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Padina pavonica for the Removal of Dye from Polluted Water

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ABSTRACT

The adsorption of fast yellow dye onto dried biomass *Padina pavonica* was studied in batch experiments. The amount of dye adsorbed (mg/g) was increased with the increase in initial dye concentration. An equilibrium time of about 90 min was achieved for dye concentrations ranging from 5 to 160 mg/L with maximum removal percentage of 73.2%. Pseudo-first and second order kinetic models have been used to analyze the adsorption data. The pseudo second-order kinetic model adequately described the adsorption data with correlation coefficient between 0.96 and 1.084. Fourier transform infra-red analysis demonstrated the chelating character of the dye molecule to different functionalities groups of the alga. Stirring speed higher than 50 rpm revealed no significant changes in dye adsorption. Temperature ranging from 15˚C to 65˚C showed stability followed by a decrease in adsorption. Scanning electron microscopy of adsorbent particles showed a high surface porosity allowing the free passage of dye molecules.

Keywords: Adsorption; Dried *Padina pavonica*; Equilibrium; Fast Yellow Dye; Functional Groups

1. Introduction

Pollution is an environmental problem of worldwide concern. Consuming of water by agricultural, industrial and domestic sectors resulted in the generation of large amounts of waste water containing a number of pollutants [1]. Organic dyes in water are one of the important classes of pollutants [2]. It is difficult to treat dyes as they have a complex molecular structure which makes them more stable and difficult to be biodegraded [3,4]. The first known use of organic colorant appeared nearly 4000 years ago, when the blue dye indigo was found in the wrapping of mummies in Egyptian tombs [5]. There are more than 100,000 commercial dyes with a roughly estimated production of millions of tons per year [6-8]. Azo-dyes are one of the important classes of dyes which are characterized by an azo group consisting of two nitrogen atoms (–N═N–) as the chromophore in the molecule [9]. These dyes find application in many industries including textile, cosmetic, food colorants, printing, and pharmaceutical industries. The effluents of these industries tend to contain dyes in sufficient quantities. They cause coloration of water bodies once released into the aquatic environment and subsequently interfere with the transmission of light affecting aquatic communities' life [10]. Various treatment technologies such as precipitation, ion exchange, and adsorption have been employed to remove dye pollutants from aqueous solutions. Adsorption using low cost biological materials is generally regarded as an effective technique for the treatment of dye-containing wastewater [11-16]. One of the promising biological materials is the use of nonviable dried organisms. They have been proposed as potential sorbents; since they are dead materials having no need of nutrition to keep the biomass [17]. Algae have been found to be potential and suitable biosorbent because of their fast and easy growth as well as their wide availability. There were various researches on the usage of micro and macro algae as sorbent materials [2,18-21]. The sorption capability of algae has been attributed to their cell walls which are often porous and allow the passage of molecules and ions in aqueous solutions [22,23]. Essentially, the extracellular biopolymers of Phaeophyta are predominately alginic acid or alginate with a smaller amount of fucoidan which seems easily permeable for small ions [24]. Whereas the Rhodophyta contain a number of sulfated galactans like agar, carregeenan and porphyran [25]. Thus, the adsorption capacity along with the dye sorption process onto the surface is due to the different long chain extracellular biopolymers. Furthermore, algae functional groups found

on the algal cell surface such as hydroxyl, carboxyl, amino and phosphate and other charged groups are considered to be responsible for dye binding and separation of contaminants from water [26-29].

The examined biological material in this study is *Padina pavonica*. It is a widely distributed Mediterranean brown seaweed commonly known as peacock tail. The present work is to investigate the potential of this alga for the removal of fast yellow dye from contaminated aqueous solution. Effects of various operating parameters such as contact time, initial dye concentration, temperature and stirring speed are considered. The pseudo-first and second order equations are used for modeling the kinetics of the dye adsorption. The functional groups involved in the adsorption process were identified using FTIR analysis. Beside, adsorbent surface examinations are performed using scanning electron microscopy.

2. Materials and Methods

2.1. Algal Material

Fresh samples of *Padina pavonica* (L.) Lamouroux were collected from the coastal zone of Abu-Quir, Mediterranean Sea, Alexandria, Egypt during spring season. The alga grows on submerged rocks up to 50 cm depth. It has flattened fan-shaped thallus up to 15 cm. The thallus is calcified with concentric bands of hairs. The algal samples were thoroughly washed under running tap water to remove the surface adhered particles. Subsequently, it was air dried for about 30 minutes and oven dried at 45˚C to a constant weight. The dried biomass was crushed and stored in airtight container at room temperature for subsequent using.

2.2. Dye Solution

Acid fast yellow dye was kindly provided by dyestuffs and chemicals company (Elbeherah, Egypt). It was used without further purification. The molecular structure of the dye is shown in **Figure 1**. Stock solution was prepared at room temperature (25˚C ± 2˚C) by dissolving 1 g of acid fast yellow in 1L distilled water. The test solutions were prepared by diluting stock solution to the desired concentrations. Dye concentrations were measured at wave length 407 nm using Perkin Elmer UV-VIS spectrophotometer.

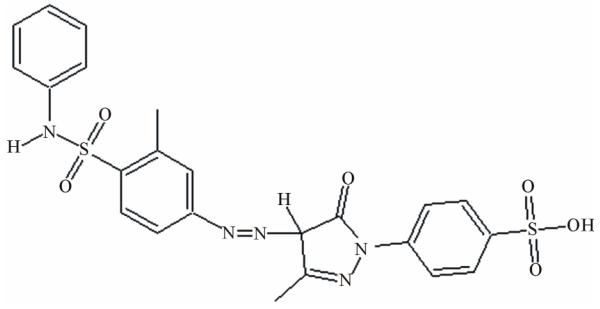


Figure 1. Chemical structure of acid fast yellow dye.

2.3. Adsorption Protocol

Adsorption experiments were carried out in 500 ml Erlenmeyer flasks by agitating 2.0 g dried alga with 200 ml desired dye concentrations (5, 10, 20, 40, 80 and 160 $mg \cdot L^{-1}$) at room temperature. The pH was kept without treatment. Samples were taken out at various time intervals (10, 20, 30, 40, 50, 60, 90, 120, 150 and 180 minutes) and sedimented by centrifugation at 5000 rpm for 10 minutes. The clear phase was subsequently analyzed for residual concentration of the dye at λ_{max}.

To express the percent of dye removal, the following equation was used:

$$\%\ \text{dye removal} = \left\{\left(C_i - C_e\right)/C_i\right\}100$$

where C_i = the initial dye concentration (mg dye$\cdot L^{-1}$), C_e = the equilibrium dye concentration (mg dye$\cdot L^{-1}$) at time t.

The amount of dye adsorbed, q (mg/g), was obtained as follows:

$$q = v\left(C_i - C_e\right)/M$$

where v = the volume of solution and M = the dry weight (g) of the adsorbent.

2.4. Adsorption Kinetic Studies

A study on the kinetics of adsorption is desirable as it provides information about the mechanism of adsorption; as well it describes how adsorbates will interact with an adsorbent. In order to characterize the kinetic behavior of the reaction and to fit the experimental data, two kinetic models are used:

1) The pseudo first-order lagergren expression [30] was the first equation for the adsorption of liquid/solid system based on the solid capacity. It can be expressed as:

$$\text{Log}\ \left(q_e - q\right) = \log q_e - \left(k_1/2.303\right)\ t$$

where q_e is the amount of dye adsorbed ($mg \cdot g^{-1}$) at equilibrium, q is the amount of dye adsorbed ($mg \cdot g^{-1}$) at time t (min) and k_1 is the rate constant of pseudo first-order adsorption (min^{-1}).

2) The pseudo-second kinetic rate law derived by Ho and Mckay [31], where the sorption capacity was assumed to be proportional to the number of active sites occupied on the sorbent. It can be expressed as:

$$t/q = \left(1/k_2 q_e^2\right) + \left(1/q_e\right)\ t$$

where k_2 is the pseudo second-order rate constant with a unit of $g \cdot mg^{-1} \cdot min^{-1}$.

The best-fit equilibrium model was selected based on the linear squared regression correlation coefficient, *R*2, values.

2.5. Sorbent Characterization

The functional groups of *Padina pavonica* were interpreted using the fourier transform infrared (FTIR) technique. A sample of adsorbent was mixed with approximately 0.5 g potassium bromide in the sample disk shortly before recording the spectra. The spectra were collected by Perkin-Elmer spectrum RXIFT-IR System within the range 500 - 4000 cm^{-1}.

2.6. Scanning Electron Microscopy

To study the surface texture, porous properties and morphology of the algal particles, scanning electron microscope examination was chosen. Samples were coated with a thin electric conductive gold film prior to use. Examinations of surface texture were performed using Jeol JSM-5300 scanning electron microscope.

3. Results and Discussion

3.1. Dye Concentrations and Sorbent Contact

The relationship between the time profiles and the dye removal percentage at various initial dye concentrations is shown in **Figure 2**. The adsorption rate of dye increased from 18.8% to 73.23% as the dye concentration increased from 5 to 160 mg/L. This can be attributed to an increase in surface area of the biosorbent, which in turn increases the binding sites. Generally, the adsorption rate is strongly influenced by several parameters including the state of the solid, availability of solute, and interference between reactive binding sites [32].

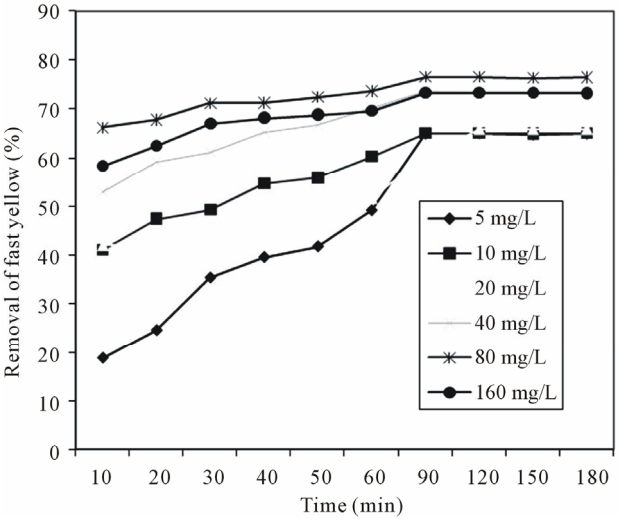


Figure 2. Effect of contact time on the adsorption rate of fast yellow by *Padina pavonica*.

3.2. Sorption Equilibrium Studies

In **Figure 3**, adsorption capacity (q) is correlated with time (min) at different initial dye concentrations (mg/L) keeping biosorbent weight as constant. It shows that most of the dye is adsorbed to achieve adsorption equilibrium in about 90 min except for 20 mg/L dye concentration; it was 120 min. At this point, the amount of dye being adsorbed onto the alga was in equilibrium state and no increase in loading capacity on the external surface of the adsorbent can take place. The overall adsorption is seen to consist of higher adsorption rate at the early period which may be due to the availability of more adsorption sites on the adsorbent surface [33]. As time passes, the adsorption rate is slow down due to the accumulation of the dye molecules in the vacant sites. This observation is consistent with the concept of non-homogeneity of the algal surface, which contains a variety of active sites. It serves as adsorption sites and may differ both with respect to the strength of the dye sorptive bond and the rate of adsorption onto the active sites [34,35]. Also it is more likely to note that the external diffusion is one of the rate-controlling steps of the initial fast adsorption of the dye onto the biosorbent [36]. In general, adsorption may be assumed to involve migration of dye from the bulk of the solution to the surface of adsorbent followed by diffusion of the adsorbate through the boundary layer and into the interior pore structure of the surface of adsorbent species.

3.3. Adsorption Kinetics

The principle behind the adsorption kinetics involves the search for a best model that well represents the interpretation of adsorption data. The pseudo-first and second order kinetic models, which are widely used to describe the adsorption kinetics, were applied. The best-fit model was determined based on the linear regression correlation coefficient values. Since the linear dependency was not

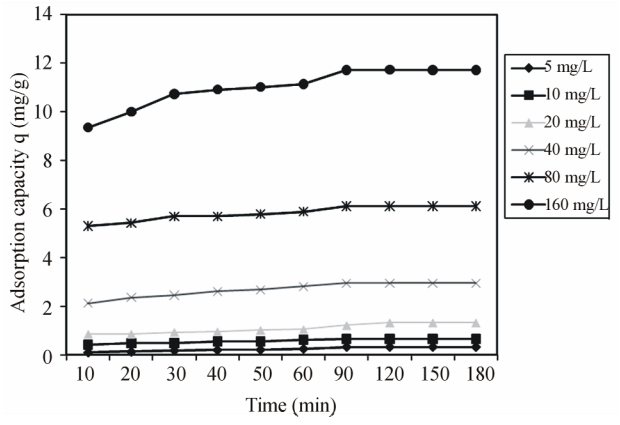


Figure 3. Adsorption capacity of fast yellow onto *Padina pavonica*.

obtained between log ($q_e - q$) and t (**Figure 4**), it can be said that the first-order equation of Lagergren does not fit well to the whole range of contact time. This suggests that the adsorption of dye onto the algal biomass is not a first-order reaction. The second order rate constant k_2 and q_e where determined by plotting t/q versus t (**Figure 5**). Correlation coefficient (R^2) together with the adsorption rate k_2 show that the pseudo second-order model is well in line with the tested experimental data. The linear plot between log ($q_e - q$) and t was detected and the correlation coefficients are nearly equal to 1 (**Table 1**). In the view of these results, the pseudo-second order kinetic model provided a good correlation for the adsorption of fast yellow onto the biosorbent in contrast to the pseudo-first order model.

3.4. Sorbent Characterization

FTIR spectroscopy involves collecting absorption information in the form of spectra. These spectra specify the absorption signals and the corresponding functional groups on the pure biomass surface to be compared with

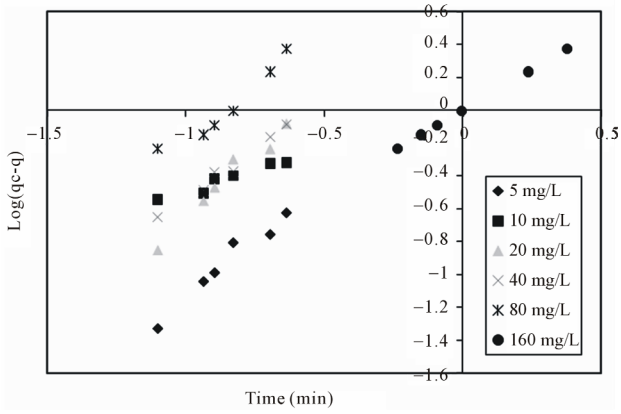


Figure 4. Pseudo-fist-order kinetic sorption of fast yellow onto *Padina pavonica*.

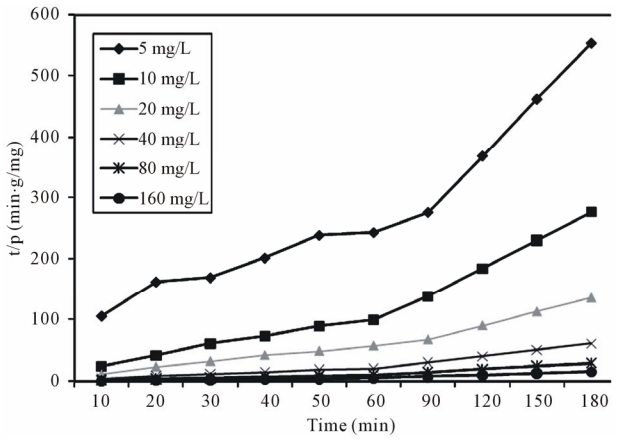


Figure 5. Plot of the pseudo-second-order model at different initial dye concentrations.

Table 1. Pseudo second order kinetic constants for the adsorption of fast yellow onto algal biomass.

Dye concentration (mg/L)	q_e (mg/g)	k_2 (g/mg·min)	R^2
5	0.325	0.0344	0.971
10	0.65	0.017	1.05
20	1.319	0.0063	1.084
40	2.95	0.0037	0.988
80	6.12	0.0018	0.96
160	11.72	0.00094	1.009

the referenced data of IR absorption. The FTIR spectra are in the range of 500 - 4000 cm^{-1}. It exhibits absorption bands at 3250, 2550, 3400, 1200, 3000, 1670 and 1128 cm^{-1} indicating the presence of OH, COOH, NH_2, S=O, C-H, C=O and C-O groups, respectively (**Figure 6**). This assumes that the adsorbent consists of a heterogeneous surface composed of different classes of adsorption sites. These sites provide information on the nature of cell wall and dye interaction. As well, they are involved in almost all potential binding mechanisms. It was found that carboxyl and hydroxyl groups are mainly responsible for dye sorption on *Padina*. However, other group functionalities showed less contribution in binding with dye. This result agreed with Murphy *et al*. [37] which reported that metal binding to brown seaweeds showed significant participation of carboxyl group accompanied by interaction of other groups. In fact, the relative importance of these functional groups depend on factors such as the quantity of sites, their accessibility, chemical state and affinity between site and dye [37,38].

3.5. Effect of Stirring Speed

The adsorption of dye as a function of stirring speed was investigated at room temperature and pH solution. Seven stirring speeds from 0 (without agitation) to 250 rpm were studied to determine the significant speed required for the optimal dye adsorption on dried alga. **Figure 7** shows that maximum dye removal percentage was obtained at 50 rpm stirring speed. Exceeding this speed, there is no significant effect on the adsorption and the dye removal percentage held almost with no variety. These results indicate that the increase in stirring speed improves the diffusion of dye molecules toward the surface of the seaweed. However, stirring speed up to 50 rpm is sufficient to assure that all the cell wall binding sites are accessible for dye uptake. Afterward, the effect of external film diffusion on adsorption rate is not significant [34,39].

3.6. Effect of Temperature

The temperature dependence of dye adsorption onto the dried biomass was studied at 15˚C, 25˚C, 35˚C, 45˚C,

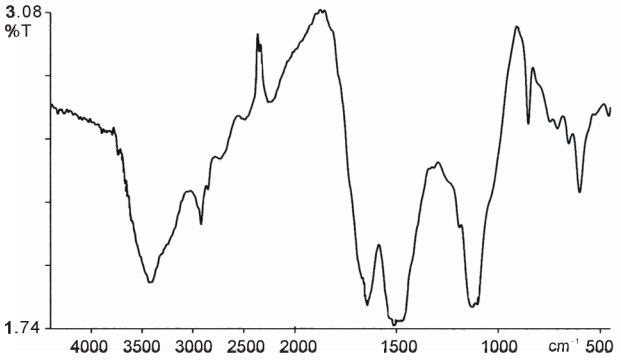


Figure 6. Peaks for nonliving biomass of *Padina pavonica* obtained from FTIR analysis.

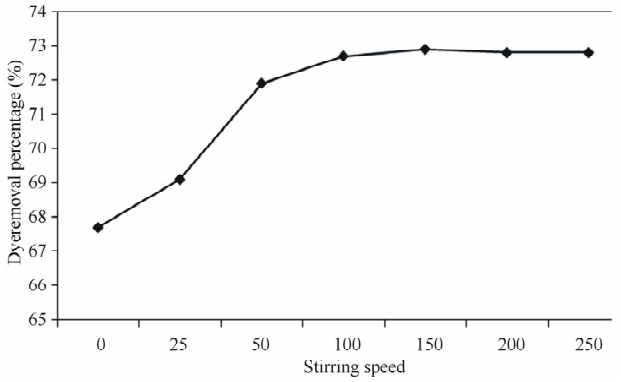


Figure 7. Effect of stirring speed on adsorption rate of dye onto non-living biomass.

55˚C and 65˚C keeping other parameters as constant. The temperature effect shown in **Figure 8** recorded relatively slight differences in dye adsorption from 68.7% to 69.5% as the temperature increase from 15˚C to 35˚C. This may be due to an increase in the mobility of the adsorbate molecules and the existence of the pores on the surface of the adsorbent particles. Hussain *et al*., Meena *et al*. and Seki *et al*. [38,40,41] noted similar observations and they suggested that the increase in temperature increase the rate of diffusion of the adsorbate molecules across the external boundary layer and in the internal pores of the adsorbent particle. This is due to the total volume and the possibility of the adsorbent pores, an increase of number of active sites for the adsorption as well as an increase in the mobility of the adsorbate molecules. However, the dye uptake was found to decrease from 69.5% to 54.1% with temperature increase from 35˚C to 65˚C suggesting that adsorption between the alga and the dye was mainly physical adsorption, dominant at lower temperature. Various authors [42,43] reported that the dye adsorption decreases with the increase of solution temperature. This can be explained by the weakening of bonds between dye molecules and active sites of adsorbents for high temperatures. Moreover the rise in temperature may damage the active binding sites in the biomass.

3.7. Scanning Electron Microscopy

Scanning electron microscopy of biosorbent particles is represented in **Figure 9**. It shows a high surface porosity with numerous macropores and mesopores. These pores exhibit hole-like with rough surfaces suggesting that the examined biosorbent can tolerate superior dye adsorption

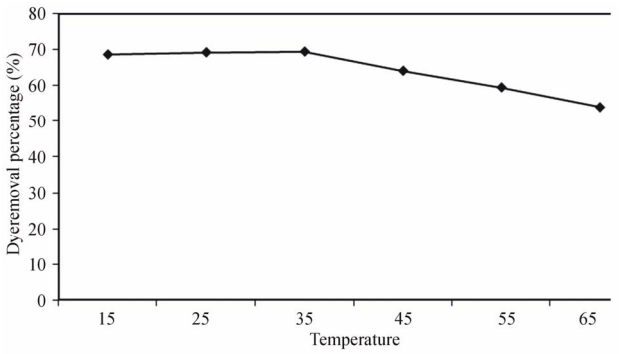
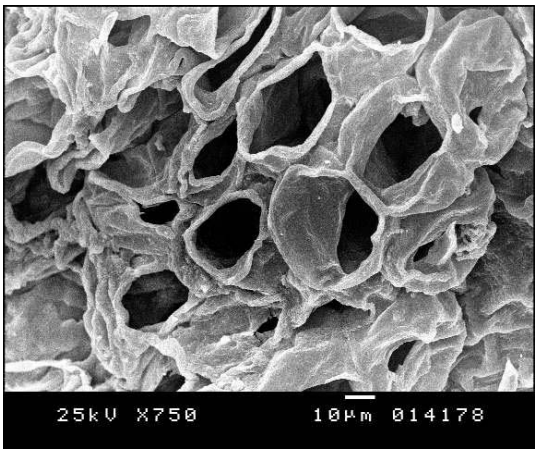
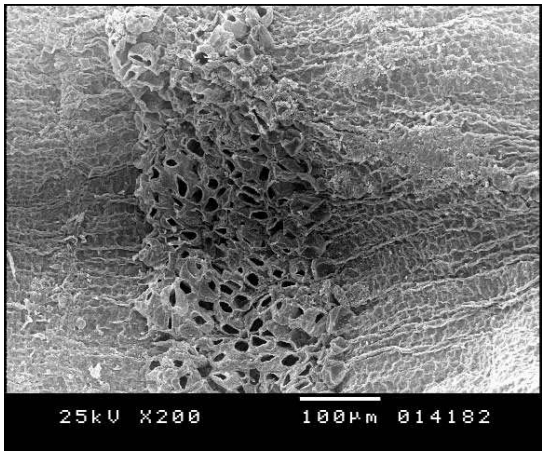


Figure 8. Dye removal percentage by dried algal biomass at different temperature.



(a)



(b)

Figure 9. Scanning electron micrographs of non-living algal particles.

and allow the free passage of dye molecules. Hussain *et al*. [38] noted similar observation as the increase in adsorption uptake of lead might be due to the possibility of porous structure on the non-living biomass *Padina pavonica* surface. In fact, the differences in adsorption capacities of different algae may be related to the morphological and compositional differences among the cell walls. Definitely, the pores may proof the increase of dye adsorption on the surface.

4. Conclusion

Focusing on the adsorptive capacity and the uptake mechanism, the ability of *Padina pavonica* for dye removal was investigated. Dye removal percentage increased as the initial dye concentration increased with the maximum removal percentage of 73.2%. The adsorption data were time-dependent and adsorption equilibrium was reached within 90 min. The sorption data corresponded well with the pseudo second-order kinetic model where the linear relationship was obvious and the correlation coefficients were between 0.96 and 1.084. Characterization of adsorbent proved the relationship between adsorptive properties and the surface groups of the adsorbent. Scanning electron microscopy informed that cell wall porosity may possibly increase the dye adsorption. No significant effect on the adsorption for stirring speeds greater than 50 rpm was observed. The adsorption capacity was optimal at 35˚C. Nevertheless, advances in knowledge to minimize dye in water body and to define more environmentally friendly chemicals are also needed.

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