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Application and Prospects of Microalgae-Based Biosorption Materials for Lithium-Containing Wastewater

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Abstract. The scale of lithium consumption in power batteries and energy storage keeps expanding, and salt lake brine, lithium ore processing and battery production generate large amounts of lithium-containing wastewater. Direct discharge causes the loss of lithium resources and also brings the risk of salt pollution in water bodies. Conventional lithium recovery processes such as precipitation and solvent extraction have practical problems of high reagent consumption, insufficient selectivity and high cost for treating low-concentration lithium-containing wastewater. Biosorption technology enriches metal ions through the functional groups on biomass surfaces, and microalgal biomass is readily available and can be cultivated on a large scale, so it has obvious potential as an adsorbent for lithium recovery from lithium-containing wastewater. This paper reviews the adsorption theory of microalgae-based adsorbents, summarizes the research progress of natural microalgae, chemical modification, composite construction and immobilized molding, analyzes the constraints that material properties, water environment parameters, coexisting ions and regeneration conditions place on lithium ion adsorption efficiency, lists the practical bottlenecks of technical implementation at the present stage, and finally makes an outlook for microalgae-based adsorption materials in the direction of resource recovery from lithium-containing wastewater.

Keywords: lithium-containing wastewater, lithium recovery, biosorption, microalgae, modification

1. Introduction

1.1. Sources of lithium-containing wastewater, pollution characteristics and the significance of lithium resource recovery

With the rapid development of the lithium battery industry chain, lithium raw material mining, battery material synthesis and the hydrometallurgical treatment of spent lithium batteries all produce lithium-containing wastewater. Besides lithium ions, such wastewater usually accompanies sodium, calcium, magnesium, potassium and other alkali and alkaline earth metal ions, the ion composition is complex, the lithium concentration of some wastewater lies in the low-to-medium range, and the salinity is generally high. Direct discharge of lithium-containing wastewater causes on the one hand the wanton loss of lithium as a strategic resource, and on the other hand the high salt components

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also change the original physicochemical conditions of the water body and bring a potential burden to the water environment of the basin.

Recovering and reusing the lithium in the wastewater can ease the supply pressure of lithium mineral resources and can also reduce the impact of wastewater disposal on the external water environment, serving the dual goals of resource circulation and pollution prevention. Exploring low-cost lithium recovery technology that adapts to complex salt systems has therefore become a research direction of attention in the field of lithium-containing wastewater treatment.

1.2. Mainstream lithium recovery technologies for lithium-containing wastewater and their limitations

At present, several technical routes have been formed for lithium resource recovery from water, including precipitation, solvent extraction, membrane separation and adsorption. Chemical precipitation has a low operating threshold, but its treatment effect on wastewater with low lithium concentration is limited, and it also produces a large amount of solid sludge; solvent extraction separates lithium well, but the organic extractant is easy to lose and the operating cost is high; membrane separation occupies little area, but under high salt conditions the membrane module is prone to pollution and scaling, and the maintenance cost is large.

Adsorption relies on a solid adsorbent to selectively enrich lithium ions and is more suitable for the treatment of low-concentration lithium-containing wastewater. Traditional inorganic adsorbents have shortcomings such as high preparation cost and cumbersome regeneration procedures, so finding new adsorbent media with a wide source and controllable cost is of key importance for promoting the recovery of lithium as a resource from lithium-containing wastewater.

1.3. Background of microalgae-based biosorbent development and a brief review of the current research

Microalgae are primary photosynthetic organisms that are widely distributed, their culture substrates are simple and their proliferation is fast, and a large number of studies have confirmed that microalgal biomass can biosorb many kinds of metal ions in water [1]. Compared with other biomass adsorbent raw materials, microalgae can use the nitrogen and phosphorus in the wastewater to complete their own proliferation and have the advantage of simultaneously assimilating and enriching pollutants.

However, the adsorption capacity of raw microalgal biomass for lithium ions is limited, and facing actual lithium-containing wastewater with high salinity and coexisting multiple ions, adsorption selectivity and cycling stability still have obvious shortcomings. Existing studies at home and abroad have mostly carried out experimental exploration around algal species screening, chemical modification, composite carrier construction and cell immobilization, trying to improve the adsorption performance of microalgal materials [2]. So far most experiments are still at the stage of laboratory simulated wastewater, and there are still many practical problems to be solved before the industrial treatment of lithium-containing wastewater.

DOI: 10.54254/2755-2721/2027.39149

2. Fundamentals of microalgal biosorption and the characteristics of lithium-containing wastewater systems

2.1. Component characteristics, ionic composition and recovery difficulties of typical lithium-containing wastewater

The water quality of lithium-containing wastewater from different sources differs markedly. Salt lake wastewater has extremely high salinity, with sodium and magnesium ion contents far above lithium ions; lithium battery production wastewater has large fluctuations in lithium concentration and is mixed with calcium, potassium and some heavy metal impurities. The main difficulties of lithium recovery concentrate on the following points: the hydrated radius of lithium ions is large and is close to the physicochemical properties of many coexisting alkali metal ions, so selective enrichment of lithium is difficult to achieve; the high salt environment brings a marked ion competition effect; under low lithium concentration conditions, the driving force for mass transfer in the adsorption process is insufficient, and ordinary adsorbents find it hard to capture the ions efficiently.

2.2. Microalgal cell structure, surface functional groups and the basic principle of biosorption

The microalgal cell wall consists of polysaccharides, proteins and lipids, and a large number of active functional groups such as carboxyl, hydroxyl and amino groups are distributed on the surface, which is also the main site where biosorption occurs. Biosorption can be divided into two processes: extracellular passive adsorption and intracellular active enrichment. Passive adsorption does not consume metabolic energy and relies on the groups on the cell wall surface to complete ion binding; active enrichment requires the metabolism of algal cells and transfers metal ions into the cell for storage. Microalgal biomass after inactivation still keeps the complete cell wall functional groups, can carry out passive biosorption, is not constrained by the survival environment, and is more commonly used in wastewater treatment experiments [3].

2.3. Main adsorption mechanisms of lithium ions on the microalgal surface

The capture of lithium ions by microalgae takes surface complexation, ion exchange and electrostatic adsorption as the main routes. Lithium ions can directly complex with functional groups such as carboxyl and hydroxyl groups on the cell wall and generate surface complexes; in the ion exchange process, Li^+ in the solution displaces cations such as H^+ and Na^+ bound to the cell wall, completing the transfer of lithium to the solid phase; a change of solution pH changes the charged state of the algal cell surface and further affects the strength of electrostatic adsorption. In real wastewater systems, the above mechanisms mostly work at the same time, only the proportion of the contribution of each mechanism differs under different working conditions [1].

2.4. Adsorption evaluation models

Kinetic models and adsorption isotherm models are two kinds of commonly used tools for evaluating the adsorption process. Pseudo-first-order and pseudo-second-order kinetic models are used to describe the change of adsorption rate and to judge whether the adsorption process is dominated by physical diffusion or by chemical binding; Langmuir and Freundlich isotherm models are used to fit the change of adsorption capacity under different lithium concentration conditions. The Langmuir model assumes that adsorption takes place at uniform sites of a monolayer, and the

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theoretical maximum adsorption capacity can be calculated; Freundlich is an empirical model and is mostly used to describe the adsorption behavior of heterogeneous surfaces. With the help of model fitting results, the adsorption performance of different microalgal materials can be compared, and reference can be provided for the optimization of process parameters [1].

3. Construction of microalgae-based lithium adsorption materials and research progress on adsorption performance

3.1. Adsorption characteristics of natural microalgal biomass for lithium ions in water

Microalgae, with their own unique physiological characteristics, show good application potential in the field of metal purification from water. As unicellular or simply multicellular photosynthetic organisms, microalgae proliferate fast and have tolerance for some extreme water environments. Facing metal stress, microalgae can complete the enrichment of metal ions in water through extracellular adsorption, intracellular detoxification, polypeptide chelation and other means, which also provides a material basis for the treatment of lithium-containing wastewater [3].

The adsorption of lithium by raw microalgae relies mainly on the functional groups of the cell wall to achieve ion exchange and surface complexation. The algal species is the core factor determining the adsorption capacity of natural microalgae; the polysaccharide and protein components of the cell walls of different microalgae differ, the numbers of functional groups and the specific surface areas differ, and the lithium adsorption performances finally shown are not the same. As can be seen from Table 1, the lithium adsorption performance of different natural microalgae differs significantly; Dunaliella salina has a better adsorption capacity than Spirulina under suitable conditions, but the overall adsorption capacity is still at a low level [4, 5]. Natural microalgae generally face shortcomings such as poor selectivity in high salt environments and the difficulty of solid-liquid separation of powdered material; relying only on the raw algal species makes it hard to apply directly to the actual treatment of lithium-containing wastewater, and modification, composite construction and immobilization are needed to further optimize the material performance.

Published data show that Dunaliella salina, at an initial lithium mass concentration of 20.0 mg/L and pH=9.0, reaches a maximum lithium adsorption capacity of 4.003 mg/g with dry algal biomass; Spirulina at pH=11 has a maximum lithium adsorption capacity of only 1.75 mg/g [4, 5]. Dead microalgae are not constrained by survival conditions, cover a wider temperature range and are more stable in actual treatment; increasing the dosage of microalgae raises the total number of active adsorption sites in the system and improves the overall lithium removal rate to some extent, but the adsorption capacity per unit mass of biomass does not necessarily increase at the same time [6].

But relying only on natural microalgal biomass for the treatment of lithium-containing wastewater also has obvious shortcomings: the lithium adsorption capacity of the raw algal body is low overall; in a high salt environment with coexisting multiple ions the anti-interference ability is weak; after adsorption is completed, the algal powder particles are fine and the solid-liquid separation is difficult, and the effect of repeated cyclic use of the material is not ideal. Natural microalgae alone can hardly meet the practical needs of treating lithium-containing wastewater, and further optimization by modification, composite construction and other means is needed [1, 4-6].

DOI: 10.54254/2755-2721/2027.39149

Table 1. Comparison of the Li^+ adsorption performance of different natural microalgae (simulated wastewater system)

Algal species	Test conditions	Maximum adsorption capacity /(mg·g^{-1})	Main limiting problem	Reference
Dunaliella salina	Initial Li 20.0 mg/L, pH=9.0	4.003	Ion competition under high salt, difficult solid-liquid separation	[4]
Spirulina	pH=11	1.75	Low adsorption capacity, weak anti-interference ability	[5]
Oocystis	Initial Li 200 mg/L, neutral pH	–	Good adaptability to high lithium concentration, insufficient selectivity	[6]

3.2. Chemical modification of microalgae: acidification, alkalization and grafting modification to strengthen adsorption performance

The adsorption capacity of natural microalgal biomass for metal ions is limited, complex wastewater working conditions also reduce the stability of the adsorption process, and relying only on the raw algal body is hard to support large scale engineering application. Carrying out chemical modification and adjusting the physicochemical characteristics of the microalgal surface is an important research direction for improving the adsorption performance of microalgae.

Acidification modification is a rather common chemical treatment method. Acid treatment can change the state of the surface functional groups of microalgae, the absorption peaks of some functional groups shift, the electronegativity of the groups increases, and the binding ability to cations is strengthened in this way. Existing studies have used hydrochloric acid to treat the cyanobacteria of Lake Taihu, and after treatment the adsorption rate of the material for the target substance in the pH 2.5-5.5 range is close to 80%, far above the adsorption level of 40% for the unmodified raw algae [7]; the increase in the concentration of acid radical ions such as NO_3^-, Cl^-, SO_4^{2-} and PO_4^{3-} in the system can further amplify the adsorption effect of acid modified microalgae.

Alkalization modification is likewise often used for the improvement of microalgal biomass. Alkaline treatment hydrolyzes some components inside the algal cell wall and exposes more active sites. A study has modified kelp with NaOH, and after modification the copper ion adsorption of the material reaches 88.0 mg/g, clearly better than the adsorption level of 59.3 mg/g for the raw algae and 38.6 mg/g for the hydrochloric acid modified sample [8]; the dried Sargassum fusiforme pretreated with alkali shows a good removal effect for Cu, Zn and Cr in electroplating wastewater [9]. Alkali treatment can also improve the adsorption selectivity of microalgae in systems with coexisting multiple cations; the blue algae modified by 0.1 mol/L NaOH show an improved selective adsorption capacity for the target cation under Ca^{2+} and Mg^{2+} interference [10].

The acidification and alkalization processes are simple to operate and the material modification cost is controllable, but the action mechanisms of the two are different. When modification is actually carried out, the suitable modification means need to be selected in combination with the ionic composition of the wastewater and the background pH condition of the water body. Besides acid and alkali modification, grafting modification attaches specific functional groups to the microalgal surface through chemical reactions and introduces in a directed way the groups that have affinity for lithium, although at the present stage the experimental cases of grafting modification for the lithium system are still relatively few.

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3.3. Composite adsorption materials of microalgae (inorganic mineral / polymer carrier composites)

Acid and alkali modification only make a chemical level change to the microalgal biomass itself. To further amplify the adsorption efficiency and at the same time solve the problems of difficult recovery of algal powder and poor cycling performance, building composite adsorption materials from microalgae and other carriers is an important idea. Loading the microalgae onto the carriers improves the mechanical strength of the material and makes repeated adsorption-desorption cycles possible, which is of practical significance for lowering the application cost and promoting the implementation of the technology.

Inorganic mineral composite selects natural minerals such as zeolite, montmorillonite and diatomaceous earth to combine with microalgae. These minerals have well developed pores and large specific surface area by themselves, and carry a large number of surface active sites. After the two are composited, the mineral acts as the attachment carrier of the microalgae and improves the overall mechanical performance of the material, and it can also produce an adsorption synergistic effect with the microalgae. Existing studies have constructed algae-clay mineral-sodium alginate composite immobilization systems, in which the mechanical stability of the gel is enhanced, the lead removal performance of the material is improved and repeated cycling is supported [11]; the montmorillonite and Chlorella composite system can induce the formation of basic lead carbonate precipitates on the surface and raise the tolerance level of microalgae to heavy metals [12]. Transferred to the lithium-containing wastewater scenario, the mineral itself can take part in the lithium adsorption process, the microalgae provide biologically active sites, and the synergy of the two gives hope of improving the lithium enrichment effect under high salt conditions, but at present the systematic experiments of microalgae-inorganic mineral composite materials for lithium ions are still few.

Polymer carrier composite generally uses polymer materials such as sodium alginate, chitosan and polyvinyl alcohol to embed the microalgae into a gel network. Polymer materials have good biocompatibility, their internal porous structure is favorable to ion diffusion, and they can also reduce the loss of microalgal cells. Existing related studies have used sodium alginate to embed marine diatoms to prepare adsorption beads for removing Pb^{2+} from water; the test results show that the concentration of the sodium alginate solution affects the adsorption rate of the beads, and under the condition of 2% sodium alginate and a dosage of 5.0 g/100 mL algal powder the formed beads have the best overall performance, with a maximum theoretical adsorption capacity of the material reaching 909.09 mg/g under these conditions, and the adsorption process conforms to the Langmuir model and the pseudo-second-order kinetic model [13]. The idea of polymer embedding can be applied directly to the treatment of lithium-containing wastewater, and the gel ratio and crosslinking conditions need to be optimized again for the lithium ion system so as to balance mechanical strength and internal mass transfer efficiency.

3.4. Preparation and molding process of immobilized microalgal adsorption systems

Directly adding microalgal powder into the wastewater makes the subsequent solid-liquid separation workload large; at the same time the fine algal bodies are easily lost with the flow. With the help of the immobilization process, restricting the microalgal cells inside the carriers or on the carrier surface can solve the above problems. At the present stage the mainstream immobilization means are divided into three categories: adsorption method, entrapment method and crosslinking method.

DOI: 10.54254/2755-2721/2027.39149

The adsorption method is also called passive immobilization. It relies on electrostatic attraction, hydrogen bonds and hydrophobic interaction to let the microalgal cells attach spontaneously to the surface of the carrier and form a biofilm. The preparation process is not costly, the mass transfer resistance of the system is small, and the activity of the algal cells can be retained to a good extent. But this mode has a weak resistance to shock loading, and the biofilm tends to fall off when the pollutant concentration is high. The method is more suitable for the treatment of low concentration wastewater and for the cultivation of biomass on a large scale.

The entrapment method belongs to active immobilization. The operation is to dissolve the polymer carrier first, then mix it fully with the microalgal suspension, and then form it through a crosslinking reaction, so that the algal cells are held in the three-dimensional gel network of the polymer. Entrapment and molding can build a buffer microenvironment for the microalgae and weaken the shock brought by high concentrations of external pollutants; the material has good performance for reversible use, and it is also the immobilization method most used in the field of wastewater treatment. The disadvantage is that the molding conditions need precise control, and a certain mass transfer resistance arises inside the gel. Research on entrapping Chlorella pyrenoidosa with sodium alginate and calcium chloride shows that 2% sodium alginate combined with 3% calcium chloride gives alginate beads with the best overall performance in bead forming, mechanical strength and resistance to disintegration [14].

The crosslinking method relies on the active groups of the crosslinking agent to form covalent crosslinks with the functional groups on the surface of the microalgal cells and realize the fixation of the cells. The operating steps are simple, but the crosslinking reaction is relatively violent, easily damages the structure of the algal cells, and interferes with the subsequent adsorption and cyclic reuse effect of the material. Before choosing this process, the actual working conditions of the wastewater need to be fully investigated and the crosslinking reagent and reaction parameters need to be selected carefully.

The molding process directly determines the mechanical strength, the ion transfer efficiency and the cycle life of the immobilized microalgal adsorbent. Existing test data show [15] that after immobilization the heavy metal removal rate of the microalgal material can exceed 89%, the nitrogen and phosphorus removal rate lies in the 70%-89.7% range, and the overall running cost can drop by 30%-40% compared with the suspended algae system. Existing results mostly concern the removal of heavy metals, nitrogen and phosphorus, and further tests are still needed to optimize the immobilized microalgal process for lithium-containing wastewater, but this technical path has already shown good application potential.

Fig. 1 shows the four main technical paths for preparing lithium adsorption materials from raw microalgal biomass. Natural microalgae are convenient in obtaining raw materials, but their own performance shortcomings are obvious; chemical modification, composite construction and immobilized molding can optimize the material performance from three dimensions, namely surface functional group modification, introduction of synergistic adsorption sites and material molding. Different modification means have their own advantages and disadvantages, actual engineering application can adopt a combined modification strategy, and the adsorption-desorption regeneration cycle performance also needs to be taken into account.

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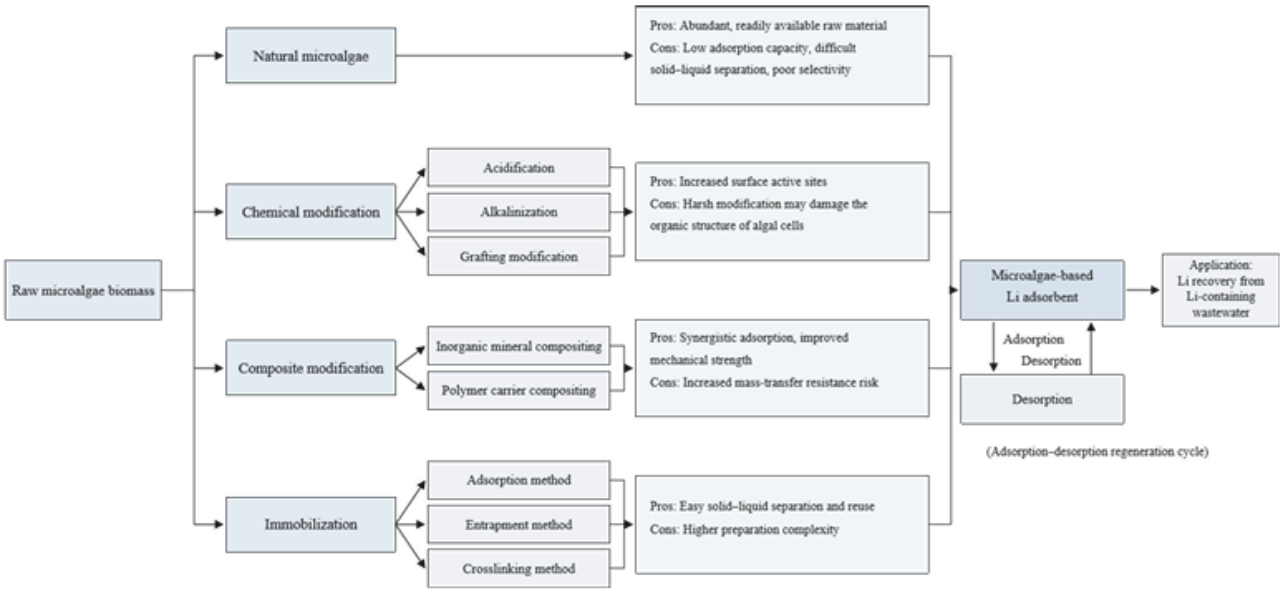

Figure 1. Technical roadmap for development and modification of microalgae-based lithium adsorbents

4. Key factors affecting the adsorption performance of microalgae-based materials for lithium ions

4.1. Properties of the adsorption material itself (algal species, pretreatment, modification method)

The properties of the material itself are the internal cause determining the lithium adsorption performance, which are mainly reflected in three levels: algal species selection, pretreatment means and modification scheme.

The cell wall components of different algal species differ, the types and numbers of the surface functional groups and the specific surface area all differ, and this directly causes the differentiation of lithium adsorption capacity. Both Dunaliella salina and Spirulina have been confirmed to have an adsorption effect on lithium ions, but under the same test conditions the adsorption capacity of the two differs clearly [4, 5]. Screening the algal species adapted to high salt lithium-containing wastewater is the basic work for the subsequent modification and composite research.

Pretreatment is divided into physical pretreatment and chemical pretreatment. Physical treatment such as drying can adjust the pore structure of the algal body and raise the adsorption capacity of the biomass for metal ions; chemical pretreatment such as acidification and alkalization can modify the surface functional groups of microalgae and change the surface charging characteristics, improving the adsorption effect in this way [7, 8]. The adsorption performance of raw algae without any pretreatment often falls short of the expectation of the experiment.

The modification method directly changes the physicochemical properties of the microalgal surface. Acid modification adjusts the state of the functional groups and raises the electronegativity of the groups; alkali modification hydrolyzes some cell wall components and exposes more active sites, and at the same time optimizes the adsorption selectivity under the condition of coexisting ions. The improvement brought by different modification means is not the same, and some modification processes even have a negative effect on specific ion systems, so the suitable

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modification method needs to be screened in combination with the characteristics of the lithium ion system [7-10].

4.2. Water environment conditions: effects of pH, temperature, initial lithium concentration and salinity

The working condition of the water environment belongs to external conditions, and interferes with the adsorption effect of microalgae-based materials on lithium ions from several dimensions.

pH is the most critical influencing parameter. On the one hand it changes the degree of ionization of the functional groups of the extracellular polymeric substances of microalgae and adjusts the charged state of the particle surface; on the other hand it also changes the form in which the ions exist in the solution. When pH is low, the carboxyl and hydroxyl groups of the cell wall are protonated in large numbers, the surface of the material carries positive charge, forms electrostatic repulsion with the cation Li^+, and the adsorption capacity declines; when pH rises, the functional groups are deprotonated, more binding sites are provided, and the adsorption of lithium is favored. But when pH is too high, lithium hydroxide precipitation is easily generated inside the solution, which interferes with the test results. The suitable pH range for the adsorption of cations by most microalgae lies between 5.0-8.5, and the optimal pH for lithium ions needs to be determined further by experiments in combination with the specific algal species and the modification method [1, 4].

Temperature mainly changes the adsorption reaction rate and the adsorption equilibrium state. Within a suitable range, raising the temperature speeds up the molecular motion, which is favorable to the diffusion of lithium ions toward the adsorption sites, and the adsorption efficiency is improved. Related tests on Chlorella and Oocystis borgei show a better adsorption effect in the range of 20-30 °C [6, 12]. Once the temperature exceeds a critical value, if a live algae system is used, the high temperature causes enzyme inactivation and inhibits cellular physiological activity; in an inactivated algae system the too high temperature may also destroy the organic structure of the biomass and cause a decline of adsorption performance.

The initial lithium concentration determines the magnitude of the mass transfer driving force in the adsorption process. If the initial lithium concentration is low, the driving force for mass transfer is insufficient and the adsorption capacity per unit is not high; as the lithium concentration keeps rising, the adsorption capacity per unit rises at first, and after the overwhelming majority of the surface sites are occupied, continuing to raise the lithium concentration no longer makes the adsorption capacity grow clearly. The optimal initial lithium concentrations corresponding to different algal species differ greatly, the optimal initial lithium concentration in the Dunaliella salina test system is 20.0 mg/L, while the optimal initial lithium concentration in the Oocystis system can reach 200 mg/L [4, 6], which also reflects that the ability of different microalgae to adapt to lithium concentration is different.

Salinity represents the ionic strength of the system. Lithium-containing wastewater generally contains Na^+, K^+, Ca^{2+} and Mg^{2+}, and the large number of cations compete for the limited adsorption sites on the microalgal surface. As the ionic strength keeps rising, the number of effective sites available for binding lithium is compressed, and the lithium removal efficiency of the material declines accordingly. Relevant calcium alginate adsorption tests can corroborate this: when the ionic strength drops from 0.5 mol/L to 0.005 mol/L, the removal rate of the target metal rises from 80% to 95% [16], which is enough to show that the interference brought by salinity cannot be ignored.

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4.3. Competitive adsorption mechanism of coexisting metal ions and the pattern of selective interference

The lithium-containing wastewater under actual working conditions does not contain only lithium ions; the system contains a large amount of cations such as Na^+, Mg^{2+}, Ca^{2+} and K^+, and the site competition brought by the coexisting ions is a major obstacle constraining the recovery of lithium by microalgal materials.

The essence of competitive adsorption is that several kinds of cations compete for the limited number of active binding sites on the microalgal surface. The strength of the interference of the coexisting ions is related to the ionic radius and the charge density; the closer the radius is to that of Li^+, the stronger the competitive adsorption capacity. Existing tests on the adsorption of zinc by Spirulina show that Mg^{2+} and Na^+ bring a significant inhibition, Ca^{2+} and K^+ also reduce the adsorption capacity, but the degree of inhibition is relatively weak [5].

The degrees of interference caused by different cations are not equal. Under fixed pH conditions the total amount of functional groups on the microalgal surface is a constant; when the salinity rises there are more cations in the system, and the binding opportunities allocated to lithium ions are squeezed out. In high salt lithium-containing wastewater the content of magnesium and sodium is far above that of lithium; how to weaken the competitive effect of the two and raise the selective adsorption performance of microalgal materials for lithium is a problem that the subsequent research needs to tackle.

4.4. Effect of the adsorption-desorption cycle conditions on the regeneration performance of the material

Whether the adsorbent can be regenerated stably directly decides whether the material can be put into actual use. An ideal regeneration process needs a high desorption efficiency and simple operation, and it also needs that repeated treatment does not seriously damage the structure of the adsorbent itself. Acid washing, alkali washing, salt washing and chelator elution are several commonly used desorption means.

Dilute acid reagents such as dilute hydrochloric acid and dilute nitric acid are applied rather widely, relying on the ion exchange between H^+ and the Li^+ already bound to the adsorption sites to realize the desorption of lithium. Some seaweed adsorption materials use 0.1 mol/L HCl for desorption, and after three adsorption-desorption cycles they still maintain a good adsorption capacity [16]. But strong acids and strong alkalis are quite destructive; after several cycles they gradually destroy the organic structure of the microalgal biomass and cause an irreversible decline of the material performance.

Salt washing uses NaCl and $CaCl_2$, relying on high concentration cations to achieve ion exchange desorption; EDTA-2Na relies on strong chelation to strip the metal ions from the sites. Existing studies have compared the effects of several desorption reagents on Chlorella pyrenoidosa, where the desorption effect of EDTA-2Na is outstanding and the damage to the algal cell structure is very small [17]; the disadvantage is the high price of the chelator and the complicated subsequent treatment of the eluate. The design of the desorption process needs to weigh several dimensions, namely the desorption efficiency, the material damage, the reagent cost and the difficulty of recovering lithium from the eluate, in order to select the adapted desorption agent and operating parameters.

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5. Existing bottlenecks and optimization paths of microalgae-based adsorption materials in application

Although microalgae-based adsorption materials for treating lithium-containing wastewater have already achieved many experimental results under laboratory conditions, really applying them to the treatment of industrial lithium-containing wastewater still faces multiple practical bottlenecks.

First, the lithium adsorption capacity and selectivity are insufficient. The adsorption capacity of raw microalgae for lithium itself is not high, the large amount of sodium and magnesium ions in the wastewater causes strong competitive interference, the existing modification and composite means still improve the selectivity for lithium only to a limited extent, and the adsorption performance of most materials for lithium is still inferior to that for other heavy metal ions.

Second, the experimental conditions are disconnected from the real wastewater working conditions. Most of the existing literature prepares simulated lithium-containing solutions for the tests, with simple system components; the actual wastewater has high salinity, complex impurities and large fluctuations in water quality, and the excellent materials screened in the laboratory often show an obvious decline in performance once they are moved to the real wastewater system.

Third, there are difficulties in the preparation, recovery and regeneration of the material. The solid-liquid separation of powdered algal bodies is troublesome; the immobilized molding material needs to balance mechanical strength and transfer efficiency, and the preparation cost needs to be controlled; after repeated adsorption-desorption cycles the biomass is prone to structural damage and the material performance declines step by step. After desorption the lithium enrichment degree of the eluate is limited, and the subsequent extraction of lithium salt still needs supporting processes.

Fourth, there is the problem of supplying algae on a large scale. Large scale application needs a large amount of microalgal biomass, and the whole set of processes of large scale cultivation, harvesting and drying of the algal species brings material and energy costs; how to obtain a stable microalgal raw material at low cost is also a practical problem that the engineering has to consider.

To optimize the above bottlenecks, work can be carried out in the following directions:

1) Directionally screen and mutagenically acclimatize microalgal species with high lithium affinity and high salt tolerance, with attention to the halotolerant microalgal resources from salt lakes;

2) Develop directional modification strategies, introduce functional groups with specific affinity for lithium, or composite with lithium-sieve type inorganic materials, to improve the anti-ion interference ability of the material and raise the selectivity for lithium;

3) Optimize the immobilized molding process, balance mechanical strength and internal transfer efficiency and lower the preparation cost of the molding material; systematically carry out adsorption-desorption cycle tests, screen low damage and low cost desorption systems, and evaluate the long term cycle life of the material;

4) Push forward the pilot test with real lithium-containing wastewater, simulate the working conditions such as water quality fluctuation and high salt, and verify the actual treatment effect of the material; explore the coupling mode of microalgae cultivation and wastewater treatment, use the nitrogen and phosphorus in the wastewater to feed the microalgae and press down the raw material cost.

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6. Conclusion and outlook

6.1. Summary

Microalgal biomass, with the characteristics of a wide source and a large modification potential, has good development prospects in the field of recovering lithium as a resource from lithium-containing wastewater. Natural microalgae can complete the capture of lithium ions by relying on surface complexation, ion exchange and electrostatic action, but the adsorption capacity of raw algae is not high and the ability to resist high salt interference is poor. Acidification and alkalization modification can modify the surface functional groups of microalgae; compositing with inorganic minerals and polymer carriers, together with immobilization molding means such as adsorption and entrapment, can both improve the adsorption performance and solve the problem of difficult recovery of algal powder.

The adsorption of lithium by microalgae-based materials is jointly constrained by the conditions of the material itself such as algal species, pretreatment and modification, and by the external water environment conditions such as pH, temperature, initial lithium concentration and salinity; the site competition of the coexisting cations in the wastewater and the structural damage of the material during the adsorption-desorption regeneration process are the key factors limiting practical application. At present this direction still has shortcomings such as a selectivity for lithium that needs to be improved, being mostly based on simulated wastewater experiments, and an incomplete large scale preparation and regeneration process.

6.2. Outlook

The follow-up research work can take the development of salt tolerant and highly selective microalgae-based adsorption materials as the core. On the one hand, local salt tolerant algal species resources need to be explored, and chemical modification and composite construction should be combined to strengthen the recognition ability of lithium ions in a directed way and weaken the competitive interference of sodium and magnesium ions; on the other hand, the laboratory simulated solution system has to be left behind, and pilot and semi-industrial studies on real lithium-containing wastewater should be carried out step by step, examining the stability of the material under water quality fluctuations and long term cyclic operation.

At the same time the whole process chain of microalgal biomass production, adsorption treatment and lithium extraction by desorption has to be coordinated, and the coupling mode of "wastewater feeds microalgae - microalgae-based material adsorbs lithium - lithium salt is recovered from the eluate" should be explored so as to control the overall process cost. With the continued improvement of the modification technology and the immobilization process, microalgae-based biosorption materials have the hope of becoming one alternative technical path for the recovery of lithium as a resource from low and medium concentration lithium-containing wastewater.

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