Wheat Straws and Corn Straws as Adsorbents for the Removal of Cr(VI) and Cr(III) from Aqueous Solution: Kinetics, Isotherm, and Mechanism

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ABSTRACT: In this paper, the adsorption properties of wheat straw (WS) and corn straw (CS) for Cr(VI) and Cr(III) in solution were studied. The effects of adsorption time, pH of the solution, temperature, and initial concentration of metal ions on adsorption capacity were investigated. The adsorption mechanism was discussed. The results showed that the adsorption isotherms of WS and CS for Cr(VI) and Cr(III) satisfied the Langmuir equation. By fitting the Langmuir equation, the saturated adsorption capacity of WS for Cr(VI) and Cr(III) can reach 125.6 and 68.9 mg g⁻¹, and that of CS for Cr(VI) and Cr(III) can reach 87.4 and 62.3 mg g⁻¹, respectively. The adsorption kinetics conformed to the pseudo-second-order kinetic equation. The effect of temperature on the adsorption capacity was not significant. Physical diffusion and chemical adsorption coexist in the process of adsorption of metal ions by straws, and chemical adsorption is dominant, and the effect of physical diffusion on the chemical adsorption rate can be neglected. It can be seen from the experimental results that the treatment of chromium-containing wastewater by using cheap and easily available wheat straw and corn straw had a remarkable effect. The adsorbed straw could be completely desorbed and had excellent recyclability, indicating that the straws are ideal adsorbents.

1. INTRODUCTION

Wastewater from mining and smelting of chromium, manufacture and plating of chromium salts, metal processing, tanning, paints, pigments, printing, and dyeing contains a large amount of Cr(VI). Natural activities such as volcanic eruptions and weathering of rocks can also cause chromium emissions. Therefore, chromium exists in water and soil environments for a long time, leading to serious environmental pollution and affecting human health.¹⁻³ The methods of precipitation, membrane filtration, amine solvent extraction, ion exchange, adsorption, electrodeposition, and various other biological processes are usually used to treat chromium-containing wastewater. Among them, adsorption is the most common method. The biosorption method is widely used because of its low cost, high efficiency, stability, and good selectivity to different heavy metal ions.⁴⁻¹²

China is one of the countries with the most abundant straw resources in the world. The annual total output of straw resources in China is 600⁻700 million tons, and the utilization rate is 60⁻70%.¹³ The comprehensive utilization and recycling of straw is one of the most important topics in China. Rational utilization of straw will become a necessary trend for future environmental protection and sustainable development strategies.¹⁶⁻¹⁷

First, the surface of the straw is rough, the cell wall capillary structure is internal, the porosity is high, and the surface area is large. These structures are favorable for the physical adsorption of pollutants.¹⁶⁻¹⁹ Second, the straw body is generally composed of macromolecules such as cellulose, hemicellulose, lignin, and protein, which consist of C, H, O, N, and S. These macromolecules contain functional groups such as hydroxyl, carboxyl, and amino groups. These functional groups have strong coordination ability.²⁰⁻²¹ Therefore, the straw has a strong chemical adsorption capacity and, in particular, absorbs heavy metal ions having an electronic structure capable of forming a coordinate bond.²² Compared with other traditional methods for treating pollutants in water, it has the advantages of good structural performance, large specific surface areas, high removal efficiency, abundant sources, low costs, and less secondary pollution.²³ A large number of the literature have confirmed that the use of modified straw as an adsorbent for
the removal of water pollutants is a good way to utilize straw resources, which not only improves the utilization rate of straw but also opens up a new water-pollution treatment mode. Therefore, the development and utilization of straw-based adsorbents has important practical significance.

In this paper, wheat and corn straw were selected as research objects, and their adsorption characteristics for Cr(VI) and Cr(III) were studied. Surface properties such as surface functional groups and pore structures of biomass carbon were analyzed. They are expected to provide a scientific basis for the efficient treatment of heavy metal pollution by biochar and also open up new ways for the rational development and recycling of straw.

2. EXPERIMENTAL SECTION

2.1. Materials. Wheat straw and corn straw, K2Cr2O7 (AR, Tianjin Fuchen Chemical Reagent Factory), and Cr(NO3)3·9H2O (AR, Tianjin Fuchen Chemical Reagent Factory). All other chemicals were of analytical grade.

2.2. Preparation of Adsorbents. The straws were washed by distilled water, dried in air, and cut into 1 to 2 cm length. They were then ground in a ball mill, sieved to particle sizes of 100 mesh, and placed in a desiccator for later use.

2.3. Characterization of Adsorbents. The materials were analyzed using a Tensor 27 FT-IR spectrophotometer (Bruker Company, Germany), scanning electron microscope (SEM, Hitachi, Japan), X-ray diffractometer (XRD, D2-phas, Dandong Haoyuan Instrument Co., Ltd.,China), Rario type II elemental analyzer (Elementary, Germany), BET specific surface meter (3H-2000PS1, Beijing, China), and an instrument of microelectrophoresis (JS94H, Shanghai Zhong Chen Digital Technology Equipment Co., Ltd.).

2.4. Batch Adsorption Experiment. An amount of 0.2 g of adsorbent was placed in a solution containing one metal ion for adsorption, and the adsorption properties of straw on Cr(VI) and Cr(III) were studied under noncompetitive conditions. All the adsorption experiments were carried out upon stirring at 200 rpm in an orbital shaker for 12 h at 25 °C. The adsorbed mixture was separated by membrane filtration, washed with distilled water, and reused in the next cycle of adsorption experiments.

2.5. Recycling Experiments. The Cr(VI)-loaded straws and Cr(III)-loaded straws were regenerated with 1 M KOH solution and 0.1 M HNO3 solution, respectively, then collected by filtration, washed with distilled water, and reused in the next cycle of adsorption experiments.

3. RESULTS AND DISCUSSION

3.1. Characterization of Adsorbents. 3.1.1. Physical and Chemical Properties of Straw. Table 1 shows the chemical and elemental composition and physical structure parameters of wheat and corn straws.

| parameters | wheat straw | corn straw |
|------------|-------------|------------|
| chemical composition (%) |              |            |
| cellulose  | 45.32       | 42.40      |
| hemicellulose | 19.89     | 29.60      |
| lignin     | 12.49       | 21.70      |
| ash        | 5.03        | 5.10       |
| elemental composition (%) |              |            |
| O          | 49.96       | 27.26      |
| C          | 42.69       | 70.07      |
| N          | 6.32        | 2.09       |
| H          | 1.03        | 0.58       |
| surface area (m2 g−1) | 12.576     | 10.235     |
| BJH cumulative volume (cm3 g−1) | 0.03    | 0.01       |
| average pore diameter (nm) | 3.045     | 4.497      |
chemical composition, elemental composition, and pore structure parameters of straws. Straws are composed of macromolecules such as cellulose, hemicellulose, lignin, and protein, which are composed of elements such as C, H, O, and N. These macromolecules contain functional groups such as hydroxyl groups, carboxyl groups, and amino groups with strong coordination ability. Therefore, straws possess strong ability of chemical adsorption. It can be seen from the pore structure parameters of straw that the straw has high porosity and a large surface area, which is beneficial to the physical adsorption of pollutants.

3.1.2. Analysis of SEM. The photo and SEM of the straws are shown in Figure 1. The straws have a rough surface, and there are cell wall capillary structures inside the straws. The structure of the straws is loose, which can expose more active sites and is more conducive to the chemical adsorption. At the same time, it can be seen that there are more small pores distributed in the loose pores of the straw, which is beneficial to increase the specific surface area.

3.1.3. Analysis of FT-IR. The IR spectrum of straw is shown in Figure 2. It can be seen from Figure 2 that there are a large number of surface functional groups in the straw, and these surface functional groups may play a very important role in the process of adsorbing heavy metal ions. Wheat straw has a strong absorption peak in 3412, 1702, 1615, 1273, 1097, and 950 cm⁻¹ (Figure 2a). The broad peak near 3412 cm⁻¹ is a stretching vibration of a hydroxyl group (−OH). The peak at 1702 cm⁻¹ is the characteristic absorption peak of an ester (C=O). The peak at 1615 cm⁻¹ represents the stretching vibration of C=C in the aromatic ring. The bending vibration absorption peak of the phenolic hydroxyl group is at 1273 cm⁻¹. The peaks near 1097 and 929 cm⁻¹ are the stretching vibrations of C=O and C−H, respectively. The infrared spectrum of corn straw is shown in Figure 2b. Among them, 3420 cm⁻¹ is the stretching vibration absorption peak of the phenolic hydroxyl group and amino group in proteins. The peak at 1720 cm⁻¹ is the stretching vibration peak of the carboxyl group. The bending vibration peak of the hydroxyl group is at 1383 cm⁻¹. The peak at 1250 cm⁻¹ represents the plane vibration absorption peak of hydroxyl in cellulose.

3.1.4. Analysis of ζ Potential. The ζ potential means that the charged particles attract the oppositely charged particles in the dispersion, and the ions near the surface of the particle are strongly bound, and the ions farther away form a difference between the internal and external potentials of a loose electron cloud. A large number of polar groups exist on the surface of the straw, and when it is dispersed in solution, a certain amount of ions can be adsorbed on the surface of the straw to form an electric double layer, thereby generating a ζ potential.

The pH−ζ potential curves of WS and CS samples are presented in Figure 3. It can be seen that the ζ potential values of wheat straw and corn decrease as pH increases. This can be explained by the mechanism of variable charge generation. Due to the presence of pH-dependent functional groups (such as hydroxyl and carboxyl groups) in WS and CS, the surface...
positive charge of CS and WS decreases, while the surface negative charge increases with the increase of pH. The hydroxyl and carboxyl groups of the surface are protonated under acidic conditions, resulting in the enhancement of positive charge in WS and CS. As the pH increases, these groups will exhibit a greater negative charge and result in the decrease of the positive charge in WS and CS.

3.2. Adsorption Property. 3.2.1. Effect of pH on Adsorption. Figure 4 shows the effect of solution pH on Cr(VI) and Cr(III) adsorption. The pH value of the solution affects the form of Cr(VI) and has a certain effect on the activity of the functional groups on the adsorbent. With the increase of pH, the adsorption amount of wheat straw and corn straw for Cr(VI) increased slowly and then decreased rapidly. When the pH is low, Cr(VI) mainly exists in the form of HCrO$_4^-$ and Cr$_2$O$_7^{2-}$, and these ions are easily adsorbed to the protonated adsorption site by electrostatic attraction, and the surface of the adsorbent is surrounded by a large amount of hydrogen ions, thereby increasing the attraction between the Cr(VI) and adsorbent surface bonding sites. As the pH increases, the concentration of OH$^-$ increases and induces competitive adsorption with Cr$_2$O$_7^{2-}$, resulting in a decrease in adsorption capacity. With the increase of the pH value, the adsorption capacity of wheat straw and corn straw for Cr(III) increased gradually. This may be because, at lower pH conditions, protonation in the solution gives the material a positive charge, while Cr(III) also has a positive charge, which affects the adsorption capacity of the adsorbent for Cr(III). As the pH value of the solution increases, OH$^-$ on the surface of the adsorbent increases, and the adsorbent can adsorb Cr(III) through a surface metal reaction, proton reaction, and complexation reaction. At higher pH, Cr undergoes a hydrolysis reaction. The higher the pH is, the stronger the hydrolysis is, which weakens the adsorption capacity of the adsorbent for Cr(III) in the solution. Therefore, pH 3.0 and 5.0 were selected as the optimum pH for subsequent adsorption of Cr(VI) and Cr(III), respectively.

3.2.2. Effect of Temperature on Adsorption. Figure 5 shows the effect of temperature on the adsorption of Cr(VI) and Cr(III). It can be seen from Figure 5 that the adsorption capacity of WS and CS for Cr(VI) and Cr(III) increased with the increase of temperature. The adsorption capacity improved with increasing temperature, probably due to the chemical interaction between the adsorbent and adsorbate, which created a new adsorption site under high-temperature conditions or accelerated the internal diffusion rate of metal ions into the pores of the adsorbent. However, as can be seen from Figure 5, the adsorption capacity did not change significantly with temperature.

3.2.3. Adsorption Isotherms. Figure 6 showed the adsorption isotherm for adsorption of Cr(VI) and Cr(III) by wheat straw and corn straw at 25 °C. With the increase of the equilibrium concentration in the system, the equilibrium adsorption capacity of wheat straw and corn straw for Cr(VI) and Cr(III) increased gradually, and it stabilized when the equilibrium mass concentration was 100 mg L$^{-1}$. The adsorption isotherm parameters fitted by the Langmuir and Freundlich models are shown in Table 2. The adsorption of wheat straw and corn straw for Cr(VI) and Cr(III) can better conform to the Langmuir model. Therefore, the
adsorption of WS and CS for Cr(VI) and Cr(III) belongs to monolayer adsorption.

3.2.4. Adsorption Kinetics. The dynamic adsorption effect of the adsorbent was investigated, as shown in Figure 7. It can be seen from Figure 7 that the adsorption process of the straw for Cr(VI) and Cr(III) was divided into two stages. The adsorption rate was very fast at 0−5 h, while the adsorption rate was slowed down after 5 h, and the adsorption capacity reached equilibrium after about 12 h.

The pseudo-first-order kinetic equation, pseudo-second-order kinetic equation, and intraparticle diffusion equation were used to fit the adsorption kinetics of the process, and the kinetic mechanism was analyzed. The fitting parameters are shown in Table 3.

It can be seen from Table 3 that the fitting effect of the pseudo-second-order kinetic equation was better than that of the pseudo-first-order kinetic equation and intraparticle diffusion equation, and the correlation coefficient $R^2$ reaches 0.99. When $R^2 > 0.980$, the model is considered to be suitable for describing the dynamics of the adsorption process. In addition, the equilibrium adsorption capacity obtained by fitting the pseudo-second-order kinetic equation was closer to the equilibrium adsorption capacity obtained in the experiment. Therefore, the pseudo-second-order kinetic model can better reflect the adsorption process of straws for Cr(VI) and Cr(III).

According to the mechanism established by the pseudo-second-order kinetic equation, it can be inferred that physical diffusion and chemical adsorption coexist in the process of adsorption of metal ions by straws, and chemical adsorption is dominant, and the effect of physical diffusion on the chemical adsorption rate can be neglected.

3.2.5. Adsorption Thermodynamics. Thermodynamic parameters such as free energy change ($\Delta G$), enthalpy change ($\Delta H$), and entropy change ($\Delta S$) can provide in-depth information about the inherent energy change related to adsorption, which can be calculated using eqs 7−9, respectively

$$
\Delta G = -RT \ln K_C \\
\Delta G = \Delta H - T \Delta S \\
\ln K_C = -\Delta H/RT + \Delta S/R
$$

where $R$ is the universal gas constant (8.314 J mol$^{-1}$ K$^{-1}$), $T$ is the absolute temperature (K), and $K_C$ is the adsorption equilibrium constant.

The results are shown in Table 4. The negative value of $\Delta G$ indicated that the adsorption of Cr(VI) and Cr(III) on WS and CS was a spontaneous process. The thermodynamic data of $\Delta H$ and $\Delta S$ proved the endothermic nature of the process and the increase of randomness at the solid/liquid interface, indicating that the temperature is a favorable factor for the adsorption kinetics. In the temperature range studied, $T\Delta S$ accounted for a large proportion of the total free energy, indicating that the increase of entropy during ion binding was an important factor to promote the interaction between metal ions and straws.

3.2.6. Reusability. To explore the desorption and reusability of straws, 1 M KOH and 0.1 M HNO$_3$ were used as eluants to carry out the recycle experiment for Cr(VI)-loaded straws and Cr(III)-loaded straws, respectively. As shown in Figure 8, there was no significant change in the adsorption capacities of WS and CS.
and CS during the repeated adsorption–desorption, indicating that the straws had excellent reusability.

4. CONCLUSIONS

In this work, the adsorption properties of wheat straw and corn straw for Cr(VI) and Cr(III) in solution were investigated. Pseudo-second-order and Langmuir adsorption isotherm models can preferably describe the adsorption process. The saturated adsorption capacity of WS for Cr(VI) and Cr(III) can reach 125.6 and 68.9 mg g\(^{-1}\), and that of CS for Cr(VI) and Cr(III) can reach 87.4 and 62.3 mg g\(^{-1}\), respectively. The straws possess excellent reusability. These results indicate that wheat straw and corn straw might serve as an efficient, simple, and low-cost material for heavy metal-ion removal from wastewaters. This research has theoretical and practical significance for the development and application of adsorbents based on agricultural straws. It also has a positive effect on solving the problem of heavy metal pollution in water and the problem of environmental pollution and safety caused by improper disposal of agricultural wastes.

Table 3. Related Parameters of the Kinetic Model

| equations                          | parameters | WS (Cr(VI)) | WS (Cr(III)) | CS (Cr(VI)) | CS (Cr(III)) |
|------------------------------------|------------|-------------|--------------|-------------|--------------|
| experiment                         |            |             |              |             |              |
| pseudo-first-order kinetic equation|            | 80.5        | 46.9         | 69.8        | 42.6         |
|                                    |            | 45.7        | 22.6         | 32.8        | 20.1         |
|                                    |            | 0.072       | 0.036        | 0.069       | 0.028        |
|                                    |            | 0.865       | 0.902        | 0.885       | 0.915        |
| pseudo-second-order kinetics       |            |             |              |             |              |
|                                    |            | 81.2        | 47.5         | 71.8        | 43.6         |
|                                    |            | 0.0059      | 0.0032       | 0.0056      | 0.0021       |
|                                    |            | 0.998       | 0.995        | 0.997       | 0.996        |
| intraparticle diffusion equation   |            | 0.215       | 0.197        | 0.193       | 0.108        |
|                                    |            | 0.957       | 0.948        | 0.962       | 0.971        |

Table 4. Thermodynamic Parameters Estimated for Adsorption of Cr(VI) and Cr(III) on WS and CS

| straw | metal ions | \(\Delta H\) (kJ mol\(^{-1}\)) | \(\Delta S\) (J mol\(^{-1}\) K\(^{-1}\)) | 25 °C | 35 °C | 45 °C | 55 °C |
|-------|------------|--------------------------------|-------------------------------------------|------|------|------|------|
| WS    | Cr(VI)     | 22.87                          | 118                                       | -12.29 | -13.49 | -14.64 | -15.86 |
|       | Cr(III)    | 16.96                          | 92                                        | -10.46 | -11.36 | -12.29 | -13.20 |
| CS    | Cr(VI)     | 19.02                          | 104                                       | -11.97 | -12.95 | -14.05 | -15.10 |
|       | Cr(III)    | 14.38                          | 82                                        | -10.06 | -10.86 | -11.69 | -12.53 |

Figure 8. Adsorption capacity of WS and CS after 10 adsorption–desorption cycles.

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Notes
The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**

This work was supported by the “Cyanine Talent” of Xianyang Normal College (no. XSXY201710), scientific research project of Xianyang Normal College (no. XSXY19047), and Science and Technology Plan Project of Xianyang (no. 2019k02-29).

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