

# Green Production of Biodegradable Mulch Films for Effective Weed Control

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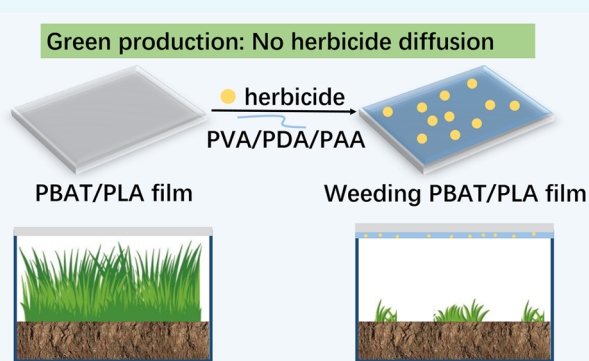


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Supporting Information

**ABSTRACT:** Biodegradable mulch films are supposed to be a prospective substitute for poly(ethylene)-based mulch films in the field of sustainable agriculture. Among them, weeding mulch films play significant roles. However, the large-scale production of weeding mulch films through the traditional high-temperature film blowing process would often cause serious pollution due to the diffusion of herbicides in the surroundings. Herein, a green and facile coating approach is developed to produce biodegradable weeding mulch films. In our strategy, a herbicide was added into a poly(vinyl alcohol) aqueous solution with dopamine in it. After the subsequent low-temperature coating procedure on a biodegradable poly(butylene adipate-*co*-terephthalate)/poly(lactic acid) film, effective weeding mulch films were obtained. The morphology, structure, and mechanical property test results revealed the robustness and stability of the coating, and the pot experiments clearly demonstrated the effective weed suppression ability of the obtained weeding films. Evidently, this strategy to produce biodegradable weeding mulch films is green and facile, exhibiting great prospects in the large-scale production of weeding mulch films and other functional biodegradable mulch films.



## 1. INTRODUCTION

Agricultural mulches have been widely used in many countries to raise the output and quality of crops. Mulch films can control the growth of weeds, increase the temperature and humidity of the soil, and protect crop species from extreme weather and pest threat.<sup>1</sup> Moreover, it has been reported that the mulches can also strengthen the efficiency of fertilizers and pesticides, which could help reduce their consumption and subsequently decrease the production cost.<sup>2–4</sup> The total amount of plastic films used in greenhouse and mulching over the world was expected to reach 7.4 million tons in 2019, and it demonstrated a huge increase of 69% compared with 4.4 million tons in 2012.<sup>5,6</sup> However, most of these plastic films are made of poly(ethylene) (PE), poly(propylene), poly(vinyl chloride), and poly(styrene), which could bring serious environmental problems since these materials are non-biodegradable.<sup>7–11</sup> For this reason, many kinds of novel biodegradable films have been developed using biodegradable polymers or regenerative resources, such as poly(butylene adipate-*co*-terephthalate) (PBAT), poly(lactic acid) (PLA), poly(propylene carbonate) (PPC), poly(hydroxyalkanoate) (PHA), or cellulose, starch, fiber, and proteins.<sup>12–18</sup> The production of biodegradable mulch films has also become a focus in the research field of sustainable agricultural practices.<sup>19–21</sup> Furthermore, to meet the need of increasing

requirements, plastic films with different functions have been manufactured.<sup>22–24</sup>

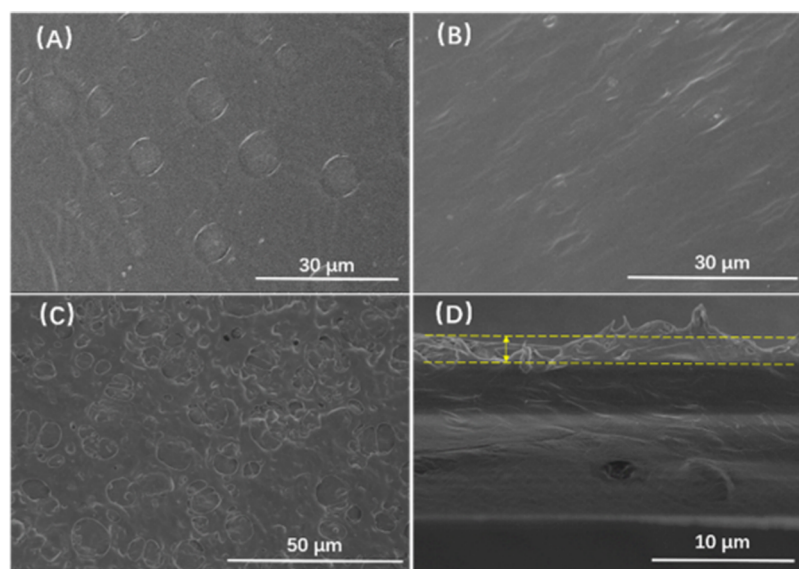
Among these various films, weeding mulch films play significant roles in the agriculture field.<sup>25,26</sup> Although plain mulch films could have a certain weeding function, for some specific areas, their weeding effect was not good enough.<sup>27</sup> Therefore, some agrochemicals are employed to improve the weeding effect.<sup>28,29</sup> Nevertheless, the direct use of herbicides can bring some difficult problems, including early decomposition or oxidation of bioactive ingredients before they were absorbed by the crop species and serious surface runoff caused by rain wash and other reasons in the period of crop growth. Based on the above situations, mulch films embedded with herbicides have been developed in recent years.<sup>30</sup> Researchers blended herbicides or modified herbicides into biodegradable monomers to produce weeding mulch films via the casting or hot-pressing method; however, these methods were not quite suitable in large-scale industrial production.<sup>31,32</sup> As a matter of

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**Figure 1.** SEM images of the (A) PBAT/PLA-PVA film, (B) PBAT/PLA-PVA-PDA film, (C) PBAT/PLA-PVA-PDA-PAA film, and (D) cross section of the PBAT/PLA-PVA-PDA-PAA film.

fact, high-temperature film blowing or multilayer coextrusion processes are often used in the production of weeding plastic films in factories. Unfortunately, during these high-temperature production processes, plenty of herbicides would diffuse out from the mulch films and spread to the whole production workshop and even the surrounding villages and places, which would bring serious pollution and harm to human beings and the ecological environment.

In this study, to solve the pollution and harm caused by the high-temperature process, a facile and green coating approach is developed to produce biodegradable weeding mulch films. Coating technology can avoid high-temperature processes, but how to form a stable and firm coating is still a challenge. In our strategy, a herbicide was added into a poly(vinyl alcohol) (PVA) aqueous solution with dopamine in it, which would help the above liquid adhere to the biodegradable mulch film better in the subsequent coating process.<sup>33,34</sup> The obtained coated film could effectively suppress the growth of different kinds of weed species. Importantly, this coating process can be finished at a temperature below 60 °C, which would efficiently avoid the harmful pollution generated from the diffusion of the herbicide in a high-temperature producing process. Especially, compared with the casting or hot-pressing method, the coating technology is more suitable for large-scale weeding mulch film production, and the achieved weeding mulch films are biodegradable.

## 2. RESULTS AND DISCUSSION

**2.1. Morphology of the Coated PBAT/PLA Film.** The thickness of commercial PBAT/PLA mulch films used in our coating experiments was about 10  $\mu\text{m}$ , and generally, it is not easy to coat on films with such a thin thickness. Herein, PVA was selected as the main coating compound, which was also biodegradable.<sup>35,36</sup> Furthermore, the hydroxyl groups of PVA could form hydrogen bonds with the residual carboxyl groups of PLA and all ester groups in PBAT and PLA, which was beneficial to the firmness of the coating. The surface morphologies and cross-sectional morphologies of the films were carefully characterized and are shown in Figure 1.

It was seen that the surface of the PBAT/PLA-PVA film was smooth but with some holes (Figure 1A). The smooth surface revealed the successful spread and adhesion of PVA on PBAT/PLA, and the holes indicated that there still existed some regions where the hydrogen bonds were not formed. To strengthen the firmness of the coating, dopamine was introduced, which could self-polymerize into polydopamine (PDA) and was facilely immobilized on the PBAT/PLA substrate as revealed in the literature.<sup>37</sup> As demonstrated in Figure 1B, there were no more holes on the PVA-PDA-coated surface. Although PVA could adhere to the substrate surface well, the abundant hydroxyl groups of PVA would also make the coated surface too sticky to be used in practical applications due to their water-absorbing quality. In that case, PAA was employed to cross-link with the redundant hydroxyl groups of PVA, which would help the surface remain dry and steady. It can be seen from Figure 1C that the coated surface of the PBAT/PLA-PVA-PDA-PAA film was not as smooth as that of PBAT/PLA-PVA-PDA, and some grains emerged on the surface.

Importantly, since the thickness of the mulch film was very thin, the thickness of the coating should also be thinner to make sure the firmness of the coating. The obtained PBAT/PLA-PVA-PDA-PAA film was quenched and broken off by liquid nitrogen for SEM characterization. However, since the PBAT/PLA-PVA-PDA-PAA film was as thin as only about 13  $\mu\text{m}$ , the section of the film was not quite smooth, and there were some deformations in the coating layer. The cross section of the PBAT/PLA-PVA-PDA-PAA film was characterized, as revealed in Figure 1D, and the yellow dotted line indicated the position of the coating. It can be seen that the thickness of the coating was as thin as only 2–3  $\mu\text{m}$ , and no obvious interfacial separation between the PVA-PDA-PAA coating and the PBAT/PLA mulch film was observed, indicating their good compatibility.

**2.2. FTIR Analysis.** The FTIR spectra of the PBAT/PLA film and the coated mulch film are demonstrated in Figure 2. The peak at about 1575  $\text{cm}^{-1}$  is ascribed to the C=C bond of the aromatic ring, most of which comes from the aromatic ester in PBAT. The peaks at 2850 and 2980  $\text{cm}^{-1}$  indicated the

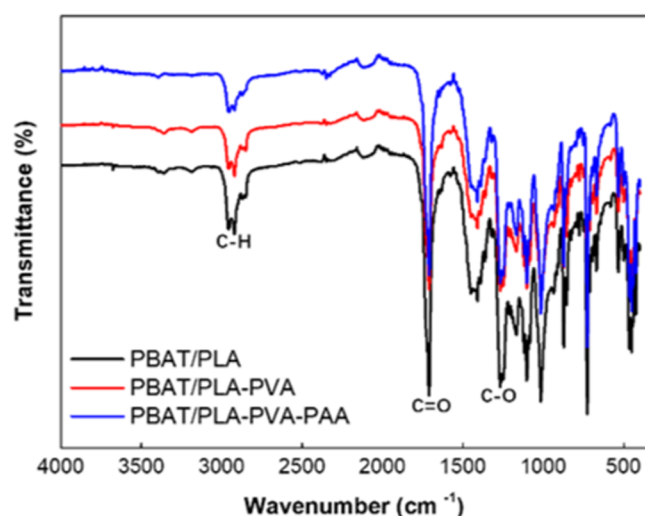


Figure 2. FTIR spectra of films with different coatings.

symmetrical and asymmetrical stretching of C–H as described in the literature.<sup>38</sup> A characteristic peak at around  $1740\text{ cm}^{-1}$  can be ascribed to all C=O group absorption of the ester bonds, including all aliphatic ester and aromatic ester from both PBAT and PLA.<sup>39</sup> It can be seen that the intensity of the C=O peak in PBAT/PLA-PVA was similar to that in PBAT/PLA, but the intensity of the C=O peak in PBAT/PLA-PVA-PAA was higher, which can be explained by the fact that the ester bond was increased in PBAT/PLA-PVA-PAA. Likewise, the peak at  $1250\text{ cm}^{-1}$  indicated the stretching vibration of C–O in the ester bond, which was also increased in PBAT/PLA-PVA-PAA compared with those in PBAT/PLA-PVA and PBAT/PLA. Therefore, PAA would react with PVA and form new ester bonds in the coating. Generally, these spectra were similar since both the film and the coating were very thin. Moreover, since the amount of PDA was very small in the coated film, there were no distinctive differences in the FTIR spectra of PBAT/PLA-PVA-PDA and PBAT/PLA-PVA-PAA-PDA films (Figure S1). Nevertheless, the coated film with PDA in it revealed a light brown color, which can be easily distinguished and proved the successful coating of PDA.

**2.3. Mechanical Properties.** The mechanical properties of the film play significant roles in their practical applications. Figure 3A shows the tensile strength of the PBAT/PLA-PVA, PBAT/PLA-PVA-PDA, PBAT/PLA-PVA-PAA, and PBAT/

PLA-PVA-PAA-PDA films. The tensile strength of the PBAT/PLA-PVA film is about 6.8 MPa, while the tensile strength of the PBAT/PLA-PVA-PDA film is about 7.9 MPa. Moreover, there existed obvious scaling off of the coatings in the PBAT/PLA-PVA film during the test, but the PBAT/PLA-PVA-PDA film remained intact. This phenomenon indicated that the employment of PDA would successfully enhance the firmness of coatings, which was consistent with the above SEM results. It can also be seen from Figure 3A that the tensile strength of the PBAT/PLA-PVA-PAA and PBAT/PLA-PVA-PAA-PDA films showed an evident increase of 10.2 and 10.1 MPa, respectively. The improvement of their mechanical properties can be attributed to the reaction between the hydroxyl groups of PVA and the carboxyl groups of PAA, and the large number of hydrogen bonds between PVA and PAA also played an important role that cannot be ignored.

The elongation at break of the PBAT/PLA-PVA, PBAT/PLA-PVA-PDA, PBAT/PLA-PVA-PAA, and PBAT/PLA-PVA-PAA-PDA films was tested, as illustrated in Figure 3B. Contrary to the tensile strength, elongation at break decreases from 422.9% for PVA-coated mulch films to 343.2% for PVA-PDA-coated mulch films, and the elongation at break also decreases from 509.3% for PVA-PAA-coated mulch films to 405.5% for PVA-PDA-PAA-coated mulch films. On the one hand, it can be seen that with the employment of PAA in the coating, the elongation at break shows an obvious improvement due to the high cross-linking density through esterification and hydrogen bonding between PAA and PVA. On the other hand, after the introduction of PDA, there was a clear decrease in their elongation at break, which can be ascribed to the fact that the addition of PDA reduced the high flexibility of the previous systems. Nevertheless, although the tensile strength and elongation at break of PVA-PDA-PAA-coated mulch films were slightly smaller than those of PVA-PAA-coated mulch films, the firmness of PVA-PDA-PAA-coated mulch films was much better than that of PVA-PAA-coated mulch films. Therefore, by comprehensively considering the influence of the above factors, PVA-PDA-PAA-coated mulch films were more prospective for practical applications.

**2.4. UV–Vis Characteristic Absorption Wavelength of Acetochlor.** The UV absorption curve of acetochlor is demonstrated in Figure S2, in which the acetochlor standard solution showed a characteristic absorption peak at 204 nm. In that case, the standard curve of the acetochlor solution was created with its absorbance value at 204 nm as abscissa and its

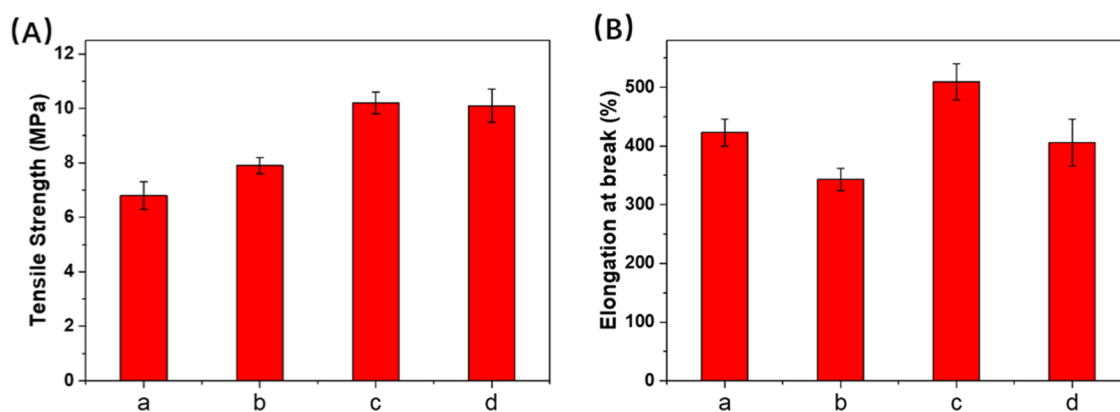
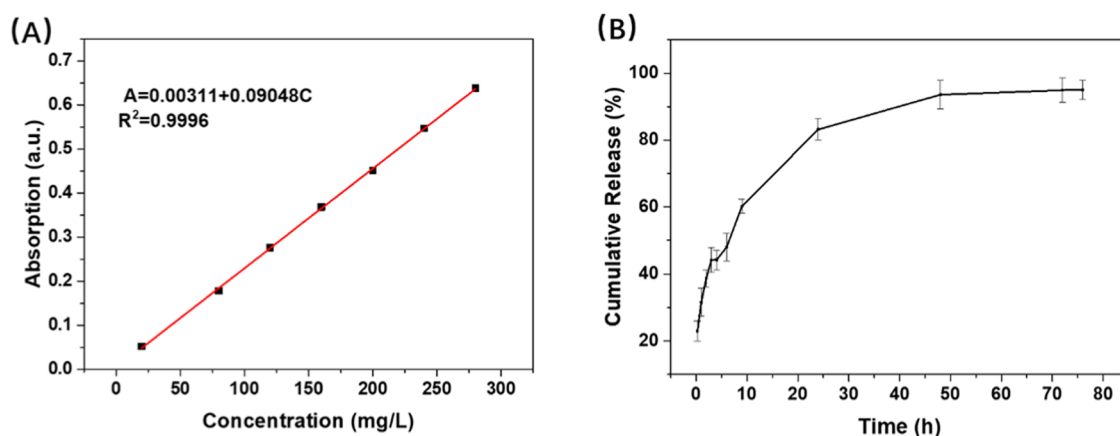


Figure 3. (A) Tensile strength and (B) elongation at break of the (a) PBAT/PLA-PVA film, (b) PBAT/PLA-PVA-PDA film, (c) PBAT/PLA-PVA-PAA film, and (d) PBAT/PLA-PVA-PAA-PDA film.



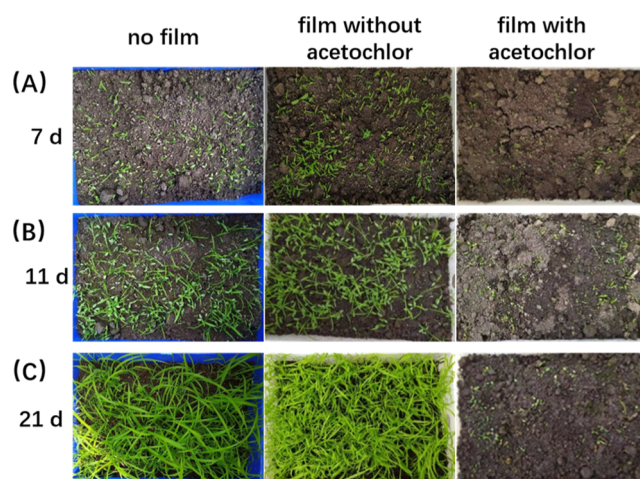
**Figure 4.** (A) Linear relationship between  $C$  (concentration) and  $A$  (absorbance) of acetochlor. (B) Releasing profile of acetochlor from the PBAT/PLA-PVA-PAA-PDA-acetochlor film.

concentrations as ordinate. As revealed in Figure 4A, the absorbance value and concentration of the acetochlor solution exhibit a linear positive correlation in a concentration range of 20–280 mg/L. After careful computation, the linear relationship between  $C$  (concentration) and  $A$  (absorbance) can be expressed by the following equation:  $A = 0.00311 + 0.09048C$ ,  $R^2 = 0.9996$ . This equation can be used for calculating the released amount of acetochlor in the subsequent releasing tests.

**2.5. Releasing Experiment.** Figure 4B shows the releasing profile of acetochlor from the PVA-PAA-PDA-coated mulch film with acetochlor loaded in it.

The releasing test was completed at room temperature by putting the film in methanol under continuous stirring. As can be seen, the cumulative release curve indicated that nearly 30% of acetochlor was released from the film within 3 h, and about 48% of acetochlor was released from the film within 4 h. This fast release of acetochlor in the early stage of the releasing test was because acetochlor had good solubility in methanol, which would lead to the burst release. After that, the releasing rate gradually decreased, and the cumulative release of acetochlor was nearly stable from 48 to 72 h, which reached 95% within 72 h.

**2.6. Weeding Effect.** Not only the production procedure of biodegradable mulch films is green without the generation of pollution, but also the weeding effect of the films is not inferior compared with other herbicide-containing films.<sup>29</sup> To test the weeding effect of the coated mulch film, the PBAT/PLA-PVA-PAA-PDA-acetochlor film was fabricated with the herbicide acetochlor mixed in the coating layer. Three groups were set for the weeding experiment, and the grass in each group had the same growth rate. Group 1 was with no mulch film, group 2 was covered with the PBAT/PLA-PVA-PAA-PDA film, and group 3 was covered with the PBAT/PLA-PVA-PAA-PDA-acetochlor film. On the first day, grass seeds were scattered in the soil of all of the groups equally. As demonstrated in Figures 5 and 6, after 7 days, many grass seedlings germinated in both groups 1 and 2. By comparison, there were also some grass seedlings germinated in group 3, but the number of grass seedlings in group 3 was quite less. On the 11th day, the grass seedlings in groups 1 and 2 showed an obvious growth, while the growth of seedlings in group 3 was very slow. Not only the number of grass seedlings in group 3 was quite less, but also the plant height was much lower. Compared with other literature studies,<sup>29</sup> we also investigated

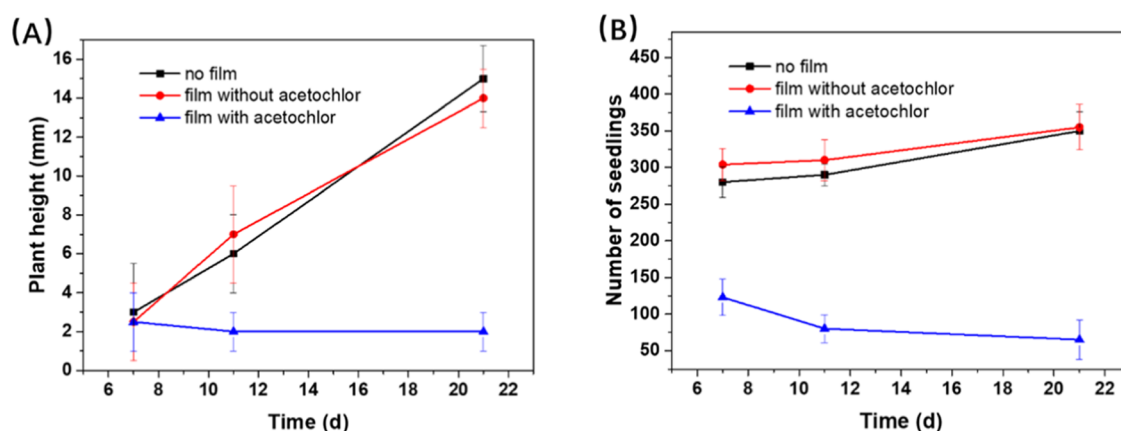


**Figure 5.** Herbicidal test of three groups (no film covering, the film without acetochlor covering, and the film with acetochlor covering): (A) 7 days, (B) 11 days, and (C) 21 days after the film covering treatments.

the weeding effect of the mulch films for a longer time. After 21 days, the grass seedlings in group 1 had grown up luxuriantly, and the grass in group 2 had also grown well under the mulch film. In sharp contrast, the height of grass seedlings in group 3 remained as it was without growth, even some grass seedlings were wilted. These records strongly proved that the PVA-PAA-PDA-acetochlor-coated mulch film had great herbicidal ability, which could effectively prevent the growth of different weeds.

### 3. CONCLUSIONS

In summary, a biodegradable weeding mulch film was produced via a green and facile strategy without any herbicide diffusion pollution in this process. In this study, a herbicide was added into the PVA systems as a weeding coating, which was also biodegradable. Furthermore, PDA was introduced to help form a robust and stable coating layer. This coating method was completed at a low temperature of below 60 °C, efficiently avoiding the harmful pollution generated from the diffusion of the herbicide in the high-temperature producing process. Moreover, the obtained weeding film showed great weed suppression ability against different kinds of weed species in the pot experiments, which strongly proved its efficient



**Figure 6.** (A) Plant height of seedlings in three groups after different days. (B) Number of seedlings in three groups after different days.

weeding effect. Importantly, compared with the casting or hot-pressing method, the coating technology in our work is green and more suitable for large-scale weeding mulch film production, which also exhibits great prospects in the large-scale production of weeding mulch films and other functional biodegradable mulch films.

## 4. EXPERIMENTAL SECTION

**4.1. Materials.** Poly(acrylic acid) (PAA) with an average  $M_v$  of 1 250 000, poly(vinyl alcohol) (PVA) with an alcoholysis degree of 86.5–89.0%, dopamine hydrochloride (98%), and acetochlor were purchased from Aladdin Biochemical Technology Co., Ltd (Shanghai, China). Tris-(hydroxymethyl) aminomethane (Tris) and methanol were obtained from Macklin Biochemical Technology Co., Ltd (Shanghai, China). The biodegradable mulch film was a PBAT/PLA film, which was prepared in our laboratory with a blown film machine (XH-432-25, Xihua Testing Machine Co., Ltd), using a commercial masterbatch (FLEX-262 F20) obtained from Kingfa Sci & Tech Co., Ltd (Guangzhou, China). All of the reagents were used directly without further purification.

**4.2. Preparation of the Coated PBAT/PLA Film.** First, 15 g of PVA was dissolved in 150 mL of water under continuous stirring for 3 h at 90 °C, which was kept at room temperature for further use. Afterward, 3 g of PAA was dissolved in 150 mL of water in the same way as described above. PAA was kept still until defoaming. Then, 120 mL of the PVA solution and 30 mL of the PAA solution were mixed and stirred for about 2 h at room temperature. After that, 200 mg of acetochlor (dissolved in 1.5 mL of acetone) and 1.5 mL of Tris-HCl buffer (pH 8.5) were added into the above-mixed solution and further ultrasonic treatment of 30 min was carried out. The obtained liquid was stored at 4 °C for the further coating procedure.

Before the coating process, 20 mg dopamine hydrochloride was added into 10 mL above prepared coating liquid with the final dopamine hydrochloride concentration of 2 mg/mL, which was used as the final coating liquid. The coating process was completed by a bar coater. Then, the achieved PBAT/PLA-PVA-PAA-PDA-acetochlor film was put in an oven of 50 °C for about 0.5 h.

**4.3. Characterization of the Coated PBAT/PLA Film.** Scanning electron microscopy (SEM) was used for the morphology examination of the mulch films using a field

emission scanning electron microscope (SU-70, Hitachi). Before the examination, the coated film was cut into long strips and frozen in  $-80$  °C liquid nitrogen for 1 h, and then the stripes were quickly broken and the cross sections were used for the SEM test. Fourier transform infrared (FTIR) spectra were recorded using a Bruker Vector 33 spectrometer. Mechanical properties were tested according to the Chinese National Standard GB/T 528-2009. The measurements were conducted using a testing machine (CMT5505, MTS Systems), and the cross-head speed was set as 50 mm/min. For each film, five replicates of film specimens were tested. UV-vis absorption spectra were monitored using an ultraviolet-visible spectrophotometer (TU-1901, Puxi).

**4.4. Release Testing.** First, the standard curve of acetochlor was tested. Standard aqueous solutions of acetochlor with concentrations of 20, 80, 120, 160, 200, 240, and 280 mg/L were prepared. After that, the UV absorption spectrum of each standard solution was measured to determine the relationship between the solution concentration and absorption.

For the release experiment, about 400 mg of the PBAT/PLA-PVA-PAA-PDA-acetochlor film was soaked in 100 mL of water under gentle stirring. At set intervals, 1 mL of release medium was removed and replenished by the same volume of fresh release medium at the same time. The released quantity of acetochlor was obtained by UV-vis spectra.

**4.5. Weeding Experiment.** To evaluate the weed control efficacy of the PBAT/PLA-PVA-PAA-PDA-acetochlor film, 1 g of grass seeds (including 0.25 g of barnyard grass seeds, 0.25 g of crabgrass seeds, 0.2 g of amaranth seeds, 0.1 g of Ox grass seeds, and 0.2 g of longclaw grass seeds) were sown in plastic pots (length 352 mm, width 250 mm, and height 95 mm). Three groups were set for the weeding experiment. The number of grass seeds in each group is the same, and the grass seeds have the same growth rate under the same growth environment. Group 1 was with no mulch film, group 2 was covered with the PBAT/PLA-PVA-PAA-PDA film, and group 3 was covered with the PBAT/PLA-PVA-PAA-PDA-acetochlor film. Each group had three replicates. The growth of these grass seeds was monitored and studied at set time intervals.

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsomega.1c05725>.

FTIR spectra of films with different coatings and the absorption curve of acetochlor (PDF)

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### Notes

The authors declare no competing financial interest.

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