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Degradation and remediation of environmental pollutants by algae

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Abstract. The usage of a large number of industrial resources and the discharge of harmful pollutants have caused serious damage to the environment and ultimately affect the ecosystem and human health. Bioremediation is a green, sustainable and low-cost technology for the treatment of environmental pollutants, which includes the process of environmental remediation using plants, animals, microorganisms and so on. Among them, algae have the characteristics of rapid growth, wide distribution, strong stress resistance, large enrichment, and can convert pollutants into chemical products, so they play an important role in bioremediation. In this review, we introduce the types of pollutants existing in the environment, analyze the mechanism of algae for environmental remediation, and then describe the research progress of algae in the process of degradation and remediation of different pollutants. Finally, the shortages and limitations of bioremediation using algae are discussed, and its future development is prospected.

Keywords: Algae; heavy metals; pesticides; organic dyes; bioremediation.

1. Introduction

In the process of industrialization and the growing demand of human beings in the 20th century, a large number of chemicals, organic materials and fossil energy have been developed and used.At the same time, a large number of pollutants emitted from the chemical industry have caused and will continue to cause environmental pollution and damage to the ecosystem.These pollutants include heavy metals [1, 2], radionuclides [3], organic pollutants (including organic dyes and hydrocarbons) [4], pesticides and pesticides [5] and so on, which affect life support functions such as plants, microorganisms, aquatic organisms, fixation, mineralization and nitrification, and ultimately affect the health of human beings and ecosystems [6] like cancer, nervous system damage, growth retardation, even death and the disruption of the normal process of plant growth and physiological metabolism [7]. As people pay attention to the living environment and the concept of sustainable development is put forward, the traditional chemical and physical remediation methods have some shortcomings, such as high cost, harmful by-products. For example, ultraviolet radiation treatment wastewater may produce aldehydes, organic bromine and bromate and other trace pollutants and radioactive waste [6], so its application is limited and more low-cost and eco-friendly green technology development is needed [8].

Bioremediation is an innovative technology that uses biological systems to deal with pollutants [9,10]. In other words, It can be used for animals, plants and microorganisms by using natural biological metabolic processes to completely remove toxic pollutants or transform them into harmless substances [11]. Green plant-mediated bioremediation refers to the use of green plants to make pollutants harmless, which can be used to repair soil or water contaminated by heavy metals, organic matter and radioactive elements [12]. In addition to higher plants, bioremediation using photosynthetic algae also plays an important role [13]. Algae, as photoautotrophic organisms, whether they are microalgae or macroalgae, have high resistance and tolerance to the toxicity of heavy metals, radioisotopes, formaldehyde, phenols and oxalic acid, and studies have shown their efficiency in removing toxic compounds Able to reach more than 90%[14].They are fast-growing, widely distributed, highly resistant, can reproduce in large numbers, and their larger surface areas absorb and enrich pollutants, which are then carried out by inhaling cells or converting pollutants into other

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compounds [15, 16]. Therefore, phytoremediation with algae is a cheap, easy to obtain, high removal rate, sustainable and will not cause secondary pollution.

In this review, we first introduce the pollutants existing in the environment, then analyze the mechanism of algae for environmental remediation, and then review the research progress of algae for environmental pollutant degradation and environmental remediation. finally, the shortcomings and limitations are discussed, and the development of algae remediation in the future is prospected. Through this review, we hope to summarize the studies on the degradation and remediation of environmental pollutants by algae in recent years, find more target strains conducive to our follow-up research, and attract more attention in algae remediation research.

2. Mechanism of environmental remediation by algae

2.1 Microalgae

Microalgae refer to tiny groups of algae that can only be identified under a microscope, belonging to Cyanophyta, Microalga, Chrysophyta and Rhodophyta. Except cyanobacteria, all the other eukaryotes contain chlorophyll and can carry out photosynthesis. It is commonly used in production and application. Microalgae have a wide range of applications, including pharmaceutical industry, food industry, animal feed, environmental testing, biotechnology, renewable energy, cosmetics [17]. Some microalgae species, such as Chlorella and Spirulina, have been used in commercial production and utilization [18]. A large number of studies have shown the prospect of microalgae as a bioremediation technology [19], they can use wastewater organic matter as a carbon source and nutrient source, so that they can treat wastewater, and finally make algae culture cost-effective [20].

Bioremediation of microalgae can be carried out through three different mechanisms: biosorption, bioaccumulation and biodegradation [21]. Biosorption is an energy-independent process that keeps pollutants on the cell surface [22] through various mechanisms, such as chelation /complexation, ion exchange, surface precipitation and adsorption[23]. Biosorption can occur either in living algae cells or in dead organisms because cell receptors remain active after cell death [24]. In addition, removal efficiency can be improved by chemical treatment, for example, base-modified algae biomass increases the functional groups of major actors in the biosorption mechanism [25]. Bioaccumulation is the detoxification of pollutants through living algae cell [26], in which algae cells absorb pollutants and nutrients through the surface [27], combine with proteins or other cell wall components to transport pollutants within the cells [21], and then accumulate or metabolize according to the type of biomass [28]. Bioaccumulation efficiency is determined by biological concentration factor (BCF), which is defined as the ratio of accumulated pollutants to external media [29], BCF value is mainly affected by biological enrichment mechanism, chemical bioavailability, physical barrier, BCF determination methods, dissolved organic matter, metabolism, interspecific variation, ionization of ionizable compounds and differences in environmental conditions [30]. However, the bioaccumulation of toxic compounds in living algae cells can lead to excessive production of reactive oxygen species (ROS), which may cause serious damage to cell components and function [31]. Photosynthetic organisms can overcome the side effects of bioaccumulation by expressing antioxidant enzymes, such as superoxide dismutase (SOD), which scavenges free radicals [80]. Biodegradation is the basic process for microorganisms to remove pollutants from sewage. Microalgae can degrade organic compounds in water into small molecules, and then use them as nutrients for their own growth. The degraded pollutants can be transformed into intermediates of other products, and can also be used by the surrounding microbial community to enhance their degradation potential [32].

2.2 Macroalgae

Macroalgae is a group of visible, chlorophyll, autotrophic plants, the vast majority of which are multicellular filaments, membranes, tubes or leaflets, including a variety of aquatic plant species, traditionally belonging to Chlorophyta, Phaeophyta and Rhodophyta [33]. In the classification of macroalgae, there are great differences in biochemical composition among species[34] and significant diversity[35]. Macroalgae account for the majority of all commercially cultivated aquatic plants (quality > 95%) and are one of the fastest growing aquaculture sectors, mainly as food commodities, but there are also a number of other uses, including as chemical fertilizers, as well as for biofuels, pharmaceuticals, mineral extraction, environmental monitoring and environmental remediation [33].

Macroalgae can achieve the effect of purification and repair mainly through biosorption, and it is an extremely effective biosorbent. The cell walls of macroalgae are rich in heteropolysaccharides, lipids and proteins, which are composed of various active sites (functional groups), including carboxyl, amino, hydroxyl and phosphate groups, which can be used as sites for binding to various pollutants on the cell surface. in order to achieve the purpose of pollutant removal. The mechanisms of its binding to pollutants include surface precipitation, chelation, ion exchange, electrostatic force, complexation or intracellular diffusion between pollutant cations and seaweed surface, intracellular bioaccumulation and binding with proteins and other intracellular components[36,37].Among them, the biosorption of brown algae biomass to pollutants in aqueous solution is mainly attributed to the high content of alginate in its cell wall[38],It is reported that the highest biological removal rate of copper in wastewater is the dried biomass algae of brown algae, especially Fucus vesiculosus and Fucus serratus[39].In addition, large algae not only absorb water pollutants to synthesize their own biomass, but also produce a large number of by-products, which has high economic value[40].

3. Research Progress on Environmental Pollutant degradation and Environmental remediation by algae

In recent years, the research on bioremediation by using different algae to remove pollutants has developed rapidly. The following will be introduced from five aspects: organic hydrocarbons, organic dyes, heavy metals, radionuclides and pesticides.

3.1 Organic hydrocarbons

Up to now, Filamentous cyanobacteria has been widely used in secondary and tertiary wastewater treatment [41].The researchers found that Phormidium valderianum grew in phenol [42] and could tolerate a concentration of 50 mg/L and remove phenol (38 mg/L) during a 7-day retention period. The mixed culture of an eukaryotic microalgae Chlamydomonas reinhardtiiand a cyanobacteria Anabaena cylindrica can remove 2-dinitrophenol and its degradation product 2-amino-4-nitrophenol (2-ANP), which is expected to be an effective application in industrial wastewater treatment [43]. In further research, unicellular marine cyanobacteriaAgmenellum quadruplicatum, Microcoleu chthonoplastes and Phormidium corium all answered whether cyanobacteria in the blue-green cushion could participate in the utilization of mixed nutrient hydrocarbons, thus solving the problem of self-cleaning along the Gulf of Mexico. It is proved that cyanobacteria can be used to degrade dibenzenes and other organic compounds [44, 45].In addition, algae species such asOscillatoria salina [46], Plectomena terebrans, Aphanocapsa sp.[47] and Synechococcus [48] have been successfully used in the bioremediation of mixed populations or oil pollutants using green algae mats in different regions of the world.

Eukaryotic microalgae also play an important role in the treatment of organic hydrocarbons. Green algae can mineralize harmful organic hydrocarbons, such as polycyclic aromatic hydrocarbons (PAHs), phenols and organic solvents, and they can support the aerobic degradation of various harmful pollutants [49]. Lima et al evaluated the ability of Chlorella vulgaris and a kind of Chlorella pyrenoidosa to biodegrade p-nitrophenol (PNP) and p-chlorophenol (PCP), and found that they could remove PCP in different light states [50]. Chrysophyta Chrysophyceae can convert phenolic cyclic hydroxyl groups to catechins and then open the benzene ring through ortho-and meta-cleavage pathways. Both ortho-and meta-cleavage pathways involve monooxygenase and dioxygenase using oxygen [50]. This specific activity can decompose phenolic compounds into pyruvate and carbon dioxide. This specific activity can decompose phenolic compounds into pyruvate and carbon dioxide

[50]. C. reinhardtii and A. cylindrica showed extremely high ability to degrade low concentration of 2,4-DNP, which was reduced to 2-ANP by these strains [43].Ankistrodesmus braunii and Scenedesmus quadricauda can degrade phenols in olive mill wastewater (OMW) with a removal rate of more than 70% [41]. They have been successfully used to treat catechol in OMW and paper industry wastewater [51].

3.2 Organic dye

Macroalgae can be used as an effective carrier to absorb organic dyes. A large number of experiments have proved that the purification of organic dyes by macroalgae is affected by many factors, such as reactant concentration, pH value, soaking time, and the soaking time and reactant concentration are positively correlated with the purification effect, while the ability of pH to absorb organic dyes by algae is related to the species of algae. Gracilaria can be used to absorb methylene blue [52]. It was found that the highest biological removal rate of methylene blue could reach 94.86% when the conditions were 6g/L Gracilaria biomass, initial pH=8, methylene blue concentration 20mg/L and treatment time 180 minutes [53]. Enteromorpha intestinalis can be used to absorb malachite green [36]. When the concentration of malachite green is 7.92 mg/L, the biomass is 4.30g / L, pH=9.92 and the treatment time is 38.5min, the maximum removal rate can reach 94.17% [36]. Ulva lactuca can be used to absorb Congo red [54]. When the concentration of Congo red dye is 100 mg/L, the algae biomass is 3 g / L, the treatment time is 120 minutes at 30 °C, and the initial pH=6, the removal rate of Congo red dye is up to 97.89% [54].

Similarly, eukaryotic microalgae showed a good effect on the adsorption of organic dyes.Some studies evaluated the decolorization effect of C. vulgaris and Nostoc paludosum on crystal violet (CV) and malachite green (MG). The results showed that C. vulgaris had the most obvious decolorization effect on CV and MG, with a decolorization rate of 93.55% for MG and 62.98%. The decolorization rate of N. paludosum to MG and CV was 77.6% and 35.1% respectively[55]. In terms of degradation of organic dyes by cyanobacteria, Priscila et al evaluated the degradation of three textile dyes (indigo, RBBR and sulfur black) by three cyanobacteria strains Anabaena flosaquae UTCC64, Phormidium autumnale UTEX1580 and Synechococcus elongatus PCC7942[56]. Three kinds of cyanobacteria all showed the potential to repair textile wastewater by removing color and reducing toxicity, but cyanobacteria grew slowly on sludge and low discoloration efficiency. P. UTEX1580 was the only strain to completely degrade indigo dye, and two metabolites, o-aminobenzoic acid and isobenzoic acid, were produced in the degradation process, but the toxicity did not increase after treatment [57].

3.3 Heavy metal

Eukaryotic microalgae is a common kind of algae used to remove heavy metals. Recent studies have found that eukaryotic microalgae can be used to remove heavy metals such as lead, cadmium and copper. Pratiwi et al used Skeletonema costatum to repair Cd and Cu. The average bioconcentration factor (BCF) were 6.15 and 7.97 respectively [58]. Makhanya et al used green algae biofilm and spectrophotometry to determine the physical and chemical parameters and heavy metal removal amount every 24 hours. The results showed that the highest removal rates of iron, zinc and cadmium were 85%, 95% and 99%, respectively, while the removal rates of copper and aluminum were 100% [59]. In terms of cyanobacteria, recent studies have found that cyanobacteria can be used to remove heavy metals such as cadmium and lead. In one study, Nostoc muscorum and Trichormus variabilis were cultured in four Cd solutions with different initial concentrations (0,0.5,1.0 and 2.0mg/L), then found that when the concentration of Cd was 0.5,1.0 and 2.0mg/L, the removal efficiency of N. muscorum was 93.4%, 82.5% and 74.5% respectively, which was better than that of T. variabilis 89.13%, 74.00% and 68.38% [60]. The adsorption effect of lead on Spirulina platensis was detected in the experiments of Homaidan et al.. The results of atomic absorption spectrophotometry showed that the removal rate of lead was higher than 91% [61].

Macroalgae have selectivity for the removal of Cd, Hg and Pb, and the adsorption efficiency reaches 75-99.05%. The priority order of other metal ions absorbed by Sargassum crassifolium is Cu > Fe >

Co > Ni > Cr, but all the removal rates are lower than 56% [62]. The researchers used four kinds of macroalgae Costaria costata, Hizikia fusiformis, Gracilaria verrucosa and Codium fragile to remove Cd. The results showed that C. costata The results showed that the metal ion removal performance of C.costata for cadmium ion, copper ion, nickel ion and lead ion was 20.7%, 42.5%, 9.8% and 9.8% respectively, showing the highest metal biosorption[63]. Another study used Gracilaria sp.to remove Al, Cr and Zn, and cultured under constant light (250 umol / m2s), photoperiod (12:12 L: d cycle) and aeration at 20 ±2 °C. The results showed that the biofiltration capacity of the algae to Al, Cr and Zn was 10.1%-72.6%, 52.5%-83.4% and 36.5% 91.7%, respectively [64].

Recently, some studies have compared the effects of eukaryotic microalgae, cyanobacteria and macroalgae on the removal of heavy metals. In the experiment of Carrera et al., 11 species of algae were used, including six species of macroalgae (Gracilaria sp., Gelidium sp., Sargassum sp., Saccharina latissima, Ulva rigida and Undaria pinnatifida), two species of eukaryotic microalgae (Isochrysis galbana and Phaeodactylum tricornutum) and three species of cyanobacteri(A. cylindrica PCC 200, N. muscorum UTAD_N7122 and Spirulina sp.). The results showed that the biomass adsorption capacity of macroalgae S. latissima, U. pinnatifida and Sargassum sp. was about twice as much as that of microalgae I. galbana and P. tricornutum, while the adsorption capacity of these two eukaryotic microalgae was slightly higher than that of the three cyanobacteria A. cylindrica PCC 200, N. muscorum UTAD_N7122 and Spirulina sp.. Overall, the performance of macroalgae was better than that of eukaryotic microalgae, followed by cyanobacteria [65].

3.4 Radionuclide

The researchers found that eukaryotic microalgae and cyanobacteria can be used as antidotes to treat radionuclide contaminated wastewater, and they have great potential to accumulate radionuclides as nutrients for their intracellular metabolism. For example, Coccomyxa actinabiotis isolated from radioactive wastewater may tolerate a wide range of radiation doses up to 20kGy [66]. Similarly, studies by Rivasseau et al have shown that the algae has almost 100% removal of 137Cs from radioactive contaminated wastewater with an initial Cs concentration of 67Bq/L [66]. In addition, Fukuda et al reported that nearly 188 species of aquatic plants and microalgae were widely used to 137Cs, 125I and 85Sr from the water environment during the Fukushima nuclear power plant accident [67]. Different species of cyanobacteria, namely Spirulina laxissima, Nostoc carneum, Oscillania geminata and Nostoc insulate can remove more than 79% of 134Cs from simulated radionuclide contaminated wastewater [68]. Moreover, the use of different chemical pretreatment techniques, such as phosphorylation, can increase the rate of radionuclide accumulation in dead algae cells. After Mashkani and Ghazvini used H3PO4 to pretreat the dead cells of to pretreat the dead cells of N. carneum and N. insulate, the removal rate of strontium was increased from 72% to 98% and 75% to 86%, respectively [69]. What's more, Fukuda et al used eukaryotic microalgae Parachlorella binoss and three cyanobacteria Nostoc commune, Scytonema javanicum and Stigonema ocellatums to remove 125I from wastewater. In this study, the maximum removal efficiency was 39%, 66%, 62% and 49%, respectively [67].

The scavenging effect of macroalgae on radionuclides is also significant. In the wastewater with initial Cs concentration of 70mg/L, the accumulation of Cs by treated macroalgae Cystoseira indica and Sargassum glaucescens were 96% and 95%, respectively. Dabbagh et al also reported that compared with untreated biomass, pretreatment with algae biomass could increase the accumulation rate of radionuclides by nearly 30 times [70]. The accumulation rates of C. indica and S. glaucescens reached 63mg Cs/g dry weight and 62mg Cs/g dry weight, respectively [70]. In another study, the maximum removal rate of 85Sr by eukaryotic microalgae Parachlorella bino s with alginate extracellular matrix was 76% [71].

3.5 Pesticide

In recent years, the role of eukaryotic microalgae in recovering important nutrients such as phosphorus and nitrogen from secondary wastewater has been studied more and more.

Bioremediation involving the use of eukaryotic microalgae to remove or convert pesticides into harmless or less harmful compounds is becoming a trend [72]. Nasiri et al discussed the potential of Dunaliella salina to remove four organophosphorus pesticides from well water. The results showed that under a certain concentration of pesticide (5 mg/L), D. salina could grow and remove 70.25%, 47.65%, 53.89% and 41.84% diazinon, parathion, probenophos and chlorpyrifos, respectively, indicating that D. salina has the ability to tolerate environmental stress, grow in polluted water and remove pesticides[73]. Scientists studied the synthesis of pyrethroid insecticide zeta-cypermethrin, which is commonly used against Curculio nucum L.. Here, A study explored the effect of zeta-cypermethrin on the growth and bioremediation of C. reinhardtii P.A. Dangeard and Lemna minor L. [74]. It was found that the application of low concentration of zeta- cypermethrin (150μg / L) produced nutritional effects (stimulants), while high concentrations (300-600μg / L) showed toxicity and inhibited growth, so the decomposition rate of species was related to the concentration of pesticides applied to the environment. When the concentration of zeta-cypermethrin was 300μg / L, the removal rate of C. reinhardtii P.A. Dangeard was the highest (98.2%), while the ratio of L. minor L. was lower (35.4%-95.9%)[75].

Cyanobacteria are sensitive organisms and effective degradation bacteria for bioremediation of soil and water pollutants [76]. N.muscorum can use malathion as the only phosphorus source, so it is considered to be a cheap and efficient biotechnology for repairing organophosphorus pesticides in polluted wastewater. In this study, three strains of filamentous cyanobacteria were used to study their growth and utilization efficiency under organophosphorus pesticide malathion. The results showed that N. muscorum had tolerance at different concentrations. It is the strain with the highest biodegradation efficiency of the compound (91%) [77]. There is excessive use of organophosphorus pesticides in agricultural production, such as pyridine phosphorus (PY) to inhibit crop losses caused by insects, resulting in soil and water pollution. Seham et al used two kinds of diazotrophic cyanobacteria (Anabaena laxa and N. muscorum) for comparative study [78]. A. laxa and N. muscorum were designed to be exposed to mild (5mg/L) and high concentration (10mg/L) PY for 7 days to compare the effects of PY toxicity on them. Compared with A. laxa, N. muscorum showed the ability to accumulate and degrade PY into safe environmental products, and 6-hydroxy-2-phenylpyridazin-3(2H)-one could inhibit plant cell growth and reduce the content of Chla and the activity of photosynthesis-related enzymes (PEPC and Rubisco). This ability had little effect on N. muscorum. PY also induced oxidative damage, especially in Anabaena, resulting in a significant increase in the levels of hydrogen peroxide, lipid peroxidation and protein oxidation, as well as the activity of NADPH oxidase. N. muscorum showed obvious induction of antioxidants, that is, induced ascorbic acid and glutathione cycle, but the increase of these antioxidants in A. laxa was not so obvious [79].

4. Conclusion and prospect

This paper comprehensively explains the progress of algae remediation as a new bioremediation technology in the degradation of environmental pollutants, explains its remediation mechanism from two aspects of microalgae and macroalgae. The latest developments in the research on the removal of these pollutants by different algae were summarized. There are still many challenges in the application of algae to bioremediation, including the adaptability and growth cycle limitations of algae, the effects of different kinds and concentrations of pollutants on the growth and reproduction of algae, high economic costs and environmental safety risks, all of which prevent algae remediation from coming out of the laboratory and put into large-scale application. To this end, the research should focus on investigations related to the large-scale development and operation of these technologies, carry out comprehensive environmental assessments, consider the balance between ecological and economic benefits, and adopt appropriate management and monitoring measures, ensure the effect of restoration while reducing the impact on the environment, so as to accelerate the commercialization of this environmentally sustainable technology. In addition, at present, the technology of bioremediation with algae is not mature enough, and we seldom see any research on improving the biodegradability and pollutant tolerance of algae through genetic engineering. It has been reported in plants that efficient and tolerant algae species developed by transgenic technology are expected to emerge in the future[80]. At the same time, we also look forward to seeing the development of efficient removal of algae by CRISPR and other gene editing technology. CRISPR gene editing technology has the advantages of simple operation, low cost and strong specificity [81]. It is believed that combining gene editing technology with algae bioremediation can develop more algae strains for repair. At the same time, we can try to use CRISPR to build an anti-escape system to repair algae strains to prevent alien algae from entering the non-local natural ecosystem.

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