Characteristics and adsorption study of the activated carbon derived from municipal sewage sludge

Tiecheng Guo, Sicong Yao, Hengli Chen, Xin Yu, Meicheng Wang and Yao Chen

ABSTRACT

Sewage sludge-based activated carbon is proved to be an efficient and low-cost adsorbent in treatment of various industrial wastewaters. The produced carbon had a well-developed pore structure and relatively low Brunauer–Emmett–Teller (BET) surface area. Adsorptive capacity of typical pollutants, i.e. copper Cu(II) and methylene blue (MB) on the carbon was studied. Adsorptions were affected by the initial solution pH, contact time and adsorbent dose. Results showed that adsorption of Cu(II) and MB on the produced carbon could reach equilibrium after 240 min. The average removal rate for Cu(II) on the carbon was high, up to 97% in weak acidic conditions (pH = 4–6) and around 98% for MB in a very wide pH range (pH = 2–12). The adsorption kinetics were well fitted by the pseudo-second order model, and both Langmuir and Freundlich isotherm models could well describe the adsorption process at room temperature. The theoretical maximum adsorption capacities of Cu(II) and MB on sewage sludge-based activated carbon were 114.94 mg/g and 125 mg/g, respectively. Compared with commercial carbon, the sewage sludge-based carbon was more suitable for heavy metal ions’ removal than dyes’.

Key words | activated carbon, adsorption, Cu(II), methylene blue, sewage sludge

INTRODUCTION

Sewage sludge, the major byproduct of wastewater treatment plants, has been produced in an ever increasing amount in recent years. Treatment and disposal of this solid waste accounted for 25–65% of the total operation cost to a wastewater treatment plant. Municipal sewage sludge handling is one of the most significant challenges in wastewater treatment processes. The traditional handling and utilization methods for sewage sludge include agriculture application, incineration, landfill, and sea dumping, however each of them has their own limitations (Huang et al. 2014).

With its carbonaceous nature, abundant in volatile matter and low-cost resource, sewage sludge has been identified as a potentially attractive material for the production of activated carbon. Many efforts have been made to produce sewage sludge-based activated carbons. Wang used activated carbons from sewage sludge by microwave-induced phosphoric acid and zinc chloride activation to adsorb copper ions. The maximum Cu(II) adsorption capacity was 10.56 mg/g (Wang et al. 2011b). Li used activated carbon made from paper mill sewage sludge by steam activation to adsorb methylene blue (MB), and the maximum MB adsorption capacity was 130.69 mg/g (Li et al. 2011). Fan used activated carbons produced from surplus sewage sludge by heat treatment for alkaline-black adsorbing. And it was found that activated carbon from sewage sludge exhibited higher adsorption capacities for alkaline-black than the commercial one (Fan & Zhang 2008). It is economical to produce activated carbon from sewage sludge instead of traditional precursors. Taking advantage of the carbonaceous nature, preparing porous carbons from sewage sludge is believed to be an environmentally benign and sustainable technique. Moreover, activated carbon derived from sewage sludge has been proved efficient enough for various pollutants’ adsorption.

Copper ion Cu(II) and MB were typical industrial pollutants presented in wastewater. Cu(II) is one of the highest mammalian toxic and most serious pollutants in industrial wastewater. MB is the most commonly used material in azo dyeing industry, and has various detrimental effects on...
human and aquatic life when released into the environment (Fu et al. 2010). Cu(II) and MB were widely used in adsorption studies to research adsorptive performance and mechanism due to their readily adsorptive capacities onto carbons (Demiral & Gungor 2016; Alt Nt et al. 2017). It's favorable for analysis and understanding the adsorption processes based on their easily adsorbed characteristics and high adsorbed capacities. Thus, the purpose of our study was to evaluate the behavior and efficiency of the sewage sludge-based activated carbons for Cu(II) and MB removal from aqueous solution. Brunauer–Emmett–Teller (BET) and scanning electron microscopy (SEM) were used to evaluate the pore structure and the surface morphology of the adsorbents, respectively. The influence of several operating parameters, such as pH, contact time and adsorbent dosage during adsorption was also investigated. The equilibrium and kinetics of their adsorption process were then analyzed to understand adsorptive mechanism on the produced carbons. This work would be useful for better understanding the beneficial utilization of sewage sludge compared to other waste disposal.

**METHODS**

**Materials and characterizations**

Dewatered surplus sewage sludge from a local municipal wastewater treatment plant was used for the preparation of activated carbon. The proximate and ultimate analysis of the raw sludge was listed in Table 1. The ultimate analysis of the raw sludge is obtained by a Vario MICRO select Elemental Analyzer, Elementar, Germany, in accordance with the standard of JY/T 017-1996 elemental analyzer method. The proximate analysis of the sample was conducted as described in the standard method (GB/T212-200, China).

The sludge was dried at 105 °C for 48 h and crushed into powder. The activating agent consisted of ZnCl₂ (6 mol/L) and H₂SO₄ (wt% 35%) in a volume ratio of 4:1. The precursor was then impregnated into the activating agent at a ratio of 2.3 (g·mL) for 24 h and placed into a ceramic crucible. The crucible was inserted in a tubular furnace (KXG-2-13A, Shanghai, China) with a constant heating rate of 10 °C/min and the final carbonization temperature was 550 °C. After 1 hour pyrolysis under purified N₂ (99.99%) flow of 200 mL/min, the sample was cooled down to room temperature and washed several times sequentially with hot distilled water (70–80 °C deionized water) to remove any residual chemicals. Activated carbon products were then dehydrated under 105 °C in an electrical heating oven, ground and sieved to a mean particle size of 75 μm for further tests.

The stock solution of Cu(II) and MB was prepared by dissolving a certain amount of Cu(NO₃)₂·3H₂O or MB (chemical formula = C₁₆H₁₈ClN₃S, MW = 320 g/mol, λ_max = 665 nm) in deionized water to get a stock concentration of 1,000 mg/L. All the chemicals and reagents used were analytical grade.

The surface area and pore structure of prepared activated carbons was measured by BET adsorption technique using a ZXF-06 sorptometer (Tianjin, China). The adsorbent's surface was characterized by SEM (JSM-5900LV, Tokyo, Japan).

**Batch adsorption tests**

Impact of operational parameters, i.e. reaction time, pH, carbon dosage and pollutants' concentration, on the produced carbon's adsorptive capacity were carried out in our work. Experiments were conducted in a series of 150 mL Erlenmeyer flasks containing 50 mL of various concentrations of Cu²⁺ or MB solution.

The flasks were agitated in a thermostatic oscillator at 150 rpm under room temperature for 4 h. The samples were then collected, filtered and analyzed for pollutants' concentration. The Cu(II) concentration in the solution was analyzed by an atomic absorption spectrometer (AAS) (PEAA400, USA). The MB concentration in the solution was determined by measuring the absorbance at λ_max = 665 nm using a double beam UV-vis spectrophotometer (UV-1100, Shanghai, China).

<table>
<thead>
<tr>
<th>Proximate analysis</th>
<th>Ultimate analysis</th>
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<tbody>
<tr>
<td>Moisture content</td>
<td>C</td>
</tr>
<tr>
<td>Volatile content</td>
<td>83.21</td>
</tr>
<tr>
<td>Ash content</td>
<td>32.61</td>
</tr>
<tr>
<td>Fixed carbon</td>
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| *Calculated by difference.
Each experiment was twice duplicated under identical conditions. And all the data in the figures are the average value of the two experiments.

The amount of adsorption at equilibrium, \( q_e \) (mg/g), was calculated as Equation (1):

\[
q_e = \frac{(C_0 - C_e)V}{W}
\]

where \( C_0 \) and \( C_e \) (mg/L) are the liquid-phase concentrations of Cu(II) or MB at initial and equilibrium, respectively. \( V \) (L) is the volume of the solution, and \( W \) (g) is the mass of dry adsorbent used.

RESULTS AND DISCUSSION

Characterization of adsorbents

Figure 1 shows the SEM micrograph of the produced activated carbon. As shown in Figure 1, the adsorbent exhibits an uneven and rough surface morphology. Pores, cavities and crevices are presented in the adsorbent’s surface, which result from the evaporation of chemical reagent ZnCl\(_2\) during carbonization and then leaving the space previously occupied by the reagents. Thus, it can enhance trapping and adsorbing dyes or heavy metals from aqueous solution to the solid layer (Chen et al., 2014).

Textural characteristics of the sewage sludge carbon (SSC) and a coconut shell-based commercial available carbon (CC) are shown in Table 2. It can be observed that SSC had a much lower specific surface area (218.37 m\(^2\)/g) compared to CC (1,412.07 m\(^2\)/g). Most commercially available activated carbons have a specific surface area of 400–1,500 m\(^2\)/g. As listed in Table 2, SSC has a relatively lower total pore volume than CC’s, while a higher micropore volume was presented in SSC than CC’s.

Batch adsorption tests

Cu(II) adsorption

To investigate the effect of solution pH on Cu(II) adsorption, the adsorption experiment ran for 4 hours with 3 g/L sewage sludge carbon to treat an initial Cu(II) concentration of 200 mg/L (Xie et al., 2013b). The solution pH was adjusted between 2 and 7 by adding 0.1 mol/L NaOH or HNO\(_3\) solutions.

As shown in Figure 2(a), Cu(II) adsorption capacity on the produced carbon kept stable and high efficient removal at pH 4–6. The Cu(II) removal rate was high, up to 97%, within this pH range. When pH was higher than 6.0, the precipitation of insoluble metal hydroxides took place and restricted the adsorption reaction. In our test, the initial pH of the tested Cu(II) solution was about 5.6 and the final pH was close to 6.0. Under such an operation, the average Cu(II) removal rate could be high, up to 98%, by the produced carbon adsorption. Wang found the optimum heavy metal adsorption by activated carbon from sewage sludge was pH = 5 (Wang et al., 2011a). Other researchers also got similar results using activated carbons prepared from non-coal materials. Demiral found the optimum operation for Cu(II) adsorption on activated carbon prepared from grape bagasse was around pH = 5 (Demiral & Gungor, 2016).

It is believed that the surface of the activated carbon is negatively charged as a consequence of the presence of acidic functional groups, and this is also found for sewage sludge-derived adsorbents (Tang et al., 2016). H\(^+\) competes with the Cu ions for the active surface sites in low pH conditions. In accordance with other studies and MINTEQ calculation, with H\(^+\) decreasing and pH increasing, Cu\(^{2+}\) and Cu(OH)\(^+\) gradually become the dominant species at acidic conditions. Other species like Cu(OH)\(_2\) and Cu(OH)\(_3\) are not considered below pH 6.0 (Bouhamed et al., 2012; Xie et al., 2013a). The negatively charged carbon surface plays a high electrostatic attraction between adsorbents and heavy metals’ positive charges.

Sewage sludge-based activated carbons ranging from 1 to 4 g/L were added in a series of 50 mL Cu(II) solutions (200 mg/L) and reacted for 4 h to study the effect of
adsorbent dosage and contact time on Cu(II) adsorption. The initial pH was ca. 5.6 without any adjustment.

The results are shown in Figure 3(a). As shown in Figure 3(a), the Cu(II) removal rate rapidly increased with the increment of adsorbent dosage ranging from 1 to 3 g/L, then slightly increased with the carbon dosage of 3–4 g/L. The final removal rate could be higher than 98%. With an increasing adsorbent dosage, more active sites were available and thus facilitated the extent of adsorption (Gupta & Gogate 2014). Insignificant change of removal efficiency at higher loading of the adsorbent might indicate overlapping of adsorption sites and possible aggregation of the adsorbent particles, which led to a decrease in the total surface area.

Also, as illustrated in Figure 3(a), the uptake of metal ion occurred in two stages. A rapid uptake of solid-liquid contact took place within the first 30 min and was followed by a subsequent slow uptake from 30 to 240 min. The Cu(II) removal rate was initially high due to the full availability of exposed sites on the adsorbent. Then, it became slower because of the repulsive forces between cations already set and the free ones in solution, which inhibited their access and increased the mass transfer resistance (Ben-Ali et al. 2017). The removal rate was slightly changed

<table>
<thead>
<tr>
<th>Characteristics of the produced carbon</th>
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<tbody>
<tr>
<td>Adsorbents</td>
</tr>
<tr>
<td>------------</td>
</tr>
<tr>
<td>SSC</td>
</tr>
<tr>
<td>CC</td>
</tr>
</tbody>
</table>

Figure 2 | Impact of pH on the (a) Cu(II) and (b) MB adsorption.

Figure 3 | Impact of the activated carbon dosage on the (a) Cu(II) and (b) MB adsorption.
and Cu(II) adsorption kept stable and reached equilibrium at 240 min.

**MB adsorption**

To investigate the effect of solution pH on MB adsorption, the initial pH of the MB solution was adjusted from 1 to 12. Then 2 g/L carbon was added into a series of 50 mL MB solutions (200 mg/L) with different initial pH at room temperature and reacted for 4 h according to our previous research (Chen et al. 2012). According to Figure 2(b), there was little difference in MB adsorption within pH = 2–12. The MB removal rate was stable and high, around 97.6 ± 1.3%, in such a wide pH range.

MB is a cationic dye with the estimated dimensions of 1.43 nm × 0.61 nm × 0.4 nm, and its adsorption by activated carbon is very susceptible to solution pH (Gokce & Aktas 2014). The same as Cu(II) adsorption, the H⁺ ions in the solution would effectively compete with MB at low pH, while an increased MB adsorptive rate would obtain with the increasing pH due to the electrostatic attraction between the positively charged adsorbate and negatively charged adsorbent. However, MB adsorption was not affected obviously in acidic conditions in our tests, which indicated that there was enough adsorbing surface for H⁺ and MB, and the influence of pH was suppressed and negligible.

Sewage sludge-based activated carbons ranging from 1 to 2.4 g/L were added in a series of 50 mL MB solutions (200 mg/L) and reacted for 4 h to study the effect of adsorbent dosage and contact time on MB adsorption. The initial pH was ca. 5.0 without any adjustment. The experiment results are presented in Figure 3(b).

It was observed that the change of MB removal rate with increasing adsorbent dosage was same as Cu(II)’s. It was rapidly increased with the increment of adsorbent dosage ranging from 1 to 2 g/L, and MB could be totally adsorbed by the produced carbon and had 100% removal with 2.4 g/L adsorbent dosage. It was also proved that the produced carbon had a much higher adsorptive capacity for dyes than heavy metals.

From Figure 3(b), the MB removal rate was insignificantly changed after 240 min. It takes a relatively long time to reach equilibrium. In the adsorption process, the dye ions were quickly adsorbed by the exterior surface of the activated carbon. The adsorption of the exterior surface reached saturation, the dye ions were exerted onto the pores of the adsorbent particles and slowly adsorbed by the interior surface of the particle (Chen et al. 2012).

**Adsorption kinetics**

To investigate the adsorption processes of Cu(II) and MB onto sewage sludge carbon, pseudo-first order, pseudo-second order, and intra-particle diffusion kinetic models were used to describe the kinetic process.

The pseudo-first order kinetics model was based on the following linear classic Equation (2) (Mahmoud et al. 2012):

\[
\ln(q_e - q_t) = \ln q_e - k_1 t
\]

where \( k_1 \) (1/min) is the rate constant of the pseudo-first order adsorption, \( q_t \) (mg/g) denotes the adsorption amount at \( t \) (min) and \( q_e \) (mg/g) is the adsorption amount at equilibrium. The adsorption rate constant, \( k_1 \), can be calculated by plotting \( \ln(1 - q_t/q_e) \) versus \( t \).

The pseudo-second order model, which is based on the assumption that the rate-limiting step involved chemisorption, can be written in a linear form as Equation (3) (Ozcan et al. 2005):

\[
\frac{t}{q_t} = \frac{1}{k_2q_e^2} + \frac{t}{q_e}
\]

where \( k_2 \) (g/mg min) is the rate constant of pseudo-second order adsorption, and \( k_2 \), \( q_e \) can be obtained from the intercept and slope by plotting \( t/q_t \) versus \( t \).

Intra-particle diffusion model, based on the theory proposed by Weber and Morris, was used to identify the diffusion mechanism as Equation (4) (Hameed 2009):

\[
q_t = k_id\sqrt{t} + C
\]

where \( k_{id} \) is the intra-particle diffusion rate constant (mg/min⁰.⁵ g) and \( C \) is a constant.

Kinetic data were calculated and are given in Table 3. As listed in Table 3, with the dosage changed, the parameters of the three kinetic models changed irregularly. The correlation coefficients (R²) of the pseudo-first order and intra-particle diffusion model were not high enough. The pseudo-second order model demonstrated good linearity agreement at each dosage. The high correlation coefficients (0.99 < R² < 1) indicated that it was suitable to describe the kinetic adsorption process of Cu(II) and MB onto the carbon (Ezzeddine et al. 2016; Liu et al. 2016). Consequently, it can be concluded that the adsorption process of Cu(II) and MB onto the sewage sludge-based carbon was partially controlled by chemisorption (Zhang et al. 2013).
Adsorption isotherm

The equilibrium data were important for the evaluation of the adsorption mechanism, which was critical to understand and improve the adsorptive efficiency of the carbon. Therefore, evaluation of the carbon adsorption capacity with different models is helpful for the process design and optimization of the operating protocol. Both Langmuir and Freundlich isotherms were adopted to describe Cu(II) and MB adsorption on the carbon.

The Cu(II) and MB adsorption isotherms under initial pH value at 25°C are shown in Figure 4, using 3 g/L sewage sludge-based activated carbon mixed with Cu(II) solution at concentrations from 60 to 500 mg/L, and 2 g/L carbon mixed with MB solution at concentrations from 50 to 400 mg/L, respectively.

Langmuir theory is based on the assumptions that adsorption takes place at specific homogenous sites of the carbon and no further adsorption occurs once the substrate molecule occupies a site. The Langmuir isotherm linear form is represented by Equation (5) (Magdy & Daifullah 1998):

\[
\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{q_m K_L}
\]  

(5)

where \( q_e \) (mg/g) is the amount of substrate adsorbed, \( q_m \) (mg/g) is the maximum amount of substrate, \( C_e \) (mg/L) is the equilibrium concentration of substrate in solution, and \( K_L \) (L/mg) is the Langmuir isotherm constant that relates to the energy of adsorption.

The Freundlich isotherm was also applied for the adsorption studies, specifically to model the multilayer adsorption and adsorption on heterogeneous surfaces. The Freundlich equation in linear form is given by Equation (6) (Aksu 2002):

\[
\ln q_e = \ln k_f + \frac{1}{n} \ln C_e
\]

(6)

where \( K_f \) (mg/g) is the Freundlich equilibrium constant and \( n \) (g/L) is the adsorption intensity determined from the intercept and slope respectively.

Table 3 | Kinetic parameters for the adsorption of MB and Cu(II) onto the produced carbon

<table>
<thead>
<tr>
<th>Substrate</th>
<th>Adsorbent dosage (g/L)</th>
<th>Pseudo-first order</th>
<th>Pseudo-second order</th>
<th>Intra-particle diffusion</th>
<th>Experimental adsorption capacities ( Q_{exp} ) (mg/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>( K_1 ) (1/min)</td>
<td>( R^2 )</td>
<td>( K_2 ) (g/min mg)</td>
<td>( R^2 )</td>
<td>( K_{id} ) (mg/min^1/2 g)</td>
</tr>
<tr>
<td>MB</td>
<td>1.0</td>
<td>0.0086</td>
<td>0.9812</td>
<td>0.0087</td>
<td>0.9901</td>
</tr>
<tr>
<td></td>
<td>1.2</td>
<td>0.0123</td>
<td>0.9093</td>
<td>0.0074</td>
<td>0.9928</td>
</tr>
<tr>
<td></td>
<td>1.6</td>
<td>0.0169</td>
<td>0.9747</td>
<td>0.0094</td>
<td>0.9969</td>
</tr>
<tr>
<td></td>
<td>2.0</td>
<td>0.0223</td>
<td>0.9708</td>
<td>0.0123</td>
<td>0.9996</td>
</tr>
<tr>
<td></td>
<td>2.4</td>
<td>0.0203</td>
<td>0.8577</td>
<td>0.0147</td>
<td>0.9997</td>
</tr>
<tr>
<td>Cu(II)</td>
<td>1.0</td>
<td>0.0061</td>
<td>0.9139</td>
<td>0.0098</td>
<td>0.9919</td>
</tr>
<tr>
<td></td>
<td>2.0</td>
<td>0.0256</td>
<td>0.9527</td>
<td>0.0125</td>
<td>0.9986</td>
</tr>
<tr>
<td></td>
<td>3.0</td>
<td>0.0175</td>
<td>0.9983</td>
<td>0.0155</td>
<td>0.9995</td>
</tr>
<tr>
<td></td>
<td>4.0</td>
<td>0.0353</td>
<td>0.9642</td>
<td>0.0195</td>
<td>1.0000</td>
</tr>
</tbody>
</table>

Figure 4 | The relationship of Ce and qe at equilibrium adsorption of Cu(II) and MB onto the produced carbon.
Table 4 shows that the experimental adsorption data were well fitted with the Langmuir and Freundlich isotherm models. As shown in Table 4, both the Langmuir and Freundlich models could describe the Cu(II) adsorption process with high correlation coefficients ($R^2 > 0.98$). And the two models’ correlation on MB adsorption was a little lower ($R^2 > 0.94$) and not much different between the two models. The statistical physics approach of anchorage number could be adopted to explain this phenomenon (Khalfaoui et al. 2006; Fabbricino & Pontoni 2016), in which dye molecules might be partially anchored to two receptor sites and partially anchored to three receptor sites. As listed in Table 4, the equilibrium experimental adsorption capacities of Cu(II) and MB are very close to the theoretical adsorption capacities. The maximum adsorption capacity obtained from the Langmuir isotherm was 114.94 and 125 mg/g for Cu(II) and MB, respectively.

Evaluations of activated carbon

Compared to other non-coal activated carbons derived by thermal method, sewage sludge-based activated carbon displayed an excellent performance. A comparative test was carried out to evaluate the performance between sewage sludge-based carbon (SSC) and the commercial carbon (CC) which was derived from coconut shells on removal of MB and Cu(II) from aqueous solution. The experiment operated for 240 min to achieve equilibrium under 25°C. The initial concentrations of Cu(II) and MB were both 200 mg/L, and the initial pH = 5, with adsorbent dosage of 3 g/L for Cu(II) and 1 g/L for MB solution. The results are illustrated in Figure 5.

From Figure 5, it can be seen that SSC was much more effective in Cu(II) adsorption than CC. The removal rate on SSC (96%) was three times larger than CC’s (30.4%). However, the MB removal by SSC (75%) was relatively lower than CC’s (99%). The adsorption difference between heavy metal ions and dyes onto SSC and CC may be mainly due to their pore structures. The microporous volume of SSC was 0.1013 cm$^3$/g, which was a little higher than CC’s (0.0933 cm$^3$/g), while the mesopore volume of SSC was 0.1506 cm$^3$/g, which was much smaller than CC’s (0.6049 cm$^3$/g). Commonly, the hydrated radius of heavy metal ions was much smaller, and the dye molecule of MB had a much larger particle size. Thus, Cu(II) was able to easily enter into the carbon and be trapped by its microporous structure, while the large dye molecule was limited in mass transfer and hardly got into the carbon. So SSC was more suitable for heavy metal ion adsorption than dyes. Moreover, with proper modification and improvement on mesopores, SSC can also be highly efficient in dyes’ treatment.

CONCLUSIONS

In this study, activated carbon was produced from sewage sludge by ZnCl$_2$ activation at 550°C. It was found that the specific surface area and total pore volume of the produced carbon were 218.37 m$^2$/g and 0.2519 cm$^3$/g, respectively. Equilibrium was reached after 240 min for both pollutants’ adsorption. With an initial concentration of 200 mg/L, and carbon dosage of 3 g/L for Cu(II) and 2 g/L for MB, the
average removal rate for Cu(II) on the carbon was high, up to 97%, in weak acidic conditions (pH = 4–6) and ca. 98% for MB over a very wide pH range (pH = 2–12). The pseudo-second order kinetic model well described the adsorption process for both Cu(II) and MB, which indicated that the rate-limiting step might be chemical sorption rather than diffusion. Isotherm data were fitted well to both Langmuir and Freundlich models for both kinds of pollutants. The theoretical maximum adsorption quantities of Cu(II) were compared with commercial activated carbon and the prepared activated carbon from sewage sludge was more suitable for heavy metal ions’ adsorption than dyes.

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