Enhanced degradation of phenol by a novel biomaterial through the immobilization of bacteria on cationic straw

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ABSTRACT

As phenol possesses a threat to human health, there is a great demand to search for fast and efficient methods for it to be discharged. In this study, a novel biomaterial was prepared by the immobilization of bacteria on a cationic straw carrier, and the remediation ability of the biomaterial on phenol-containing wastewater was investigated. The free bacteria could degrade 1,000 mg/L phenol within 240 h, while the prepared biomaterial was 192 h, shortening by 48 h that of free bacteria. In addition, the degradation tolerance of biomaterial increased from 1,000 mg/L to 1,200 mg/L than the free bacteria, within 216 h, which shortened by 24 h the degradation time of 1,000 mg/L phenol by free bacteria (240 h). Further, under different pH conditions, the degradation efficiency of phenol by prepared biomaterial was much higher than that of free bacteria. Especially for the lower pH 5, the degradation efficiency of biomaterial was nearly twice that of the free bacteria. This investigation demonstrates that this biomaterial has great potential in the field of remediation of organic pollution.

Key words: bacteria immobilization, biomaterial, cationic straw, phenol degradation, resistance pH

HIGHLIGHTS

• A novel biomaterial was prepared by the fixation of bacteria on cationic straw.
• The biomaterial can shorten the time and increase phenol concentration for degradation, compared to the free bacteria.
• At a lower pH, the degradation efficiency of biomaterial is nearly twice that of the free bacteria.

INTRODUCTION

Phenol is an important industrial material, but its high biological toxicity results in a serious environmental and health threat for the whole world (Stoilova et al. 2006; Nikl et al. 2019). As the extensive applications and pollution in the fields of oil refining, coking, and papermaking, and excellent water solubility of phenol, solutions for phenol-containing wastewater and soil pollution must be raised (Saha et al. 1999; Michalowicz & Duda 2007; Naguib et al. 2019). At present, physical adsorption, biodegradation, chemical catalysis and solvent extraction are the main treatment for phenol-containing wastewater (Yang et al. 2018; Liu et al. 2019). Because of the advantages of complete degradation and no secondary pollution,
biodegradation is widely regarded as a practical, economical and promising method (Azadi & Shojaei 2020; Lee et al. 2020; Nogina et al. 2020). Various kinds of microorganisms with the ability of degradation of phenol have been favored, such as Pseudomonas, Acinetobacter, yeasts, and activated sludge with a wide variety of microorganisms (Filipowicz et al. 2020; Lin & Cheng 2020; Liu et al. 2020; He et al. 2021). In general, as the concentration of hazardous pollutants increases, the ability of natural microorganisms for degradation is decreased. Except for high concentrations of phenol, the inevitable factors in wastewater such as adverse pH, temperature, and metal ions, which poison the growth of microorganisms, have significant effect on the removal of phenol (Nouri et al. 2020; Rongsayamanont et al. 2020; Barik et al. 2021). Thus, the development of methods to rapidly and efficiently remove phenol in a high concentration that can also maintain or improve the survival and degradation efficiency of bacteria in a harsh pH or temperature condition, is very necessary.

The immobilization of microorganisms which can protect microorganisms or cells from high levels of pollution is an alternative strategy to facilitate the degradation (Zhao et al. 2020). Many carriers have been studied for the immobilization, including inorganic materials, polymers, biochar, natural biomass, and so on (Lv et al. 2016; Basak et al. 2019; Xia et al. 2019; Swain et al. 2020; Zolair et al. 2020). Biomass and its derivatives have been widely studied as immobilized carriers of microorganisms, due to their non-toxicity, excellent mechanical strength, structural stability, and permeability, which allows the penetration and diffusion of substrates and metabolites (Zhao et al. 2020). Straw, with its large annual output, is a representative biomass material, which has micro-porous structure and large specific surface area (Xu et al. 2011a, 2011b, 2011c). The large number of active groups such as hydroxyl groups and carboxyl groups distributed on its surface, can realize the chemical modification of straw materials, to introduce new functional groups and improve their reactivity (Xu et al. 2011a, 2011b, 2011c). So far, straw is directly used as the carrier to fix microorganisms, but modified straw with cationic groups, which can more efficiently immobilize microorganism through electrostatic interaction, has not been reported (Xu et al. 2011a, 2011b, 2011c).

Herein, a novel composite material of bacteria immobilized on cationic straw through electrostatic interaction between negative Pseudomonas aeruginosa and positive straw grafted by amino groups, for the bioremediation of phenol, has been prepared. This strategy possesses some advantages. Cationic straw was firstly applied to fix bacteria to degrade phenol. Comparing with the free bacteria, the immobilized bacteria can effectively shorten the degradation time, increase the degradation rate, and enhance the degradation concentration of phenol. In addition, under some harsh environments, especially low pH, fixed bacteria can more effectively degrade phenol. Further, microorganisms with different abilities can be employed to degrade various kinds of pollution. It should be noted that using modified biomass materials to immobilize microorganisms is an effective, green and promising method for the treatment of polluted wastewater, providing the unity of social benefit, economic benefit and environmental benefit.

MATERIALS AND METHODS

Chemicals

The corn straw was taken from the test field of Jilin Agricultural University. Ethylenediamine (99%, v/v) and triethylamine (99%, v/v) were from Beijing Chemical Works. Phenol (99%, w/v), epichlorohydrin (99%, v/v), N, N-dimethylformamide (DMF) (99.5%, v/v), 4-aminooantipyrine (98.5%, w/w) and potassium ferricyanide (99.5%, w/w), were the products of Tianjin Hengxing Chemical Preparation Co. Ltd, Xian Regeatn, Sinopharm Chemical Reagent Co. Ltd, Shanghai Yuanye Biotechnology Co. Ltd and Tianjin Kermel Chemical Reagents Development Center, respectively. Pseudomonas putida (21906) was purchased from China Industrial Microbial Species Preservation Management Center.

Preparation of cationic straw

The cationic straw was prepared according to the literature (Xu et al. 2010a, 2010b) with some modifications. Herein, corn straw was used as a modified biomass material to replace wheat straw in the reported method. Firstly, after drying and crushing, straw with a diameter of 75 μm was obtained, followed with washing with distilled water and drying in the oven at 65 °C for 24 h, to obtain clean straw powder. In order to efficiently immobilize bacteria, the straw was crushed to a smaller size of 75 μm before modification, different to the 250 μm (Xu et al. 2010a, 2010b). Then, 4 g straw was reacted with 20 mL 99% epichlorohydrin (w/w) and 20 mL 99.5% N, N-dimethylformamide (w/w) in a 250 mL round-bottom flask at 85 °C for 60 min. After that, 4 mL 99% ethylenediamine (w/w) was added and reacted at 85 °C for 60 min, following the addition of 20 mL 99% triethylamine (w/w) with a reaction time of 60 min. Finally, after cooling to room temperature, the straw was
filtered and washed with ethanol three times, and washed with distilled water until the filtrate was neutral, then dried at 80 °C to a constant weight (Swain et al. 2020).

**Culture, acclimatization and phenol degradation of bacteria**

Luria-Bertani (LB) medium (tryptone 10 g/L, NaCl 10 g/L, yeast extract 5 g/L, deionized water 1 L) and minimal salt medium (MSM) (glucose 5 g/L, NH₄NO₃ 1 g/L, KH₂PO₄ 0.5 g/L, Na₂HPO₄ 1.5 g/L, NaCl 1 g/L, MgSO₄·7H₂O, deionized water 1 L) were used for activation and acclimation, respectively. The medium was treated with autoclave sterilization at 121 °C for 20 min. The pH of both media was adjusted to 7.0 with 6 mol/L NaOH aqueous solution and 6 mol/L HCl aqueous solution (Umashankar et al. 2018).

The acclimation process of the strain was carried out in MSM by gradually increasing the concentration of phenol 100 mg/L per three days and decreasing the concentration of glucose in MSM medium (100 mg/L) per three days, until the concentration of glucose reduced to 0 mg/L and phenol increased to 1,000 mg/L. The acclimation was accomplished as inoculation 10% (v/v) at 37 °C and 180 rpm. Then, the strain was used to degrade 1,000 mg/L phenol, as the sole carbon source with several circulations, to obtain a stable bacteria. Finally, the bacteria were activated in MSM medium for phenol degradation, inoculated for 10% (v/v) and cultured at 37 °C and 160 rpm. The concentration of phenol was detected by 4-aminoantipyrine method (Baird et al. 2017).

**The immobilization of the strain and phenol degradation**

The fermentation broth with the strain was centrifuged at 5,000 rpm for 20 min, then the supernatant was discarded, followed by washing with sterile water. After that, the strain was re-suspended with sterile water, whose optical density was 1.0 ± 0.13. The linear relationship between the number of bacteria and the optical density can be obtained by the spread plate method. After the sterilization of cationic straw, the straw and bacterial suspension were mixed at a solid-liquid ratio of 1:100 at 30 °C and 120 rpm for 6 h. The amount of immobilized bacteria was determined by the above linear curve (Yue et al. 2013). The number of immobilized bacteria on straw was 1.3 x 10⁹ CFU/g. After filtration, the immobilized material was mixed with 5% (w/w) trehalose, then freeze dried to constant weight and stored at 4 °C (Xu et al. 2010a, 2010b). For phenol degradation, the immobilized material was added into MSM medium where phenol acts as the sole carbon source at a solid-liquid ratio of 1:100. In addition, degradation with phenol concentrations of 1,000 and 1,200 mg/L were carried out, respectively.

**The effect of pH on phenol degradation**

After sterilization, pH of the culture medium was adjusted to 5, 6, 7, 8 and 9 under aseptic conditions, respectively. Then, strains and immobilized material were added in the MSM medium, respectively, to degrade phenol at 37 °C for 5 days. Then, the culture medium was centrifuged, and the supernatant was used to determine the concentration of phenol.

**Measurements**

Zeta potential data were recorded on a Malvern Instruments Zetasizer Nano ZS. Fourier transform infrared (FTIR) data were performed on a Bruker IFS66 V FTIR spectrometer equipped with a DGTS detector (32 scans), using KBr pellets, and the spectra were recorded with a resolution of 4 cm⁻¹. Element analysis was carried out on a Flash EA1112 analyzer from ThermoQuest Italia S.P.A.

**RESULTS AND DISCUSSION**

**Characterization of cationic straw**

Straw is a common agricultural waste, whose utilization is not effective. However, straw possesses many functional groups, such as carboxyl, hydroxyl and other groups, which provide possibilities for further modification. Herein, a kind of cationic straw was synthesized after the reaction between epichlorohydrin and ethylenediamine by using triethylamine as modifying agents, as shown in Figure 1(a). Compared with untreated straw, a new peak at 1,332 cm⁻¹ belongs to C-N stretching vibration of amine groups, proving the successful graft reaction between the amine reagent and cellulose, as shown in Figure 2(a). While the appearance of a band at 1,392 cm⁻¹ associates with N-H groups, further explaining the introduction of amine groups (Xing et al. 2011). The band C-Cl appears at 618 cm⁻¹, which further indicates that epichlorohydrin successfully grafts on the straw in the amination process and plays the role of intermediate substitute in the reaction process (Ma et al. 2013). In order to understand the degree of amination modification, the elemental analysis was also carried out to compare the elemental changes of straw before and after amination treatment. Compared with original straw (Supplementary
Material, Table S1), the C and H amount in modified straw are increased, while the N content is significantly increased from 0.58% to 8.79% (Xu et al. 2011a, 2011b, 2011c; Bera & Mohanty 2020). In the process of preparing cationic straw, the amination reagent substitutes hydroxyl groups, resulting in a large number of nitrogen-containing groups grafting onto the straw, which is consistent with the FTIR results. Zeta potential was used to analyze the surface charge properties of the material. The zeta potential decreases with the increase of pH, which is attributed to the functional groups such as hydroxyl and carboxyl, which show larger negative charges at higher pH (Umashankar et al. 2018). Compared with the original straw, the zeta potentials of modified straw are much higher, indicating the existence of increased positive-charge functional groups and the successful graft of amino groups on the straw (Figure 2(b)). In addition, the original straw has a negative charge in the range of pH 2–12, while the isoelectric point of the cationic straw is 8.89, demonstrating that the surface of the straw is positively charged at pH lower than 8.89 and indicating the ability of this straw as a novel anion adsorption material (Yue et al. 2013).
The degradation of the free strain
Among the various methods for phenol treatment, microbial degradation is welcome due to its advantages of a high removal rate, more thorough degradation, and no secondary pollution to the environment. *Pseudomonas putida* is a kind of microorganism for effective degradation of phenol, which can grow on the culture medium where phenol acts as the sole carbon source (Umashankar *et al.* 2018). As the initial strain we used could not survive on the sole carbon source of phenol, acclimation of the strain was raised by gradually increasing the concentration of phenol and decreasing the concentration of glucose medium. In order to study the degradation ability of the acclimated strain, phenol degradation at different initial concentrations from 500 to 1,250 mg/L was investigated, as shown in Figure 3. The highest degradation concentration of the strain is 1,000 mg/L. With the increase of the concentration, the delay period and the degradation time of phenol gradually increase. At the concentration of 1,000 mg/L, the bacteria could totally remove phenol within 240 h, while for 500 and 750 mg/L phenol, 144 h is used. With the rising phenol concentration, both the degree of poisoning of bacteria and the inhibition of enzymes are increased. Thus, the bacteria need more time to adapt to a high concentration of phenol and improve the number of bacteria, resulting in a slower degradation rate. In addition, when the phenol concentration exceeds 1,000 mg/L, the bacteria are beyond sufferance and death occurs.

The degradation of biomaterial for phenol
Although the microbial degradation has a good ability to remove phenol, it is vulnerable to stress from some factors in practical application, such as heavy metal ions, unsuitable environmental pH and temperature. Bacteria immobilization that fix the bacteria on the carrier with protective effect, is a good approach to reduce or avoid the influence of adverse factors. Herein, bacteria have been immobilized on the cationic straw through electrostatic interaction, whose number was $1.3 \times 10^9$ CFU/g. When the initial concentration of phenol is 1,000 mg/L, the degradation time of immobilized bacteria is 192 h, while the free bacteria is 240 h, as shown in Figure 4(a). Compared with the free bacteria, the immobilized bacteria make the degradation time shorten by 48 h. In addition, free bacteria could not tolerate the concentration of phenol at

![Figure 3](image)

**Figure 3** | Degradation of strains at different initial phenol concentration.

![Figure 4](image)

**Figure 4** | The degradation of immobilized and free bacteria at the concentration of (a) 1,000 mg/L and (b) 1,200 mg/L, respectively.
1,200 mg/L, while the immobilized bacteria could degrade phenol within 216 h, which is less than the degradation time of 1,000 mg/L phenol by free bacteria (240 h), shortening by 24 h (Figure 4(b)). Thus, the immobilized bacteria on cationic straw are not only more tolerant to a higher concentration of phenol, but also shorten degradation time, compared to the free bacteria. Moreover, due to the porosity of straw, it has the ability to adsorb phenol, but the adsorption is limited. At the initial concentration of 1,200 mg/L phenol, the adsorption capacity of cationic straw is 303 g/L, indicating that the straw not only acts as the carrier of bacteria, but also has a certain degree of adsorption effect on phenol, which reduces the toxicity of phenol on bacteria (see Supplementary Material, Figure S1). Herein, the degradation efficiency of phenol by immobilized bacteria is higher than that of free bacteria. The increase mainly relates to two points, one refers to the accelerated diffusion of substrate molecules and degradation products across the pores on straw and the improvement of degradation enzyme utilization efficiency, the other involves the improvement of the survival ability of immobilized bacteria (Zheng et al. 2017; Ke et al. 2018). In addition, the cationic straw carrier can adsorb the concentration of phenol to a level that bacteria can effectively degrade, maintaining smooth bioremediation. Moreover, the carrier can provide a shelter for the bacteria, so their degradation ability is less affected. Further, investigations have been reported that immobilized materials have the ability to treat soil pollution (Ke et al. 2018; Li et al. 2019). The existence of carriers enables microbes to survive and colonize better. Thus, the prepared materials may have the ability to be further applied in polluted soil.

The effect of pH on phenol degradation by bacteria

In a practical environment, the pH of polluted water, which is often not a neutral environment, has a great influence on the removal of phenol by bacteria. Therefore, it is meaningful to explore the effect of pH on the degradation of phenol by immobilized and free bacteria (Xu et al. 2010a, 2010b; Zhao et al. 2020). On the 120 h of degradation for 1,000 mg/L phenol, the degradation efficiency of immobilized bacteria is 63.3% at pH 7, while the free bacteria is 50.7% (Figure 5), which is weaker than that of immobilized bacteria. However, the degradation efficiency in acidic or alkaline conditions is obviously decreased, especially in the case of free bacteria. For example, in the environment of pH 5, the degradation efficiency of free bacteria is less than 20%, while immobilized bacteria is approximately 35%, much higher than free bacteria. Thus, both the free bacteria and immobilized bacteria possess excellent ability at the pH of 7 and 8, because bacteria prefer to live in neutral or weak alkaline conditions, and too much acid or alkali will affect their growth and reproduction, further affecting the degradation of phenol. In addition, as excellent physical and chemical features, straw carrier can be seen as a buffer to reduce the acidity and alkalinity to some extent in the wastewater, then provide a protective environment for bacteria to survive (Zheng et al. 2017; Zhao et al. 2020). Moreover, the neutral pH environment for bacteria in straw could reduce toxicity, promote the metabolism of bacteria, and maintain high enzyme activity, which directly affects the biodegradation of phenol and the dissolution and stability of degradation products.

CONCLUSIONS

In conclusion, a novel biomaterial combined of cationic straw and bacteria was developed, which displayed excellent ability for the degradation of phenol pollutants. Compared with free bacteria, the biomaterial containing bacteria can degrade a higher concentration phenol, shorten the degradation time and improve the degradation efficiency. Phenol solution at
1,200 mg/L can be completely removed by the immobilized bacteria within 216 h. The immobilized bacteria biomaterial has higher degradation efficiency for phenol than the free bacteria under acidic or alkaline conditions. The present design demonstrates the successful realization of an efficient phenol removal biomaterial. It can be envisioned that this biomaterial has great potential in the field of remediation of organic pollution.

**DATA AVAILABILITY STATEMENT**

All relevant data are included in the paper or its Supplementary Information.

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