Study of pulping technology on disposable tableware using withered leaves as raw material

Liu bo-fan¹, Liu Ming-dan¹*, Jin Peng², Zhong Yi-xin³

College of Mechanical and Electrical Sichuan Agricultural University 625014, Yaan Sichuan, China

e-mail: 276591355j@qq.com, 2296482900@qq.com, 1048721651@qq.com, 844929149@qq.com
TEL: 13086317002

Abstract. Disposable tableware is widely used around world due to its advantages such as sanitation, thermal preservation and low price. However, more and more countries are restricting the use of disposable tableware due to its difficulties in recycling and degradation in the natural environment. If disposable tableware is made of photobiodegradable PP plastic or high-impact polystyrene (HIPS), it is difficult to degrade and has a great impact on the earth environment. In this study, four kinds of withered leaves were selected as raw materials and mechanical chemical pulping method and caustic soda pulping method were used for experiments. The main processes include pulp preparation, adsorption and drying. The experiment indicated that withered leaves of magnolia grandiflora was the best raw material for making recyclable tableware.

1. Introduction
At present, there are four types of disposable degradable tableware on the market: degradable polypropylene tableware, pulp moulded tableware, all-starch tableware and vegetable fiber tableware [1]. The raw material of degradable polypropylene tableware is still plastic. By adding degradable material such as and photosensitizer or degradable starch material, plastic can be degradable [2]. However, its degradability is constrain by many aspects: long degradation phase, non-completing degradation; pulp-molded tableware faces limited sources and pricey material with severe pollution during manufacture, which make it unpopular in China; Although all-starch tableware possess advantages such as completing degradation, high food security, its low thermal stability, high flexibility, difficulty in preservation and high price make it unfavorled in market[3]. Thus, plant fiber tableware is the direction of this study because of its recyclable, degradable, and reusable characteristics [4, 5]. However, if the raw materials come from crops, other substances like pesticide residues and animal waste are inevitable. In recent years, Tan Wei and others have also carried out an analysis and Research on papermaking process using corn straw, rice husk, Pineapple leaves and banana fiber as raw materials [6, 7, 8], which proves the universality of plant fiber materials. Recently, a German team degraded a plate made of leaves in 28 days [9], which inspired us to use dead leaves as a disposable dish material. In this study, fallen leaves around the school were used as the raw material and mechnochemical pulping method was used to carry out the single-factor experiment to compare disposable plates made from different falling
leaves under different reaction conditions \cite{10, 11}, were compared to find out ideal material and parameter for pulp preparation.

2. Material and Methods

2.1 Material
The experimental materials collected from dry leaf of yulan, sycamore, palmetto and bamboo in Sichuan agricultural university.

2.2 Instrument
Steaming and boiling container, induction cooker, multi-functional breaker, dryer (model: DHG-9070A, Shanghai yiheng scientific instrument co. LTD), electronic scales, 0.45mm stainless steel mesh, gauze, compressor (hydraulic multi-functional testing machine with microcomputer display, model WEW-600D, Jinan hengsi shanda instrument co. LTD)

2.3 Reagent
Concentrations were 6\%, 16\%, 26\% and 36\% sodium hydroxide solution, 30\% hydrogen peroxide solution, 15\% sodium sulphide solution, Anthraquinone reagent.

2.4 Method
Both mechanical and chemical pulping method were used in this experiment to ensure enough quantity of pulping while removing more lignin to ensure pulping quality \cite{12}.

Chemical pulping adopts caustic soda pulping in paper manufacturing \cite{13}. Leaves are streamed after drying (dry conditions of 75 °C, 30 min), then using sodium hydroxide to decompose mesophyll cell obtaining vein nest in leave. In this process, lignin is extracted from fibrous cell walls and intercellular layers as result of disconnection of chemical bond in cellular lignin molecule by adding defused OH- SH- ions in the cooking liquid to lignin in leaves lignin and expansion of cellular wall caused by cooking liquid. Leaving vein nest become fibrous after delignification process. HS- and S2- from dissolved sodium sulphide can accelerate delignification while protecting cellulose from strong alkali. During steaming and boiling, Small amount of cellulose and hem-cellulose are dissolved and lost due to Alkaline hydrolysis and peeling reaction caused by alkali. Therefore, the addition of anthraquinone \cite{14}, in cooking liquid, which equivalent to 0.1\% fibrous material weight can prevent harmful peeling reaction and facilitate delignification.

Mechanical pulping is to obtain single strain fiber from raw material that are chemical treated withered leaves. The withered leaves are broken by breaking machine to separate leave veins and remove mesophyll.breaking machine also create heat and friction to soften intracellular. By mechanical treatment, lignin in leave veins become single strain fiber. After dring (80 °C drying conditions, 80 min), the weight of fibers is quantity of pulp. The final quantity of pulp is showed by using mass fraction W (%) $w = \frac{m_{1}}{m_{2}} \times 100\%$

m1- quantity of dried leaf (75 °C, 30 min)  
m2- quantity of dried pulp (80 °C, 210 min) \cite{15}

2.5 Experimental Design
The experiment was carried out in three different trials. Trial one is the comparison between pulp yield rate of different leaves. Selected dry leaf of yulan, sycamore, palmetto and bamboo were put into forced hot air oven for 30 min. After drying, 100g of each dry leave were cleaned and put into boiling container with 2.5 L distilled water, 500ml 16\% sodium hydroxide solution, 250ml 15\% sodium sulphide solution and 0.1\% Anthraquinone reagent for 2 hours.
Trial two is to compare influences of different sodium hydroxide solution concentration on the leaf pulp yield rate.

The selected leaves of yulan were put into forced hot air oven for 30 minutes. The dried leaves were divided into four groups, each group had 100g cleaned dry leaves. Each group was put into individual boiling containers with 2.5L distilled water, 500ml sodium hydroxide solution, 250ml 15% sodium sulphide solution and 0.1% anthraquinone reagent for 2 hours. In each boiling container, the concentrations of sodium hydroxide solution were 6%, 16%, 26% and 36%.

Trial three is to compare influences of boiling time on pulp yield rate.

Selected leaves of yulan were put into force hot air oven for 30min. Dried leaves were divided into four groups, each group was 100g cleaned dry leaves. Every group was put into individual boiling containers with 2.5L distilled water, 500ml 16% sodium hydroxide solution, 250ml 15% sodium sulphide solution and 0.1% anthraquinone reagent. The boiling time of each group were 1h, 2h, 3h and 4h.

After boiling trial, pulp was cleaned by using distilled water, broken by breaker and screen by mesh to remove leave vein residue. After that, pulp was boiled and bleached with 1L water and 67ml 30% hydrogen peroxide solution for 30min, then pass through 0.45mm sieve and molded by mold in 80°C forced hot air oven for 4 hours to produce paper fiber. The final products produced by compressing paper fiber using multifunctional compressing machine.

2.6 Degradation experiments

The common disposable dishes on the market at present: plastic dishes, Degradable Polypropylene dishes, paper dishes, and withered leaf dishes made in this experiment were used as sample materials. Cut 40*40 mm square pieces on the flat part of the bottom of the plate as experimental materials, and put them into 80 mm soil to simulate natural degradation for degradation test. Degradation was recorded every two days.

3. Results

3.1 Influences of different leaves (withered leaves) on pulp yield rate

Table 1. comparison of pulp yield rate among different leaves

| Species of leaves | Pulp yield rate (%) |
|-------------------|---------------------|
| Phoenix Tree      | 13.30%              |
| Magnolia grandiflora | 15.60%            |
| Bamboo            | 28.60%              |
| Species of leaves | 37.30%              |

From experiment, pulp was successfully extracted from withered leaves of four different plants. Based on this figure, the pulp yield rate from high to low are palmetto, bamboo, yulan, sycamore. Although palmetto had highest pulp yield rate, its fiber was thick. Meanwhile, bamboo was hard to collect.

3.2 Influences of different sodium hydroxide concentration on pulp yield rate
Table 2. Influences of different sodium hydroxide concentration on pulp yield rate

Based on figure 3, when sodium hydroxide concentration rose, the pulp yield rate declined. Thus, sodium hydroxide concentration highly impacts pulp yield rate. When sodium hydroxide concentration fell into range from 6% to 16%, the yield rate had obvious decline and fiber in pulp was too thick. When sodium hydroxide concentration fell into range from 16% to 26%, the change of yield rate was moderate while diameter of fiber was favorable. When sodium hydroxide concentration fell into range from 26% to 36%, the declination of yield rate increased while diameter of fiber was thinner. To save reagent and reduce pollution, the concentration of sodium hydroxide should be 16% to 36%. Therefore, 16% sodium hydroxide was used in next trial.

3.3 Influences of different boiling time on pulp yield rate

Table 3. Influence of different boiling time on pulp yield rate

Based on figure 4, with prolonged boiling time, the pulp yield rate raised firstly and then declined. When boiling time was 1-2 hours, the pulp yield rate had a minor improvement. In this period, the diameter of fiber was thick initially, but with gradually lignin precipitation, the fiber became thinner. When boiling time was 2-3 hours, the diameter of fiber was ideal. When boiling time was 3-4 hours, due to long time reaction, lignin was largely precipitated, thus fiber was too thin and pulp yield rate dropped quickly. Therefore, the most ideal boiling time is 2 hours: both saving time and ensuring yield rate.
3.4 Analysis of Degradation Experiments
Degradation experiments showed that the material of the withered leaf plate degraded significantly on the 5 day, and almost completely (fusion with soil) on the 14 day. Because the whole plate is not taken for testing, the actual degradation time should be increased, but the degradation rate of the leaves made into disposable dishes is much faster than other materials on the market at present, and the withered leaves will not cause ecological damage.

![Figure 1](image1.jpg)
Figure 1. Analysis of Degradation Experiments
(From left to right are paper plate, dead leaf plate, Degradable Polypropylene plate and plastic plate)

![Figure 2](image2.jpg)
Figure 2. Degradation after 14 days (fusion with soil)

4. Discussion
Although, livistona chinesis leaves has highest pulp yield rate, the thickest fiber leads to fragile disposable tableware, thus it is impractical. Sterculiaceae and Magnolia leaves are easy to collect and
has ideal pulp yield rate, thus is best option for producing pulp for disposable tableware.

Sodium hydroxide concentration has obvious influences on pulp yield of Magnolia leaves. 36% Sodium hydroxide concentration has low yield rate with thick fiber diameter, while 6% Sodium hydroxide concentration has higher yield rate with thin fiber diameter. Therefore, 16% Sodium hydroxide concentration has both high yield rate and suitable fiber diameter.

To some degree, boiling time has influences on pulp yield of Magnolia leaves. When boiling time is 1 or 4 hours, the fiber quality is poor with low yield rate. When boiling time is 2 or 3 hours, the fiber quality is much better with higher pulp yield rate. To save time, 2 hour boiling time is recommended.

5. Conclusions
In this paper, dry leaf of yulan, sycamore, palmetto and bamboo were selected, and the crude fiber was obtained by mechanical chemical pulping and caustic soda pulping. After processes of breaking, compressing, drying etc., the disposable food container that is environment-friendly and easy to degrade was produced. Also, optimistic processing condition is studied in experiment. It was found that, in four kind of leaves, that yulan dry leaf was the best material for the type of disposable dish. Boiling under 100 ℃ with 16%NaOH, 15% sulfidation parameter, 0.1% anthraquinone for 2 h has the best pulp yield rate and fiber quality, which process enough tenacity to meet strength requirement. This study provides a fundamental basis for developing disposable tableware using plant-sourced fiber.

Acknowledgements
This research was financially supported by my instructor, Liu Ming-dan (Corresponding author), a respectable, responsible and resourceful scholar, who has provided me with valuable guidance in every stage of the writing of this paper. Thanks to my team, we find and solve all kinds of problems together, meet setbacks and never give up, and work unremittingly for common goals.

Corresponding author: Liu Ming-dan 1, male, (1965—), professor. Major in interests include theory teaching, application and control of motor, College of Mechanical and Electrical Sichuan Agricultural University Ya an Sichuan, 625014
E-mail:276591355j@qq.com, TEL:13086317002
The first author: Liu bo-fan 1 Major in agricultural mechanization, sichuan agricultural university
Layout fees are paid by the correspondent author Liu Ming-dan

Reference
[1] Cai Huijuan, Yan Nahan, Su Qinpo, Analysis of disposable tableware use in college students [J]. Journal of Henan Science and technology,2013(17):170.
[2] Zhou Logan, Discussion on the development of disposable tableware [J]. Technology development of interprise,2001(03):4-5.
[3] Chen Yanling, Not easy popular for environment-friendly tableware [J]. Rural agriculture and farmers,2018(05):41-43
[4] Liu Tianshu, Li Shujun, Zhou Shuhui, Disposable ware technology in China [J]. Farm Machinery,2011(12):9-11.
[5] Shi Chang. Disposable tableware with plant fiber is sold at home and abroad [J]. National business,2001(Z2):8
[6] Tan Wei, Changjiang, Bian Zhiqi, Zheng Qi, Liu Chang, Guo Yue. Analysis and Research of Corn Straw Papermaking Process [J]. Journal of Harbin Commercial University (Natural Science Edition), 2015, 31 (04): 458-459+464.
[7] Amit Ramdhonee, Pratima Jeetah. Production of wrapping paper from banana fibres[J] Journal of Environmental Chemical Engineering5 (2017) 4298–4306
[8] Stephen Sibaly, Pratima Jeet. Production of paper from pineapple leaves [J]Journal of Environmental Chemical Engineering, 5 (2017) 5978–59
[9] From 730000 to 28 days: Leaf trays shorten the degradation time of tableware [J]. China Social Organization, 2016 (20): 52-55.
[10] Jia Jingyu, Wu Ting, Han Xiaohui, Miao Li. Leaf reuse pattern of urban garden in new period [J]. Agriculture of Jilin[J].,2018(05):57.
[11] Chen Kefu, Zhang Hui. Green progress and engineering technology of China's paper industry [M].China light industry press 2016（01）.136-173.
[12] Huang Junyan, Tian Yuanyuan, Zhu Tingting, Guo Runlan, Packaging paper processing and inspection technology[M], Printing industry press, 2009（01）:24
[13] Li Xushengs. Studies and progress on anthraquinone pulping mechanism [J]. East China Pulp & Paper Industry 2016（04）:30-33
[14] ZhanHuaiyuany, LiuQiju, Jinfuming, Pulping technology[M]. China light industry press,2012(01:37-45)
[15] Wunan,Wulei, Pulp and Paper Mill Laboratory Test Methods[M]. China light industry press,2009(01:253-256)