



# A novel process for obtaining high quality cellulose acetate from green landscaping waste

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## ABSTRACT

As a typical lignocellulosic biomass, abundant amount of green landscaping waste can be used as low-cost feedstock to produce value-added chemicals. This work developed a novel process to obtain industrially important cellulose acetate from green landscaping waste after separation of hemicellulose as a substrate rich in monomeric xylose. Sixteen samples of selected green waste were pretreated with diluted acid at moderate temperatures. Afterwards, the residues were treated with acetic anhydride and a small amount of sulfuric acid as catalyst to make cellulose acetate. The quantity and quality of cellulose acetate were analyzed by Fourier Transform Infrared Spectroscopy (FT-IR), Nuclear Magnetic Resonance (<sup>1</sup>H NMR), and Gel Chromatography (GPC). The activation mechanisms of the diluted-phosphoric-acid pretreatment on yields and quality of cellulose acetate were also investigated using Thermogravimetric Analyzer (TGA) and Scanning Electron Microscope (SEM). Without the pretreatment, the yields of cellulose acetate from green landscaping waste ranged from 4.0% to 8.6%. After the pretreatment the mean yield of cellulose acetate from all sixteen green waste significantly increased to approximately 35%, illustrating the high efficiency and wide applicability of our proposed method.

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## 1. Introduction

The current disposal methods for municipal and livestock waste have led to a serious problem due to their high potential for pollution and increasing amount of production. Green landscaping waste (GLW) is an important component of municipal solid waste. By the end of 2015, China's urban-landscape-area is more than 1,888,000 ha, indicating a green coverage rate of 36.34% and a per capita green area of 13.16 m<sup>2</sup>. The annual GLW output of China is more than 250 million tonnes, of which less than 10% is recycled (Lyu and Chen, 2016). With the continuous support for ecological constructions and urban greening processes, the disposal of GLW will considerably increase owing to the insufficiency in recycling program. Most GLW are disposed together with household waste,

which disrupt the ecological cycle of the soil, resulting in soil infertility and environmental pollution (Chauhan and Singh, 2016). Landfill, incineration, composting, and carbonization are the traditional methods for the treatments of GLW (Lazzerini et al., 2016). Although landfill operation is simple, extensive land resources are necessary and the generation of leachate, stench, and greenhouse gases harms the surrounding environment (Oppio and Corsi, 2017). Incineration can reduce the volume of waste but air pollution is often associated with improper technical means and operations (Lokonon, 2016). More countries are introducing a ban on open burning of GLW (Lyu and Chen, 2016). The efficiency of composting is low owing to the intrinsic anti-microbial-degradation property of cellulose and lignin (Karnchanawong et al., 2017). The energy consumption of GLW carbonization is high and much technical study is still needed (Lin et al., 2017). Developing a new, cost-effective, and eco-friendly technology to utilize GLW is important.

As a typical lignocellulosic biomass, GLW contains about 30–60% carbohydrate polymers (cellulose and hemicellulose) that can be used as a low-cost feedstock for the production of bio-oil

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## Nomenclature

GLW	green landscaping waste
CA	cellulose acetate
DS	degree of substitution
DP	degree of polymerization
UP	untreated pinewood
P22	pretreated pinewood under the conditions of run No. 22 listed in Table 3
UP-CA	CA derived from UP
P22-CA	CA derived from P22
HPLC	High Performance Liquid Chromatography
FT-IR	Fourier Transform Infrared Spectroscopy
<sup>1</sup> H NMR	Nuclear Magnetic Resonance
GPC	Gel Chromatography
TGA	Thermogravimetric Analyzer
SEM	Scanning Electron Microscope

(Cao et al., 2016b), bio-ethanol (Chen and Fu, 2016), and chemicals (Yu and Tsang, 2017). The conversion of lignocellulose to ethanol includes four steps: (1) pretreatment, (2) enzymatic hydrolysis, (3) mixed sugar fermentation, and (4) product recovery. Lignocellulosic biomass is generally resistant to enzymatic attack (Lozano and Lozano, 2017). It is essential to implement proper pretreatment prior to enzymatic saccharification. Diluted acid pretreatment at moderate temperatures (100–180 °C) has become one of the most widely employed technologies for various biomass feedstocks (Liu et al., 2017b). This process can be achieved by the hydrolysis of hemicellulose to obtain monosaccharides (mainly xylose). Although acid helps to reduce the crystallinity of cellulose, it has no capacity to transform cellulose to glucose. Cellulases are used to hydrolyze cellulose to glucose, which is slow and not economical (Chng et al., 2016). It is estimated that the cost of cellulase needs to be reduced by at least 10 fold in order to make the process competitive (Domínguez et al., 2017).

As an alternative, we aimed to convert cellulose to value-added cellulose acetate after removing hemicellulose as a substrate rich in monomeric xylose and arabinose. The market price of cellulose acetate is approximately 11 USD/kg (Cellulose acetate on Alibaba, 2017). Xylose is a kind of pentose that is an important raw material for producing xylitol (Araújo et al., 2017) and furfural (Lopes et al., 2017). Arabinose is also an important pentose that can be used for furfural production (Hongsiiri et al., 2015). This substrate

can also be used for ethanol fermentation (Caballero and Ramos, 2017). Converting the cellulose from biomass residues into cellulose acetate instead of glucose may reduce the total cost of ethanol production from lignocellulosic biomass. It is estimated that the annual production of cellulose acetate reaches 680 thousand tonnes around the world. Because of its low cost, high toughness, gloss, transparency, natural feeling, and other good aesthetic properties, cellulose acetate has been widely used in manufacturing paint, textile fiber, cigarette filter, packing material, film, artificial kidney, and reverse osmosis membrane (Goetz et al., 2016). There have been many recent publications on the production of cellulose acetate from lignocellulosic biomass. The primary raw materials for the synthesis of cellulose acetate are virgin wood (1300 USD/t in China) and cotton pulp (1050 USD/t in China) which are rather expensive (Khaparde, 2017). The exploration for alternative and economical raw materials to reduce costs has become the focus of cellulose acetate research (Candido et al., 2017). In China, the total collection and transportation cost for GLW is only 90 USD/t (Tong and Tao, 2016). Although some scholars have reported the utilizations of lignocellulosic waste like sugarcane straw (Candido and Goncalves, 2016) and rice husk (Fan et al., 2013) for cellulose acetate production, the alkali pretreatment was employed to obtain cellulose by removing lignin and hemicelluloses with no value-added byproducts was produced.

In the present work, sixteen selected GLW were pretreated with diluted phosphoric acid at moderate temperatures. Xylose and arabinose were obtained as value-added byproducts during this pretreatment. Afterwards, the treated and untreated feedstocks were subjected to acetylation reaction to produce cellulose acetate. The activation mechanisms of the diluted-phosphoric-acid pretreatment on cellulose acetate yields and quality were also investigated.

## 2. Materials and methods

### 2.1. Materials

All the GLW were collected in the campus of Fudan University in March 2014 (spring) and were fresh materials from landscaping plants trimming (0.3–1.5 m in length, 0.5–1.5 cm in diameter). The selected GLW were different with each other in state of leaves, morphology, growth habit, wood type, and botanical classification (Table S1, Supporting Information), making their good representativeness. The plants samples were dried in an oven (104 °C, 8 h), pulverized and screened into particles with a diameter of 40–60 mesh. Cellulose, hemicelluloses, and lignin content in the plants (Tables 1 and 2) were analyzed following the protocol from the

**Table 1**  
Composition and ultimate analysis of leaves (wt%).

Leaves	<i>Pinus</i>	<i>Cupressus funebris</i>	<i>Platanus</i>	<i>Cinnamomum camphora</i>	<i>Pittosporum tobira</i>	<i>Distylium racemosum</i>	<i>Viburnum odoratissimum</i>	<i>Salix babylonica</i>
Components								
Cellulose	22.9	24.8	22.7	23.5	18.2	23.7	8.3	23.2
Hemicellulose	15.0	16.9	16.1	12.9	17.6	16.3	17.5	17.3
Lignin	31.0	16.8	23.6	16.8	10.6	23.8	20.1	18.3
Ash	1.3	7.4	6.0	8.0	10.7	10.0	9.8	8.8
Ethanol extracts	8.5	8.5	5.4	5.4	9.1	6.1	8.1	4.9
Others	21.4	25.6	26.3	33.5	33.8	20.1	36.2	27.6
Ultimate								
C	46.6	43.0	45.3	43.5	45.2	46.9	44.6	45.4
H	7.4	6.8	7.7	7.4	7.3	7.3	7.6	7.4
O <sup>a</sup>	44.7	49.6	46.2	47.8	46.4	44.8	45.0	47.0
N	1.3	0.7	0.7	1.4	1.1	1.0	2.8	0.2
S	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2

<sup>a</sup> Calculated by difference.

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