解难以生物降解的有机化合物，这将在接下来的章节中深入阐明。

## 浮选机理及改进

浮选被认为是去除含有脂肪、油类、低密度有机悬浮固体和胶体的悬浮液的最可靠和实用的方法 (Colic 等人, 2007)。广泛用于去除废水处理系统预处理单元中的各种有机物，以减轻后续处理工艺的负荷。分离机理归因于气泡吸附在细小的悬浮颗粒表面，从而形成气泡-颗粒聚集体，从而显着降低污染物的重力。因此，颗粒更容易上升到水面，并被刮掉，实现固-液或液-液分离，达到去除目的 (Zimmerman et al. 2011)。气泡-颗粒聚集体的形成取决于多种因素，包括气泡/颗粒尺寸之比、浓度等，其中气体分散参数被认为是分离中最重要的参数。传统的气浮方法对于粒径范围较窄的颗粒是有效的，当超出最佳粒径范围时，气浮效率显着降低 (Fan et al. 2010)。低浮选率归因于气泡-颗粒碰撞的可能性，并且当颗粒尺寸较大或较小时，气泡-颗粒的分离分别较低和较高 (Tao 2005)。实验证据表明，纳米气泡的存在拓宽了浮选粒度范围，增加了颗粒的表面疏水性，并提高了泡沫浮选效率 (Fan et al. 2010; Sobhy 和 Tao 2018; Tao 和 Sobhy 2019)。浮选的总体效率由三个连续步骤决定：气泡-颗粒碰撞、附着和分离。各种模型和研究表明，减小气泡尺寸的需要与气泡与颗粒之间碰撞可能性的增加密切相关，并且由于尺寸小，可以大幅提高浮选在污染物分离过程中的效率。纳米气泡的存在以及相反表面电荷的存在，增加了粒子和气泡之间碰撞和附着的可能性 (Luttrell 和 Yoon 1992; Xiao 等人 2018)。米什丘克 (2005) 对含纳米气泡的系统中颗粒间相互作用进行了理

论分析，并报道纳米气泡的出现和消失影响了宏观体之间液层的机械和其他特性，包括密度、液体流动性和薄层破裂条件非均质薄膜。他们的结果进一步表明，两个相似颗粒之间间隙中的纳米气泡增加了它们的吸引力，从而改善了聚集形成 (Mishchuk 2005)。纳扎里等人。(2019)研究了气泡尺寸分布对粗石英颗粒浮选行为的影响，发现直径为171 nm的纳米气泡存在时，石英颗粒表面十二烷基胺的吸附量大于在存在尺寸为110和293 nm的纳米气泡以及普通气泡的情况下 (Nazari 等人, 2019)。蔡等人。(2007)研究了纳米气泡浮选技术 (NBFT) 结合混凝工艺处理实验室规模和中试规模的化学机械抛光 (CMP) 废水，他们的实验结果表明，与传统的混凝/絮凝工艺相比，NBFT结合混凝技术使废水除浊效率提高了40% (Tsai et al. 2007)。此外，卡尔加罗托等人。(2016)通过纳米气泡浮选去除癸基三甲醚胺。纳米气泡在pH 10.8时粘附在胺形成的胺沉淀上，通过浮选分离夹带在絮凝体内部，胺的去除率达到80%，表明纳米气泡在浮选技术中的应用具有处理残留含胺废水的潜力 (卡尔加罗托等人, 2016)。此外，周等人。(2019)研究了十二胺 (DDA) 存在下白云母表面水动力空化产生的块状纳米气泡的吸附行为 (Zhou et al. 2019)。结果表明，纳米气泡大部分被 DDA 覆盖，并且确实被吸附在白云母表面上，可能通过三相线钉扎而稳定，从而导致白云母浮选性能的广泛增强。埃切帕雷等人。(2017)研究了微气泡 (30~40  $\mu\text{m}$ ) 和纳米气泡 (150~350 nm) 对盐水中乳化原油的分离；孤立纳米气泡的输入 ( $3 \times 10^8$ 、3mg/L Dismulgan絮凝后，增加了团聚体的疏水性，促进了气泡与油性絮体的粘附，浮选去除率分别由73%提高到84%和92%提高到95%，分别 (Etchepare 等人, 2017)。基于上述研究，散装纳米气泡的应用提高了浮选效率，从而更好地去除油脂等物质。

## 好氧生物处理

对于可生物降解性好的有机废水，生物处理方法，包括生物活性炭过滤、生物活性污泥、膜生物反应器 (MBR) 等，与电化学法等其他物理化学技术或需要额外投入的物理化学技术相比，能耗低、环境友好。添加化学品，已成为一种优惠处理技术。生物法是利用微生物的代谢作用，分解水中有毒有害化学物质和某些超标成分。好氧生物过程的主要限制和成本来自曝气、污泥处理和膜污染。

氧气在好氧生物的新陈代谢中起着至关重要的作用，是污染物氧化降解的生化反应底物，它直接关系到废水处理过程中可生物降解有机物的去除效果，不仅在生物活性炭过滤中，而且在活性炭过滤中也是如此。污泥和膜生物反应器。因此，需要加快氧气的传质速率，以提高水中的溶解氧浓度，促进有机物的降解。通气在氧气输送中的重要作用已得到广泛认可。传统的机械曝气器或扩散器消耗大量电能，机械维护成本较高 (El-Zahaby and El-Gendy 2016)，而氧气传输效率极不理想，不利于节能降耗。纳米气泡的浮力可以忽略不计，并且在水中的停留时间较长，因此可以更有效地扩散氧气。张等人。(2018)研究了湖泊沉积物的修复，其结果表明，通过将材料暴露于氧气过饱和环境条件下，然后在颗粒微孔中重新负载  $\text{O}_2$  来实现含氧纳米气泡改性沸石/土壤的供应，以达到缺氧的湖泊沉积物可以将溶解氧浓度维持在大于6毫克/升的状态6个月以上 (Zhang et al. 2018)。李等人。(2013)比较了微纳米气泡对地下水生物修复的影响，发现微纳米气泡中氧气的传质速率比大气泡快125倍，溶解氧比后者持久16倍。此外，研究探讨了纳米气泡对好氧生物降解水中污染物的影响，结果表明，纳米气泡曝气合成废水处理装置的氧气利用率和体积传质系数几乎是传统气泡曝气装置的两倍，并且纳米气泡曝气装置中有机物降解的保留时间不到传统系统中使用的保留时间的一半。基于这些结果，有人

提出纳米气泡可以促进好氧生物降解过程来净化受污染的地下水 (Li et al. 2013)。最近, Xiao 和 Xu (2020) 尝试在好氧生物膜系统中提供更多的氧气, 以提高其性能并降低废水处理的曝气成本 (Xiao 和 Xu 2020)。实验表明, 纳米气泡具有较大气泡优越的供氧能力和1.5倍的传氧效率, 促进了生物膜的生长, 实现了更好的化学需氧量和氨氮去除效率。此外, 纳米气泡曝气可节省约80%的能源。拉梅什库马尔等人。(2019) 在自来水、池塘水、生活废水和工业废水四个系统中产生纳米气泡, 探讨纳米气泡对水处理效率的影响。令人惊讶的是, 所有四个系统的水处理后溶解氧浓度都急剧增加, 其中溶解氧分别增加到自来水和池塘水的2倍和1.5倍。同时, 生活污水中化学需氧量由处理前的399mg/L降至检出限以下4mg/L, 工业废水中化学需氧量由处理前的4520mg/L降至124mg/L, 效果显著。纳米气泡在水处理中的应用 (Rameshkumar 等人, 2019)。如上所述, 毫无疑问, 纳米气泡在增强通气和微生物生长以进行生物降解方面具有广阔的应用前景。

## 生物活性炭

吸附辅以生物降解被认为是生物活性炭 (BAC) 过滤过程中去除溶解有机化学品 (DOC) 的主要机制。活性炭广泛应用于去除或控制令人不快的味道和气味问题以及水中的氯仿和三卤甲烷等有机污染物, 以及各种污染物, 包括芳香族化合物、碳氢化合物、清洁剂、可溶性染料和酚类, 因为它具有高孔隙率和广泛的内表面积 (Rattier 等人, 2012)。随着时间的推移, 活性炭的吸附位点被有机物饱和, 导致最初的有效性丧失, 同时由于细菌栖息在碳的孔隙中形成微生物群落, 从而形成具有生物活性的生物膜。这确保了碳通过代谢活动和该阶段对去除作用的生物氧化反应不断去除DOC。基于此, BAC过滤器的生物降解性应受到重视。如前所述, 纳米气泡显著提高了用于微生物生物降解的好氧生物膜系统中的通气和氧气传质效率。值得注意的是, 块状纳米气泡不仅具有生物降解作用, 而且还具有吸附作用。基扎斯等人。(2019) 研究了纳米气泡对活性炭吸附水中铅离子的影响, 并验证了纳米气泡存在或不存在时吸附能力大致相似; 然而, 吸附过程令人印象深刻地加速了 366%, 预计这将减少吸附平衡时间, 从而增加吸附-解吸循环; 从这些结果得出的机制是, 纳米气泡将带电粒子吸引到其界面上, 从而有助于铅离子扩散和渗透到活性炭孔中 (Kyzas 等人, 2019)。然而, 块状纳米气泡对活性炭吸附有机化合物的影响及其潜在机制仍需探索。

## 活性污泥

活性污泥系统由生物降解和沉淀过程组成, 分别在曝气池和沉淀池中进行。曝气池内充满充分混合的曝气废水, 长时间内实现溶解和胶体底物生物转化为稳定的新生物质细胞。这个过程是由多种微生物在氧气存在下完成的。肖等人评估了纳米气泡曝气对两种最常见的微生物聚集体 (活性污泥和生物膜) 的影响。(2021), 他们的结果表明, 纳米气泡的利用成功地为微生物聚集体提供了额外的氧气, 并实现了总氮去除率的10.58%的增量。此外, 微生物聚集体的结构得到改善, 表明随着生物膜厚度和活性污泥絮体尺寸的增加, 细胞外蛋白质和多糖在生物膜和活性污泥中分别增加到最高水平, 分别为3.40倍和1.70倍。该结果表明, 纳米气泡通过加速微生物聚集体的结构发育来优化功能微生物的分布及其代谢途径, 同时导致微生物聚集体的适应性过程, 特别是对于活性污泥, 当在纳米气泡添加比例较低的情况下, 当比例超过 50% 时, 结果变为正值 (Xiao et al. 2021)。污泥分解和溶解是活性污泥工艺长期运行中的棘手问题。污泥臭氧化对减少废污泥体积具有良好的效果。污泥絮体由复杂的微生物群落组成, 包括细菌、原生动物和后生动物, 并与细胞外聚合物 (如蛋白质、碳水化合物、腐殖酸和其他有机

分子)结合形成称为絮体的微生物聚集体 (Semblante 等) (2017)。在臭氧与细菌细胞反应之前,它首先被溶解的有机物和细胞外聚合物消耗。污泥絮体的尺寸从几十微米到几百微米不等,间隙约为20 $\mu\text{m}$ 。作为臭氧载体,传统的微气泡太大,无法穿透污泥絮体的间隙,限制了臭氧减少污泥体积的能力,而纳米气泡的直径比间隙小两个数量级。因此,避免了絮体外部的臭氧消耗,而臭氧在杀死细菌方面起着关键作用。桥本等人。(2021年)以污泥中细菌的死亡率作为污泥减量效果的指标,并比较了纳米气泡和微气泡为污泥减量提供臭氧的能力;结果表明,在由纳米气泡携带臭氧的系统中,达到80%死亡率所需的臭氧剂量为15 mg- $\text{O}_3$ /g-MLSS,而当臭氧由纳米气泡携带时,所需臭氧剂量为25 mg- $\text{O}_3$ /g-MLSS。螺旋状液体型微气泡。此外,在臭氧消耗率相同的情况下,从污泥絮体表面到内部的死细胞层深度,在具有纳米气泡的系统中比在没有纳米气泡的系统中更大,这表明纳米气泡臭氧载体提高了污泥的效率减少 (Hashimoto 等人, 2021)。

## 膜生物反应器

好氧MBR利用好氧消化与膜过滤相结合,被认为是传统活性污泥处理的替代品,其中二级澄清器被膜取代,以减轻不需要的生物膜形成的沉降问题。其优点是出水水质好、占地面积小、减少土地使用总量、较高的容积负荷率、污泥产量少等, Terfasa (2017) 计算了纳米气泡系统中生物质的衰减率值,发现:它几乎比传统气泡支持系统快三倍,表明纳米气泡减少了好氧处理中过量污泥的产生 (Terfasa 2017)。然而,除了需要改善曝气以提高消化效率外,MBR还面临有机废水处理中的重膜生物污垢问题,这阻碍了操作灵活性并导致维护成本显著增加 (Iorhemen 等, 2016)。因此,必须考虑MBR中可持续的膜污染缓解策略。Terfasa (2017) 在传统系统和纳米气泡支持系统中对膜过滤进行了测试,结果表明,两种系统的膜过滤流速均随着时间的推移而下降,但传统系统下降得更快。此外,纳米气泡支持的系统的累积过滤体积比传统系统增加得更快,这可以归因于由于纳米气泡的存在而堵塞膜孔的污泥的减少 (Terfasa 2017)。加迪姆哈尼等人。(2016)尝试通过使用腐植酸作为模拟有机污染物,将空气纳米气泡送入膜单元来清洁污染的膜表面;结果表明,渗透通量恢复到初始水平,表明纳米气泡显著疏通了膜的孔隙,这归因于 $\cdot\text{OH}$ 与有机污染物结合并分解有机物 (Ghadimkhani et al. 2016)。

大体积纳米气泡集成好氧生物技术的应用能够促进有机物的生物降解,但尚未实现工业规模应用,这是未来研究的重要课题。此外,还应考虑纳米气泡的布朗运动以及纳米气泡破裂产生的高水平热和冲击波是否会对微生物细胞造成损害。

## 高级氧化

### 基于块状纳米气泡的高级氧化

如前所述,一些研究已经认识到块状纳米气泡能够产生 $\cdot\text{OH}$ ,其具有很强的氧化能力,并且在气泡破裂时对几乎所有种类的有机污染物都没有选择性,因此可以在处理含废水中发挥重要作用。通过夺氢、自由基-自由基反应、亲电加成和电子转移对常规生物处理具有抵抗力的有机化合物。传统上, $\cdot\text{OH}$ 是通过大量能量和化学密集型过程产生的,例如紫外线照射、过氧化氢氧化和芬顿氧化;因此,纳米气泡可以成为一种令人鼓舞的创新替代品。Bui和Han(2020)利用三种纳米气泡体系(纳米气泡、超声纳米气泡和纳米气泡/ $\text{H}_2\text{O}_2$ )降解深绿色Rit染料,并通

过检测甲磺酸谱验证了体系中 $\cdot\text{OH}$ 的产生。研究发现, 纳米气泡与 $\text{H}_2\text{O}_2$ 共存体系中生成的 $\cdot\text{OH}$ 会进一步与 $\text{H}_2\text{O}_2$ 反应生成 $\text{HO}_2\cdot$ 和 $\text{O}_2\cdot$ , 破坏染料分子, 获得良好的脱色率; 在该系统中, 60分钟后Rit染料的脱色率达到90%以上 (Bui和Han 2020)。此外, 除了活性物质的作用外, 还提出了静电吸引来驱动纳米气泡系统中Rit染料的脱色。结果证实, 当纳米气泡和染料溶液的电荷相反且纳米气泡的表面电荷大小与溶液中染料表面电荷大小几乎相似时, 两种条件下都可以提高除色效率。田崎等人。(2009)研究了8 W 低压汞灯在纳米气泡(直径720 nm)存在下对十二烷基苯磺酸钠(SDBS)分解的影响。使用带或不带纳米气泡的臭氧灯(185-254 nm)进行降解实验。实验结果表明, 在185-254 nm照射下, SDBS与氧纳米气泡结合时, 氧化和矿化率显著提高, 并且在集成纳米气泡/真空紫外(VUV)系统中SDBS被有效去除。SDBS后氧化率为99.8%, 24小时后总有机化合物(TOC)去除率达到76.8% (Tasaki等人, 2009)。王等人。(2020年)在光反应体系中添加氧纳米气泡, 提高可见光照射下土霉素的光降解效率。结果表明, 反应4 h后, 纳米氧气泡水中的光降解效率提高至60%, 当调节溶液pH至11.0时, 光降解效率甚至达到98%, 而普通曝气的光降解效率仅为40%左右。氧纳米气泡/光解过程的协同机制归因于氧纳米气泡向反应体系同时提供溶解氧和ROS, 在光反应过程中, 氧气的消耗促进了纳米气泡的破裂, 从而促进了ROS的生成, 其中 $\cdot\text{OH}$ 起到了重要作用。在土霉素的光降解中起主导作用 (Wang et al. 2020)。

## 基于臭氧纳米气泡的高级氧化

Advanced oxidation technology based on ozone nanobubbles can remove pollutants either through direct reaction of the compounds with ozone molecules or oxidation by  $\cdot\text{OH}$ , which is generated from the decomposition of ozone and bubble collapse. Both simultaneously promote the efficiency of oxidative degradation (Khuntia et al. 2015), although the latter plays a predominant role during the process because the standard oxidation potential of ozone (2.07 V) is lower than that of  $\cdot\text{OH}$  (2.80 V). In ozone-nanobubble based advanced oxidation processes (AOPs), enhancing both types of reaction mechanisms is essential to increase the overall oxidation efficiency. Different studies have reported that ozonation and ozone-based AOPs are affected by water properties, such as pH, concentration and type of organic matter. Regarding pH, it is known that the decomposition of ozone is accelerated when the pH or concentration of  $\text{OH}^-$  increase, which facilitates the chain reaction resulting in the formation of  $\cdot\text{OH}$  (Tomiyasu et al. 1985). Lucas et al. (2009) studied the degradation of organic matter involved in winery wastewater and found that the reduction rate of the chemical oxygen demand at alkaline and neutral pHs was more accelerated than at acidic pHs due to the formation of radical species from the decomposition of ozone (Lucas et al. 2009). Similarly, the pH of the medium strongly influences the stability of the bulk nanobubbles as discussed in previous section. Interestingly, the reaction between ozone and water molecules ( $\text{O}_3 + \text{H}_2\text{O} \rightarrow \text{O}_2 + \text{OH}^- + \text{OH}^\cdot$ ) increases  $\text{OH}^-$  ions, which are responsible for producing more negatively charged nanobubbles, resulting in considerable delivery efficiency of ozone molecules for the oxidation process. The restricted efficiency of ozone is due to its relatively low solubility and rapid decomposition in the aqueous phase. Nanobubbles as ozone delivery carrier enhance ozone concentration and prolong the ozone half-life in solution, leading to reduction of ozone wastage and increment reaction rates with

contaminants. Batagoda et al. (2019) compared the life of ozone bubbles produced by a regular diffuser and a nano-diffuser, showing that after 1 h of stabilisation, nano-ozone bubbles retained ozone in water for approximately four times longer than those produced using a regular diffuser (Batagoda et al. 2019). Hewage et al. (2020) designed an ultrasound and ozone nanobubble-coupled method to remediate the organic pollutants in sediments, p-terphenyl as simulated contaminant, of which the ultrasound was used to desorb the contaminants from sediments to suspension and then the pollutants were degraded by ozone. The results showed that ozone concentration delivered by nanobubbles was much higher than that with regular diffuser, concurrently, the reduction rate of ozone concentration of the former was far slower than the latter. The p-terphenyl treatment efficiency with ultrasound but without ozone nanobubbles was only 76.7%, while that of 91.5% was achieved when combined the two steps (Hewage et al. 2020). On the other hand, the possibility of improving the oxidative efficiency of an ozone-based water treatment unit depends largely on the  $\cdot\text{OH}$  exposure. Terfasa (2017) reported that a reduction in bubble size to the nanoscale resulted in an increase in the concentration of  $\cdot\text{OH}$  by a minimum of 3.5-fold when compared to the microbubble system, even in acidic media (Terfasa 2017), promoting the oxidation of small molecular organic matter such as olefins, having  $\text{C}=\text{C}$  bonds to  $\text{CO}_2$  and converting large molecular content to small sizes for subsequent combined biological processes such as MBRs, if necessary. 1,4-Dioxane, which has low microbial degradability and shows potential carcinogenicity to humans, was oxidised by the ozone-nanobubble system; the concentration decreased in the exponential decay model and dropped from 0.33 mg/L to the detection limit (0.005 mg/L) in 2–3 h, which was ascribed to the large amount of  $\cdot\text{OH}$  generated (Maie et al. 2020). As we can see, different types of gas source in bulk nanobubble-based system all get impressive removal efficiency of various organic pollutants by advanced oxidation which are briefly shown in Table 1.

**Table 1 Typical researches of removal efficiency of organic pollutants by bulk nanobubble-based advanced oxidation**

[Full size table](#) >

## Ozone-integrated biological technologies

Ozonation hybrids with sustainable biological methods, as discussed in the previous section, such as biological activated carbon and membrane bioreactors, are powerful for reducing recalcitrant chemicals and removing toxic pollutants. Wang et al. (2019) explored the advanced treatment of bio-treated dyeing and finishing wastewater (BDFW) in textile industries through the  $\text{O}_3$ -BAC system. The dissolved organic carbon and chemical oxygen demand removal rates were only 6% and 16.5%, respectively by individual ozonation, and 33% and 40% by BAC alone, while 43% and 45.8% reduction were achieved by the  $\text{O}_3$ -BAC combined system, respectively, the addition of ozone had no obvious improvement for removal efficiency (Wang et al. 2019). BAC alone preferentially degrades biodegradable fractions such as small molecular acids, ketones, alcohols, and halogen-containing compounds but rarely degrades fluorophores, whereas ozonation is capable to transfer

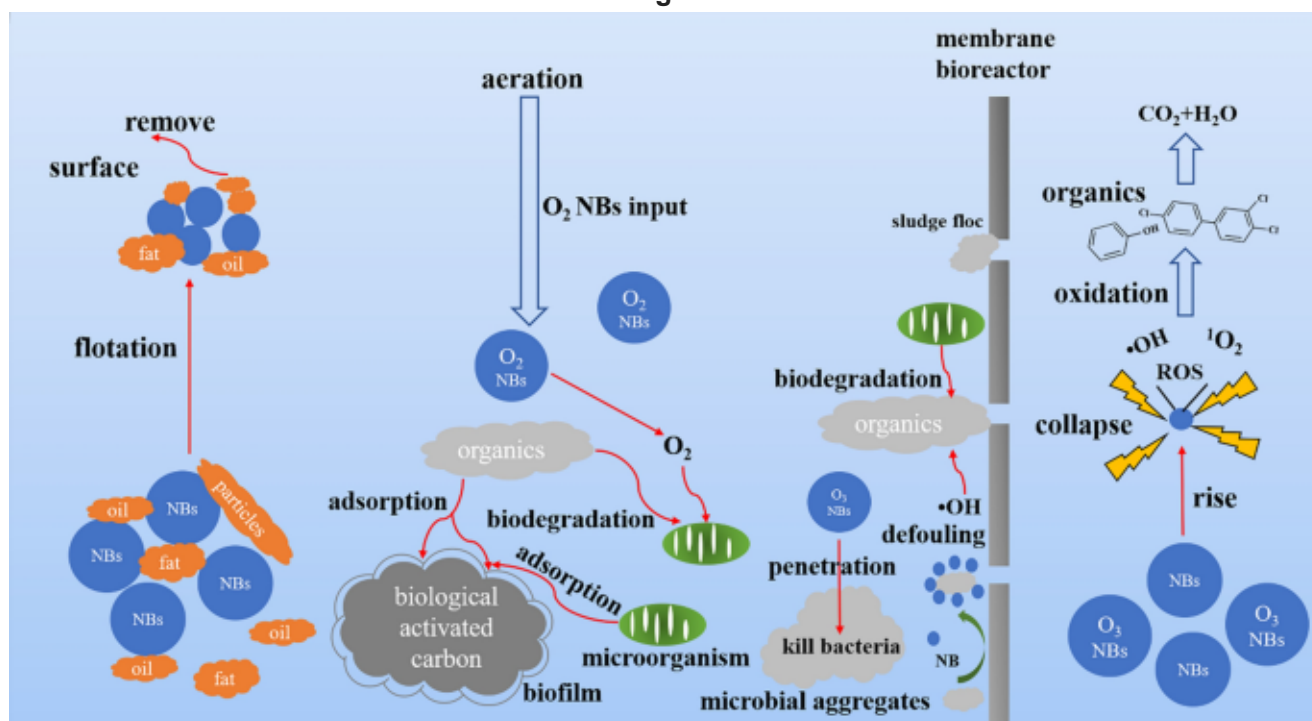
hydrophobic or hydrophilic protein-like fluorophores to less fluorescent transformation products that are more susceptible to biodegradation via BAC treatment, which explains the superior performance of the O<sub>3</sub>-BAC combination system. Hu et al. (2021) integrated ozone with co-immobilised microalgae-activated sludge bacterial biodegradation to remove organic compounds in meat processing wastewater and reported that bacterial growth was encouraged in ozone-pretreated wastewater (7.1–8.1 log CFU/mL) compared with the non-pretreated control (6.0 log CFU/mL). This was found to be due to the enhanced biodegradability of contaminants in the same way as algal biomass growth; following pre-treatment with a favourable concentration of ozone, the soluble chemical oxygen demand achieved a removal rate of 60.1% by microalgal biotreatment, while it is only 34.4% in control (non-ozonated wastewater), confirming the synergistic effects of these two technologies (Hu et al. 2021). Nonetheless, the removal rate remains restricted and dissatisfactory; thus, it is of great need to investigate whether bulk nanobubbles, which are beneficial to individual ozonation or biological treatment methods, can facilitate hybrid treatment efficiency.

## Conclusions

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Although the initial proposal of bulk nanobubbles was controversial, their existence in water has since been proven experimentally owing to the continuous exponential increase in research efforts in recent years. It is clear that the unique features of nanobubbles such as high stability, long lifetimes, large surface-volume ratio, high mass transfer efficiency, and capability to generate free radicals, can provide various means to improve conventional technologies in the water treatment field. For future perspectives, considering the wide applications of nanobubbles in order to further exploit their usage and gain a better in depth understanding of the parameters that having profound impacts are crucial. Nanobubbles have significant potential as a new environmentally friendly method to remove organic compounds as shown in Fig. 1 through their small size and existence of a surface charge, which effectively improve the air flotation process to separate suspensions; the large specific surface area and durability of nanobubbles allow the enhancement of the oxygen mass transfer efficiency to promote aeration for aerobic microorganisms to biodegrade organics and decrease the production of excess sludge in the activated sludge process, and alleviate fouling on the membrane of MBRs; moreover, the ability to generate free radicals with considerable oxidation function promotes the degradation of organic compounds. In this regard, this technology has significant value for treating wastewater containing organic pollutants.

Fig. 1



The pathways for removing organic pollutants by bulk nanobubble-integrated technologies in wastewater  
[Full size image >](#)

Nevertheless, further investigation of bulk nanobubbles is required. Only by solving these challenging issues can we make a breakthrough to provide a solid theoretical basis for the industrial scale operations of bulk nanobubbles. The topics of subsequent research can be determined based on the following issues:

(1) Many researchers have proposed that a large amount of active oxygen will be produced when nanobubbles rupture, and the existence of  $\cdot\text{OH}$  during the application of nanobubbles has been proven. However, other ROS species such as  $\text{O}_2^{\cdot-}$  and  $^1\text{O}_2$  have not been confirmed through mature techniques; thus, their effectiveness has yet to be evaluated, and the interpretation of ROS generation remains controversial. Additionally, various reports verify that ROS concentration has different influences including promotion or toxicity to plants, but the synergistic and antagonistic interaction of ROS with microbial activities during application of nanobubble techniques are uncovered.

(2) The advantages of bulk nanobubbles in aeration during the MBR biodegradation process have been highlighted. Nevertheless, few studies have focused on the probable mechanism of nanobubbles in membrane cleaning, which leads to reduce the requirement of chemicals such as antiscalants, prolong the membrane lifetime, and decrease operational expenses.

(3) Advanced oxidation processes which mainly depend on  $\cdot\text{OH}$ , such as ozonation, have been integrated with MBR systems for the treatment of various wastewaters, including oils and pharmaceuticals, dyeing or textile wastewater, municipal effluent, and domestic and

industrial wastewater. Further investigations are required to elucidate the influence of bulk nanobubbles on the treatment efficiency of these hybrid systems.

(4) Organic compounds, which cause taste and odour problems, such as 2-methylisopropanol and geosmin, generated from microorganisms that persistently exist in tap water even after it has been treated by water plants, have attracted great attention from the public in recent years. Thus, new effective methods are of urgently required, and how to combine bulk nanobubbles with biological activated carbon to adsorb and oxidise these taste and odor compounds via  $\cdot\text{OH}$  generated by bulk nanobubbles, deserves more attention.

(5) Some investigations have indicated nanobubbles consume lower energy than other methods, but there is no specific data for reliable quantification. A comprehensive cost-benefit analysis should consist of the whole processes of the nanobubble treatment system, including the production of devices, operation, maintenance, disposal, etc. The production of devices which are low-cost competitive devices that show superior performance and can be used in practical engineering is of great importance.

(6) So far, research on the application of bulk nanobubbles to the treatment of organic pollutants is limited. The existence of various substances in wastewater and the influence of the operating conditions on the treatment need to be investigated, and the process of conversion of organic matter remains to be discussed.

## Availability of data and materials

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Not applicable.

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