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环境中 N -(1, 3 -二甲基丁基) - N′-苯基-对苯二胺及其醌的去除研究进展*

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摘 要：N -(1, 3 -二甲基丁基) - N′-苯基-对苯二胺[N-(1, 3-dimethylbutyl)-N′-phenyl-*p*-phenylenediamine，6PPD]是橡胶轮胎生产中广泛使用的抗老化剂。随着橡胶产业的快速发展，6PPD 及其氧化产物 6PPD -醌[N-(1, 3-dimethylbutyl)-N′-phenyl-*p*-phenylenediamine quinone，6PPD - Q]在环境中的暴露水平持续升高，已成为备受关注的新型污染物。6PPD 和 6PPD - Q 均具有生物毒性，可通过环境介质、食物链等多种途径威胁生态系统和人类健康。因此，开发高效去除技术已成为环境科学领域的研究热点之一。通过概述 6PPD 与 6PPD - Q 的基本毒性情况，系统综述目前主要的去除策略，分析不同方法的处理效果。尽管现有研究已为 6PPD 和 6PPD - Q 的污染治理提供了多种可行方案，但在实际应用中仍存在明显局限、二次污染风险以及成本较高等问题。未来的研究可重点关注高效生物降解技术的开发、多技术协同优化体系的构建以及借助大数据深入解析其在环境中的迁移与转化规律，从而推动更经济、绿色、高效的污染治理体系的开发，为生态环境修复与人类健康保障提供理论依据和技术支撑。

关键词：环境工程学；N -(1, 3 -二甲基丁基) - N′-苯基-对苯二胺；N -(1, 3 -二甲基丁基) - N′-苯基-对苯二胺醌；毒性危害；去除策略

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0 引 言

N -(1, 3 -二甲基丁基) - N′-苯基-对苯二胺[N-(1, 3-dimethylbutyl)-N′-phenyl-*p*-phenylenediamine，6PPD]是一种广泛应用于橡胶轮胎中的抗老化剂[1-2]。如图 1 所示，在光照和臭氧条件下，6PPD 可快速氧化生成 N -(1,3 -二甲基丁基) - N′-苯基-对苯二胺 - 醌[N-(1, 3-dimethylbutyl)-N′-phenyl-*p*-phenylenediamine quinone，6PPD - Q][3-4]。6PPD 和 6PPD - Q 均难溶于水，易溶于甲醇、苯等有机溶剂，其基本信息见表 1[5]。随着橡胶工业的发展和汽车保有量的增长，6PPD 的使用量持续上升，据统计，2020 年中国 6PPD 的年产量已达 20 万 t[6]。环境中的 6PPD 和 6PPD - Q 主要来源于汽车轮胎及橡胶制品。这些产品在日常使用过程中由于老化磨损产生橡胶颗粒，并通过降水、粉尘和河流等媒介迁移到环境中[7-9]，释放出 6PPD，进而氧化生成 6PPD - Q。6PPD 和 6PPD - Q 对生物体均存在毒性，其中 6PPD - Q 的化学性质更为稳定，并对水生生物表现出更强的致死毒性[3, 10-11]。

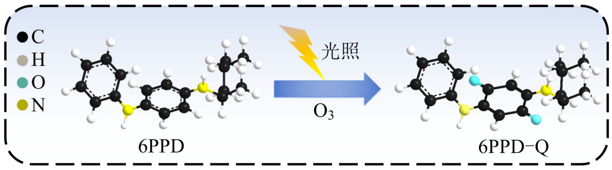

图 1 光照与臭氧条件下 6PPD 到 6PPD - Q 的转化

Fig. 1 Transformation of 6PPD to 6PPD - Q under light and ozone conditions

近年来，6PPD 与 6PPD - Q 已在全球多个地区的水体中被检出[12-13]。据有关报道，世界范围内不同地区的河道水域普遍存在不同程度的 6PPD 和 6PPD - Q。例如，中国浙江台州椒江流域 6PPD 质量浓度为 4.0 ~ 72 ng/L，6PPD - Q 的平均质量浓度为 7 ng/L[14]；中国香港的径流水域中 6PPD 的质量浓

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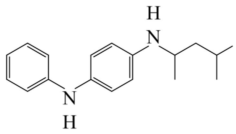
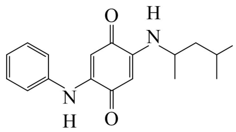
表 1　6PPD 与 6PPD－Q 的基本信息

Table 1　Basic information of 6PPD and 6PPD－Q

中文全称	英文全称	缩写	分子结构式	分子式	相对分子质量	CAS 号	辛醇水分配系数
N－(1，3－二甲基丁基)－N′–苯基–对苯二胺	N-(1，3-dimethylbutyl)-N′-phenyl-*p*-phenylenediamine	6PPD		$C_{18}H_{24}N_2$	268.40	793－24－8	4.47
N－(1，3－二甲基丁基)－N′–苯基–对苯二胺醌	N-(1，3-dimethylbutyl)-N′-phenyl-*p*-phenylenediamine quinone	6PPD－Q		$C_{18}H_{22}N_2O_2$	298.39	2754428－18－5	3.98

度为 0.21～2.71 μg/L，而 6PPD－Q 的质量浓度为 0.21～2.43 μg/L[15]；美国西雅图道路径流中 6PPD－Q 的质量浓度为 0.8～19 μg/L，洛杉矶附近流域则达 4.1～6.1 μg/L[16]。

随着 6PPD 及其醌类转化产物 6PPD－Q 在环境中不断检出，其潜在生态与人类健康风险也不断被揭示。Du 等[17]首次报道了中国华南地区人类尿液中存在 6PPD 与 6PPD－Q。戴情园等[2]也探讨了其潜在的人类健康风险。鉴于 6PPD 与 6PPD－Q 在环境中的广泛存在与生态毒性，开发其高效降解与去除技术成为当前研究的迫切需求。然而，以往研究多集中于其毒性机理与环境分布，缺乏对去除方法的系统总结。本文在概述 6PPD 与 6PPD－Q 生态毒理的基础上，重点综述近年来在光催化法、直接氧化法、吸附剂技术和生物降解技术等方面的去除策略进展，并分析各类方法的去除机理、效率与局限性，以期为相关环境修复材料与治理策略的开发提供参考。

1　6PPD 与 6PPD－Q 毒性概况

由于轮胎在使用过程中的磨损、老化以及废弃处置等原因，轮胎颗粒中的 6PPD 和 6PPD－Q 已广泛分布在多种环境介质中(图 2)，包括降水、尘埃、土壤、水域以及道路等。此外，植物[18-20]、水生动物[21-23]、陆生动物[24-26]等多个生物类群也受这类污染物的影响。面对如此广泛且日益严峻的污染态势，人类自身也面临 6PPD 和 6PPD－Q 所带来的潜在健康风险。

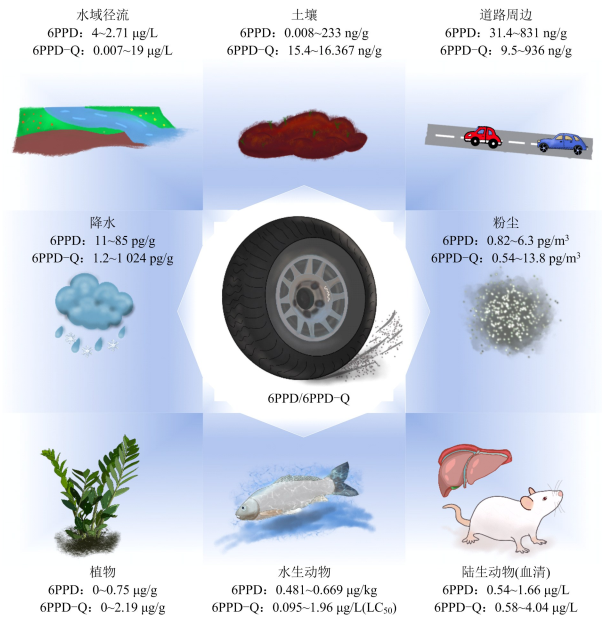


图 2　6PPD 与 6PPD－Q 的环境与生物质量浓度分布

Fig. 2　Environmental and biological mass concentration distribution of 6PPD and 6PDD－Q

1.1　水生动物毒性风险

6PPD 和 6PPD－Q 会对鱼类造成多种毒性效应，包括生长发育异常、行为改变、氧化应激和器官损伤等[27]。2021 年，Tian 等[16]首次报道并证实 6PPD－Q 是导致美国太平洋西北地区银鲑鱼在城市径流中洄游产卵时急性死亡的主要原因，其半数致死质量浓度(Median Lethal Concentration，LC_{50})为(0.8±0.16) μg/L。值得注意的是，目前部分水域中 6PPD－Q 的质量浓度已显著超过该阈值。在非

致死浓度下,6PPD 会降低斑马鱼胚胎自发运动频率、心率及色素形成,并引发凸眼球、躯干弯曲等畸形表型;而 6PPD – Q 除具有类似毒性效应外,还会引起肠道血细胞聚集和结构改变[28]。此外,6PPD 可通过干扰甲状腺信号通路和视网膜结构,诱发斑马鱼小眼症,其对眼部的毒性强于 6PPD – Q[29]。不同鱼类对这类物质的敏感程度不同,如银鲑鱼和虹鳟鱼等物种表现出较高的敏感性,而红鲑则表现出相对较强的耐受性[30]。除鱼类之外,6PPD – Q 还会抑制浮游生物海洋轮虫的群落增长[31]。同样,水蚤等无脊椎动物的生长也会受 6PPD 和 6PPD – Q 的显著抑制[32]。这些研究结果揭示了 6PPD 对水生物种的生理毒性。

1.2　陆生动物毒性风险

6PPD 和 6PPD – Q 在环境相关浓度下已表现出广泛的生物积累与多器官毒性。研究显示,6PPD – Q 可在黑斑蛙肝脏中积累,并导致其脂质代谢紊乱[33]。在无脊椎动物中,Zhou 等[24]发现近似道路周边环境浓度的 6PPD – Q,可诱导蚯蚓发生氧化应激反应,其在 1 000 μg/kg(干质量)的污染质量比下存活率显著下降。此外,土壤中的弹尾虫也受 6PPD – Q 的毒害,基于环境土壤质量,其 6PPD – Q 慢性半数致死质量比为 16.31 μg/kg[34]。在哺乳动物中,He 等[35]发现小鼠在反复暴露于 0.4 mg/kg 和 4 mg/kg 剂量的 6PPD – Q 后,肝、肾、肺等多个器官出现毒性效应,且该物质主要积累于肝脏和肺组织。Fang 等[36]通过转录组学分析进一步揭示,6PPD 和 6PPD – Q 可通过干扰糖脂代谢、免疫应答及谷胱甘肽代谢等通路引发小鼠肝毒性。Jia 等[37]通过基质辅助激光解吸/电离质谱成像(Matrix-Assisted Laser Desorption/Ionization Mass Spectrometry Imaging, MALDI – MSI)技术证实,高剂量和低剂量的 6PPD – Q 均会导致小鼠多器官(包括肝脏、肾脏、脾脏和睾丸)代谢紊乱,并破坏与氧化应激及生物损伤密切相关的关键代谢路径。这些发现系统揭示了 6PPD 与 6PPD – Q 对无脊椎动物、哺乳类多个类群的潜在健康风险,其毒性机制涉及代谢干扰与氧化损伤等多重通路。

1.3　植物毒性风险

轮胎磨损产生的颗粒已对农作物的生长产生不利影响[38-39],尽管 6PPD 和 6PPD – Q 对植物的直接毒性尚未完全明确,但已有研究指出其潜在的风险。Ge 等[40]报道称,6PPD 可降低植物的光合色素含量,抑制光合作用。低剂量的 6PPD – Q 会显著提高蟋蟀草根中活性氧(Reactive Oxygen Specie, ROS)水平,而过量的 ROS 会造成生物损伤[41],金鱼藻也发生了类似现象[21]。此外,6PPD 可通过阻断小球藻对无机氮的摄取而抑制其光合作用,并导致藻细胞生长呈现“抑制–恢复–刺激”3 个阶段变化:在初期,细胞密度随 6PPD 浓度的增加而急剧下降;随着 6PPD 由于水解、光降解和生物降解过程而浓度降低,其对微藻的生长抑制率下降;在后期,微藻细胞进入补偿性生长刺激阶段[42]。此外,研究证实 6PPD 还会导致叶绿素 a 含量降低,并干扰叶绿素的荧光参数[43]。Liu 等[20]发现,即使在较低质量浓度(1 μg/L)下,6PPD、6PPD – Q 污染物仍会对小白菜等农作物造成显著的氧化损伤。这些研究凸显了轮胎衍生的这类新型污染物在植物生态系统中的潜在风险。

1.4　人类健康风险

6PPD 和 6PPD – Q 可以通过食物链、皮肤接触、呼吸等途径进入人体[12-13, 44]。研究显示,6PPD 与 6PPD – Q 在脱氯饮用水中的半衰期分别为 5 h 和 33 h[23]。如今已在人类尿液中普遍检出这两种物质,其中孕妇尿液中含量高于普通成人和儿童,女性尿液中的 6PPD 含量比男性更高[17, 45]。基于网络药理学以及分子对接的研究结果提示,6PPD – Q 可能会增加前列腺疾病的潜在风险[46]。此外,研究在人体血清和脑脊液中检测到 6PPD – Q 及其他对苯二胺醌(*p*-phenylenediamine Quinones, PPD – Qs),并发现这些物质可诱导脑血管内皮细胞发生内皮–间质转化,从而导致血脑屏障功能障碍[25]。Qi 等[47]利用人原肝细胞模型 L02 的暴露试验证实,6PPD – Q 可诱导肝细胞氧化损伤并干扰氨基酸代谢。另有调查显示,从事交通相关工作的户外工作者体内 6PPD 和 6PPD – Q 的含量显著高于室内工作者,这可能是户外工作者肝脏疾病高发的原因之一[48]。值得注意的是,6PPD – Q 在潮湿土壤中可能与重金属反应产生致癌物质[49]。因此,长期暴露于 6PPD 和 6PPD – Q 可能对人体免疫系统、神经系统造成威胁,并对孕妇和胎儿健康构成潜在风险。

2　6PPD 与 6PPD – Q 的去除策略

由于 6PPD 与 6PPD – Q 在环境中的广泛分布与生态毒性,因此,开发其高效的去除和控制方法对维护生态与环境安全具有重要意义。2024 年,Yan 等[50]最先揭示了水环境中 6PPD – Q 在自然条件下的衰减主要是通过羟基化、醌环断裂、键裂解以及重

排这 4 个路径，其中直接光照和间接光照是其降解的主要驱动力。

6PPD 在臭氧作用下主要转化为 6PPD – Q[51-52]。相较于 6PPD，6PPD – Q 对水生生物毒性更强，且在环境中性质更稳定。因此，目前针对该类污染物的去除主要为 6PPD – Q。已有研究证实，醌类化合物可被臭氧进一步氧化[53]，例如，Seiwert 等[54] 在 6PPD – Q 的臭氧氧化试验中鉴定出 12 种转化产物，表明臭氧主要攻击其醌环系统。基于现有研究，6PPD 及 6PPD – Q 等污染物的去除策略主要包括光降解法、直接氧化法、吸附剂法和生物降解法（图 3）。

2.1 光降解法

2.1.1 单紫外光降解

紫外光催化作为一种常见的消毒降解技术，在环境污染物去除中具有独特优势[55-56]。现有关于 6PPD 及 6PPD – Q 的去除研究主要集中在光驱动的高级氧化过程。对醌类物质在 254 nm 处具有显著的紫外吸收，可在水相激发产生羟基自由基（·OH）和半醌自由基[57-58]，并通过氧化还原循环机制将对醌转化为对苯二酚等产物，从而实现降解[59]。2024 年，Wang 等[60] 证实紫外线照射可有效去除水环境中的 6PPD – Q 及其他 PPD – Qs，在初始质量浓度为 100 μg/L 时，经过紫外线照射处理 40 min 后，6PPD – Q 的去除率可达 94%。其转化路径为：6PPD – Q 在紫外光照射下激发为三联态后，在持续的水分子和紫外光共同作用下最终形成 6PPD – Q 羟基化产物。该研究结果为光催化技术应用于此类污染物的实际控制提供了理论基础。

2.1.2 光激活高级氧化

高级氧化过程（Advanced Oxidation Processes，AOPs）由于产生活性自由基，可以实现有机污染物的快速降解甚至矿化，无论是利用过硫酸盐还是高碘酸盐进行反应的 AOPs，均展现出优异的污染物去除效果[61-63]。光催化与高级氧化工艺协同作用效果显著。2024 年，Chen 等[64] 首次利用太阳光激活高碘酸盐，在环境质量浓度水平（10 ~ 100 μg/L）实现了 6PPD – Q 的有效降解，处理 30 min 后，其质量浓度下降至 5.7 ng/L。研究指出，高碘酸盐（IO_4^-）活化后产生的 IO_3·是降解过程中的主要活性物种，可实现 6PPD – Q 分子结构的裂解。Yu 等[65] 采用紫外线激活的过氧单硫酸盐（Ultraviolet-Activated Peroxymonosulfate，UV/PMS）体系降解 6PPD – Q，当过氧单硫酸盐（Peroxymonosulfate，PMS）与 6PPD – Q 的初始物质的量比为 60∶1 时，6PPD – Q 可完全降解，该过程主要由 SO_4^-·和·OH 驱动，且降解产物

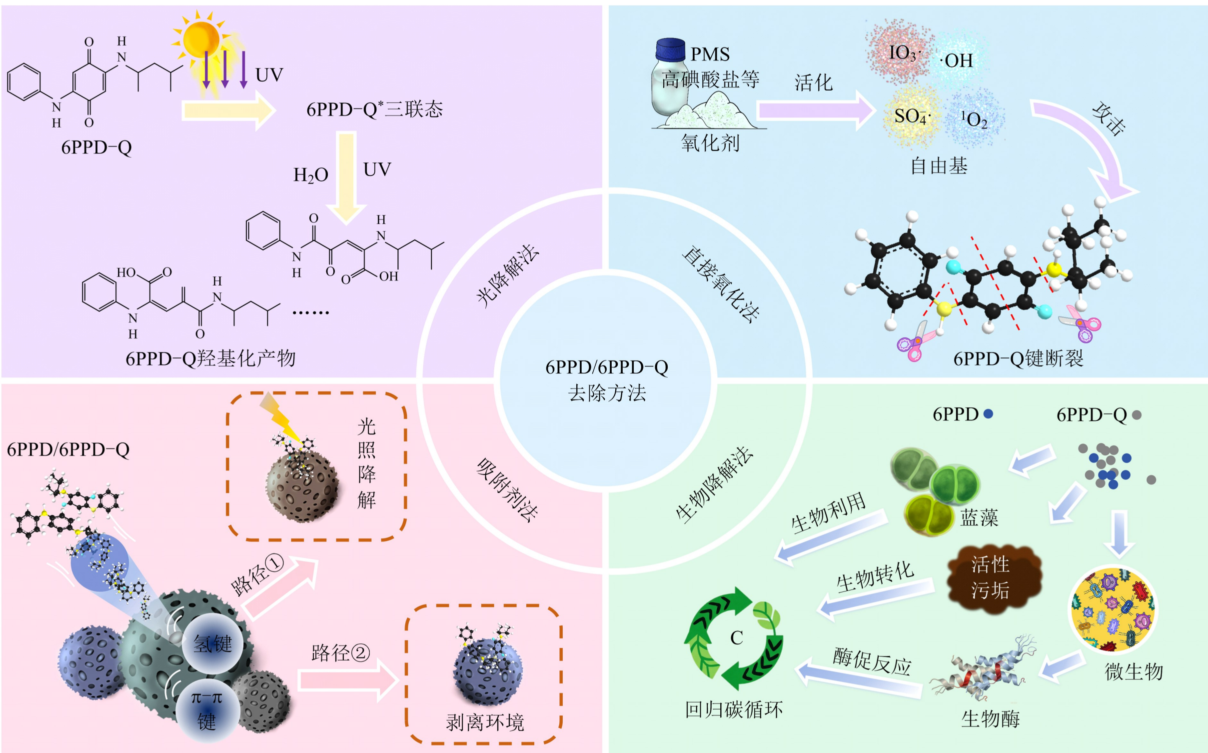


图 3 6PPD/6PPD – Q 去除策略及机理示意图

Fig. 3 Schematic diagram of 6PPD/6PPD – Q removal strategy and mechanism

对水生生物毒性较低。

2.1.3　吸附剂协同光降解

光催化与吸附剂协同作用可有效降解多种有机污染物[66],2025 年,Chen 等[67]首次构建了一种吸附剂耦合光催化降解 6PPD - Q 体系,通过制备绣球状空心 O、Cl -掺杂石墨氮化碳微球(Hydrangea-Like Hollow O, Cl - Codoped Graphite-Phase Carbon Nitride Microspheres,HHCN),增强了对水体中 6PPD - Q 的捕获能力,并在光照条件下实现了有效降解。该体系在 1 h 内对初始质量浓度为 10 mg/L 6PPD - Q 的降解率超过 90%。

光降解技术操作简便,主要利用光照对污染物进行直接或间接降解,易与其他方法联用从而提高整体处理效率。然而,该方法对于地下水系统和黑暗环境中的应用存在局限性。

2.2　直接氧化法

单线态氧($^1O_2$)在水溶液中能够促进 6PPD - Q 的降解[68]。由于苯醌与 6PPD - Q 具有相似结构,且能够刺激 PMS 分解产生$^1O_2$[69-70],前人开发出一种无需光照条件、直接利用未活化 PMS 降解 6PPD - Q 的新策略。2025 年,Yu 等[71]最先开发了一种直接利用未活化 PMS 降解 6PPD - Q 的方法,与利用光照激活的高级氧化工艺相比,该方法无需外部能源输入和活化剂等投入,更加节能简便,结果显示未活化 PMS 的降解效果与活化的 PMS 相当。该研究还考察了降解体系中各种因素(初始物质的量比、温度、pH 值、阴阳离子等)对降解效果的影响,并发现 pH 值的影响最为显著。当 6PPD - Q 与 PMS 的初始物质的量比为 1∶50 时,在碱性条件下的降解效果显著优于酸性条件;在 pH 值为 9 和 11 时,体系中的 6PPD - Q 均被完全去除。根据试验数据分析,$^1O_2$氧化和直接氧化是 6PPD - Q 降解的主要机制,在 pH = 9.0 时,$^1O_2$ 对 6PPD - Q 降解的贡献率达到 79%。氯消毒是城市污水处理的常用方法,直接利用次氯酸盐、二氧化氯等强氧化剂可实现 6PPD 和 6PPD - Q 的降解。但该方法产生的 6PPD - Q 氯取代产物毒性反而增强,易造成二次污染,并使后续解毒处理面临更大挑战[72]。

从作用机理上看,直接氧化法与光激活高级氧化过程相似,均依靠氧化剂产生活性自由基,进而攻击醌环结构与 C—N 键,最终导致 6PPD - Q 分解。

相较于光降解技术,直接利用氧化剂降解 6PPD 与 6PPD - Q 具有操作简单和能耗低的优势,但该方法存在氧化剂残留以及有毒副产物生成的风险。因此,需开发更为高效、环境友好且二次污染风险较低的氧化剂,以实现此类污染物的安全、高效去除。

2.3　吸附剂法

吸附剂主要通过 π - π 键、氢键、静电作用以及孔隙填充等机制发挥作用[73],在有机污染物废水处理中,吸附技术由于高效、设计简单和操作便捷被广泛应用,添加吸附材料已成为污水净化的常用策略[74]。

2025 年,Wu 等[75]开发了一种新型三维双金属 MOF 纳米片材料,能够高效去除 6PPD 及 6PPD - Q。该研究采用两步法,通过金属化合物与四(4 -羧苯基)卟啉[Tetrakis (4-carboxyphenyl) porphyrin, TCPP]反应,制备了三维双金属纳米片材料 Al - TCPP - M(M = Co,Cu,Zn)。与其他吸附材料相比,该纳米片材料对 6PPD 和 6PPD - Q 表现出优异的吸附性能,吸附率分别超过 97% 和 98%。这种强大的吸附性能主要归因于吸附过程中氢键的形成,以及过渡金属与 6PPD 或 6PPD - Q 的 π - π 络合作用。

碳质材料由于良好的化学稳定性、结构多样性、密度低和易于大规模生产,在吸附领域备受关注[76]。Chen 等[67]制备的碳基 HHCN 材料的比表面积为 66.8 m^2/g。其多级多孔结构不仅增大了比表面积,促进了活性位点的暴露与反应物的吸附,还通过延长光反射路径进一步提升了光催化性能,使其对 6PPD - Q 的去除率是块状石墨相氮化碳($g - C_3N_4$)的 2.5 倍。此外,在潮湿土壤中添加小麦壳生物炭也可以显著降低 6PPD 和 6PPD - Q 的暴露风险,同时有助于稳定土壤中的生物多样性、缓解氧化应激并提升土壤肥力[26]。

Yu 等[77]发现,某些微塑料颗粒可通过氢键和 π - π 键等作用有效吸附 6PPD - Q(吸附率约 80%),并具备将其迁移到他处的潜力,尽管这可能导致 6PPD - Q 扩散,但微塑料在吸附 6PPD - Q 后的整体迁移能力会下降。

除人工吸附材料外,自然界中某些天然颗粒物也表现出优异的吸附性能。2025 年,Shi 等[78]率先发现,土壤中天然的铁锰结核对 6PPD 和 6PPD - Q 具有良好的吸附能力,能促进 6PPD 向 6PPD - Q 的转化。由于该材料对 6PPD - Q 的吸附性能更强,可有效截留此污染物,阻断其环境迁移,因此可为新型吸附剂的开发提供启发。

总体而言,吸附法能有效将 6PPD 及 6PPD - Q 从环境中分离,且具备易于规模化的优势。作为辅

助材料,吸附剂可灵活与其他技术联用,展现出广泛的适用性。然而,吸附剂的回收利用及最终处置问题仍须进一步研究。

2.4 生物降解法

生物技术已在污水处理领域得到广泛应用[79-80],这也为 6PPD 及 6PPD-Q 的去除提供了潜在可能。目前,这类污染物去除的生物手段包括微生物降解、植物与藻类修复和生物酶降解。

2.4.1 微生物降解

在污水处理厂的沉淀池淤泥中,存在许多能够降解有机污染物的微生物,这些富集于活性污泥中的微生物在水体净化过程中发挥着重要作用[81-82]。2024 年,Foscari 等[83]首次利用污水处理厂的活性污泥作为生物接种物,研究了轮胎颗粒浸出物的转化过程。结果表明,在含轮胎颗粒的悬浮液中,无论是否接种污泥,6PPD 在样品中的质量比均能降至接近 0,而 6PPD-Q 则最终会达到释放-降解的动态平衡,且接种组的平衡值低于未接种组。进一步对比上清液 6PPD-Q 的质量浓度变化可以发现,其在接种组中的下降速度更快。研究还指出,接种组出现了氧化程度更高的 6PPD-Q 转化产物,表明微生物显著促进了 6PPD-Q 的转化。

除活性污泥中存在可有效降解 6PPD 和 6PPD-Q 的微生物外,某些生物体内也可能存在对这类污染物的耐受物种。Xu 等[34]研究显示,土壤弹尾虫肠道微生物组在 6PPD-Q 暴露的条件下可以实现协同反应以减轻 6PPD-Q 毒性。

2.4.2 植物与藻类修复

植物和藻类在环境修复的作用已被证实,其中利用蓝藻处理废水的技术发展迅速[84-86]。尽管目前针对新型污染物 6PPD-Q 的植物修复研究仍较为有限,但通过评估植物对该污染物的适应性,可初步判断其去除潜力。You 等[87]在研究醌类对蓝藻光合作用的影响时发现,虽然 6PPD-Q 会抑制光合作用使得生物体内碳固定量持续减少,但蓝藻最终表现出一定的适应性。具体而言,在 0.1 mg/L 6PPD-Q 暴露下,蓝藻在初期(0~16 代)呈现低剂量刺激效应,细胞增殖加快、光合作用增强;在中期(17~240 代)出现毒性响应,细胞密度显著下降;而在后期(240 代后)则逐渐恢复适应性,细胞密度回升至对照水平。在整个 100 d 的试验周期中,总体生物量损失为 9%。该研究中,蓝藻对 6PPD-Q 表现出适应性,表明植物物种在此类污染物去除领域具备一定的应用潜力。

2.4.3 生物酶降解

生物酶作为一种绿色环保的催化剂,已广泛应用于环境污染物的去除,且其开发与应用潜力仍在不断扩大[88-90]。研究表明,酶在 6PPD-Q 的生物转化过程中具有关键作用。2025 年,Yu 等[91]首次发现真菌 *Phanerochaete chrysosporium* 分泌的木质素过氧化氢酶同工酶结合吸附作用,在 7 d 内对 6PPD-Q 的去除率可达 99.06%。该研究为开发可持续的 6PPD-Q 降解方法提供了重要范例。此外,人类、小鼠以及鲫鱼的肝脏微粒体均可将 6PPD-Q 降解约 50%,其中细胞色素 P450 酶系发挥主要作用[92]。Zhang 等[93]也指出 P450 酶中的 CYP1A2、3A4 和 2C19 等在 6PPD-Q 降解中的贡献显著。这些体外试验结果表明,生物体内存在可催化降解 6PPD-Q 的酶类,这也是生物体实现自身解毒的重要途径之一。

生物降解法的核心在于利用生物技术将 6PPD、6PPD-Q 等污染物作为碳源进行吸收和代谢。相较于其他处理方法,生物降解后一部分污染物被转化为生物量固定,另一部分重新进入碳循环,具有能耗低、处理效果好、二次污染风险小等优点,是一种环境友好型的治理策略。然而,该方法目前处理周期较长,仍需进一步优化以提高降解效率、缩短处理时间。

根据上述报道的去除策略,本文总结了对 6PPD 和 6PPD-Q 的去除效果(表 2),从现有数据来看,大部分方法对此类污染物的降解和去除率均可达到 90%以上。

3 结论与展望

随着轮胎抗老化剂 6PPD 及其氧化产物 6PPD-Q 在环境中暴露水平的不断上升,引发的生态环境问题日益严峻,并对人类健康构成潜在风险。目前,已有多种方法被用于去除环境中的 6PPD 和 6PPD-Q,主要包括光降解、直接氧化、吸附剂和生物降解等 4 类技术。这些方法在试验条件下均能有效降解和去除相应浓度范围内的目标污染物。然而,在实际应用中仍存在明显局限性:光降解受光照条件制约;直接氧化法虽然高效节能,但存在氧化剂残留及有毒副产物的风险;现有高效吸附剂种类有限,其循环性能、耐久性与回收问题尚不明确,部分金属有机框架吸附剂还可能导致重金属二次污染;生物降解技术依赖于特定微生物的耐受性与降解能力,处理周期通常较长。

表 2　6PPD 与 6PPD - Q 的去除研究总结

Table 2　Summary of the research on the removal of 6PPD and 6PPD - Q

去除方法	目标污染物	处理条件	去除效果	参考文献
光降解法	6PPD - Q	光照 40 min(紫外线通量为 5 860 mJ/cm^2)	去除率为 94.2%	[60]
	6PPD - Q	IO_4^- 在 10:00 自然光下活化(光照强度为 45 mW/cm^2),60 min	降解率为 94.0%	[64]
		IO_4^- 在 13:00 自然光下活化(光照强度为 60 mW/cm^2),60 min	降解率为 94.8%	
		IO_4^- 在 16:00 自然光下活化(光照强度为 60 mW/cm^2),60 min		
		IO_4^-(0.2 mmol/L)在模拟太阳光下活化 60 min	100 μg/L 下降到 16.7 ng/L	
	6PPD - Q	ρ(PMS):ρ(6PPD - Q) = 60:1(PMS 质量浓度为 60 mg/L),在紫外线下照射 120 min	降解率为 100%	[65]
直接氧化法	6PPD - Q (50 μg/L)	钼酸盐(1 mmol/L)和过氧化氢(3 mmol/L)体系,20 ℃,黑暗条件	半衰期为(0.055 ± 0.004) h	[68]
	6PPD - Q (3.35 μmol/L)	初始 c(PMS):c(6PPD - Q) = 50:1, c(6PPD - Q) = 3.35 μmol/L, t = (25 ± 2) ℃,180 min	降解率为 95.0%	[71]
		pH = 9 或 11,60 min	降解率为 100%	
吸附剂法	6PPD (90 mg/L)	3D 双金属纳米片材料:Al - TCPP - M(M = Co, Cu, Zn) 吸附条件:T = 298 K,pH = 6.0 持续 120 min	吸附质量比达 331.9 ~ 355.9 mg/g,去除率超过 97.0%	[75]
	6PPD - Q (60 mg/L)		吸附质量比达 260.6 ~ 335.0 mg/g,去除率超过 98.0%	
	6PPD - Q (10 mg/L)	绣球状空心 O,Cl 掺杂石墨相氮化碳微球(HHCN)暗反应测试 60 min,光照 1 h	降解率为 94.0%	[67]
	6PPD - Q (100 mg/L)		降解率为 75.0%	
	6PPD - Q	天然铁锰结核(Natural Fe-Mn Nodule,NFMN),293 K 下吸附反应 10 min	吸附质量比达 719.2 μg/g	[78]
生物降解法	6PPD - Q (1 mg/L)	真菌 *Phanerochaete chrysosporium* 降解 7 d	去除率为 99.06%	[91]

鉴于 6PPD/6PPD - Q 污染物普遍存在,开发高效、经济且环境友好的去除技术尤为迫切。在众多处理策略中,生物转化技术展现出良好的应用前景,尤其是其可持续性优势显著。尽管目前相关研究仍较为有限,但已有研究显示,酶协同吸附作用对 6PPD - Q 具有优异的降解性能。未来可借助固定化酶技术进一步提升其操作稳定性与回收效率。此外,某些微生物(如蓝藻、淤泥微生物等)对 6PPD 和 6PPD - Q 具有耐受和降解能力,具备对此类污染物去除的潜力。需要注意的是,6PPD - Q 在转化过程中可能生成毒性不明的副产物,系统评估其转化产物的生态毒性至关重要。

另外,由于污水中污染物组分复杂,在实际处理中,为了尽可能多地去除 6PPD 及 6PPD－Q,还需进一步改善污水处理工艺。未来研究可进一步关注多种治理技术的协同策略。例如,在污水沉淀池中添加适量的吸附剂使污染物聚集,再经过滤分离,对聚集的污染物进行光解、氧化降解等后续处理;将对 6PPD 与 6PPD－Q 具有耐受性的微生物定植于吸附剂上,或将有效的固定化酶投入目标环境以进行治理,既能实现 6PPD 和 6PPD－Q 的针对性处理,又能提升稳定性,将固定化基质过滤分离后经过一定处理还可以循环利用。

现今,大数据快速发展,未来应积极探索多技术协同的处理路径,例如融合大数据、人工智能等新兴技术优化相关治理策略。在此过程中,可借助计算机技术与机器学习等方法,进一步改进轮胎生产工艺以减少 6PPD 释放。同时,可借助地理信息系统(Geographic Information System, GIS)建立区域污染实时监测与精准治理平台。此外,在开发新方法的同时,仍须进一步深入探究 6PPD 和 6PPD－Q 在环境中的迁移转化规律以及长期生态风险,从而构建更加科学、全面的风险防控体系提供理论支撑,助力实现绿色安全的生态环境治理目标。

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Research progress on the removal of N-(1, 3-dimethylbutyl)-N′-phenyl-*p*-phenylenediamine and its quinone from the environment

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Abstract: N-(1, 3-dimethylbutyl)-N′-phenyl-*p*-phenylenediamine (6PPD) is an anti-aging agent widely used in the rubber industry due to its effectiveness in preventing rubber aging. With the growth of the rubber industry and the increasing number of automobiles, the scale of 6PPD usage has also expanded, leading to increased environmental exposure to both 6PPD and its oxidation product, N-(1, 3-dimethylbutyl)-N′-phenyl-*p*-phenylenediamine quinone (6PPD-Q). Both 6PPD and 6PPD-Q are biotoxic; notably, 6PPD-Q exhibits greater toxicity and poses significant risks to fish populations. These compounds can diffuse into the environment through mechanisms such as river runoff and atmospheric dust, and they can enter the human body via food chains and respiration, thereby threatening human health. The ecological problems arising from these pollutants have garnered increasing attention in recent years. This paper begins by introducing the basic toxicity characteristics of 6PPD and 6PPD-Q, highlighting their substantial biological hazards, particularly the serious threat they pose to fish survival. It further emphasizes the potential risks to human health, supported by relevant literature, and underscores the necessity for effective removal strategies. Subsequently, this paper reviews the progress made in research on existing methods for the removal of 6PPD and 6PPD-Q, discussing the effects and advantages of various techniques. Following the oxidation of 6PPD to 6PPD-Q, its decay under natural conditions primarily occurs through four pathways: hydroxylation, quinone ring cleavage, bond cleavage, and rearrangement. In addition to degradation and bioabsorption removal, these pollutants can also be separated from the environment using embedded adsorbents. Based on these degradation mechanisms and removal methods, the main strategies for removal include photodegradation, direct oxidation, adsorption, and biodegradation. The removal efficiency of these methods generally exceeds 90%, and using a combination of approaches often yields even better results. However, challenges remain, including limited operational conditions, high risks of secondary pollution, and elevated costs. Future research should focus on developing high-efficiency biodegradation technologies, optimizing multi-technology collaborations, and utilizing big data to analyze environmental migration and transformation patterns. These efforts will help establish an economical, green, and efficient pollution control system, laying a solid theoretical and technical foundation for ecological restoration and human health protection.

Key words: environmental engineering; N-(1, 3-dimethylbutyl)-N′-phenyl-*p*-phenylenediamine; N-(1, 3-dimethylbutyl)-N′-phenyl-*p*-phenylenediamine quinone; toxicity hazards; removal strategy