# Development of Mycelial Leather Alternative and Sustainable Utilization of its Solid waste

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Development of Mycelial Leather Alternative and Sustainable

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Utilization of its Solid waste

Abstract

Mycelium has been considered as a promising biobased material for developing sustainable leather alternatives, driven by the increasing demand for environmentally friendly materials. In this study, we demonstrate that the mycelium can be successfully tanned with chromium-free tanning method to improve its structural stability, and significantly improved physical mechanical properties, including tensile strength and tear strength. The tanned mycelial leather alternative also exhibits excellent thermal stability exceeding 1500C. The tanning mechanism was investigated, and the environmental impact of the obtained mycelial leather alternative were evaluated. It showed a soil biodegradation rate of approximately 60 % in 60 days and does not exert and adverse effects on plant growth. In addition, the tanning process generates solid waste in the form of shavings. Owing to the inherent composition and microstructure, these wastes were utilized to produce porous carbon material. The synthesized mesoporous biochar has a high specific surface area of 3151.9 m2/g, indicating potential applications in energy storage and catalysis adsorption. This work demonstrates the feasibility of using mycelium to develop green, economical, and biodegradable leathe alternatives, while simultaneously providing a value-added way of process wastes.

Key words: Mycelial leather alternative; Strength improvement; Waste Utilization; eco-friendly material; Sustainable leather alternatives

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## 1. Introduction

In the recent years, there has been an increased interest in employing bio-based non-animal fibers, such as plant and microbial fibers to develop leather-like materials, due to the growing trend of reducing the use and toxicity of chemical agents in leather processing 1. Naturally grown mycelium, due to their reticulated structure, are considered as promising materials for producing desirable leather alternatives 2. Polysaccharides, including chitin, glucans, and chitosan are the main components of mycelium, along with a small amount of protein and polyelectrolyte acid 3. Chitin, found in the cell walls of mycelium, grows into hyphae via formation of elongated tubular structures. The N-acetylglucosamine units in chitin, due to their linear molecular structure, provides elasticity and strength to the hyphe due to strong nanofiber tensile strength 4. These linear polysaccharide hyphae entangle further to grow into a dense network structure and thereby forming the mycelium 5. Rather than relying on synthetic leather methods which incorporate plastic materials, tanning methods has the potential to stabilize the naturally largesized mycelium similar to those in traditional leather production. However, the strength of tanned mycelial leather alternative was still short in comparison to genuine leather. Hence, alternative tanning methods is required to provide an effective solution for improving their mechanical properties and broadening their practical application 6.

The disposal of mycelium waste as garbage in the development of emerging mycelium-based leather composites leads to a wastage of biomass resources 7. Therefore, efficiently utilizing biomass resources to

achieve high added value is crucial for addressing the energy resource crisis 8. The discarded Ganoderma lucidum mycelium (GLM) from experimental preparation of mycelium-based leather composites primarily consists of biopolymers (dextran, chitin, and proteins), with its microstructure characterized by linear mycelia including generative mycelia, skeletal mycelia, and connecting mycelia 9. Additionally, the multilayered pore structure formed by intertwined interactions between the mycelium and numerous tubular structures provides

favorable conditions for activator infiltration 10. This creates favorable conditions for producing porous carbon materials with a high specific surface area.

In this paper, the tanning mechanism of genipin on mycelial fibers was investigated to enhance the physicochemical properties of mycelium leather alternative, and subsequently crust leather was obtained via fatliquoring with glycerol. Tanning efficiency was evaluated by analyzing the changes in structural and functional groups in the mycelial fibers. Additionally, the application prospect and environmental impact of the genipin-tanned mycelial leather alternative were evaluated. This study aims to contribute to the development of sustainable leather-like materials and promote environmental friendly leather alternatives. Afterwards, high-performance porous carbon materials were synthesized through a two-step process that utilized mycelial wastes generated during the production of mycelial bio-based leather composites from ganoderma lucidum mycelium as raw materials.

2. Methodology

2.1 Preparation of mycelial leather substitutes

In the tanning process, the incompletely dried original mycelial sheets (ML) were tanned with glutaraldehyde solution. The reaction was carried out in a tanning drum (GZGS4 Xinda China), obtaining the tanned mycelium sheets (ML-T). Subsequently, the mycelial leather substitutes (ML-TG) were prepared by fatliquoring MLT with 3 g/L glycerol solution. Besides, The ML were treated with 3 g/L glycerol for 5 hours to produce of

mycelial leather after greasing (ML-G) to verify the effect of fatliquorling process. After tanning, unfixed glutaraldehyde was removed through water washing at 30° C for 5 min. Subsequently, the ML-TG and ML-G were naturally air-dried at 25° C.

2.2. Preparation of porous carbon material from Ganoderma lucidum residue

The waste mycelium of Ganoderma lucidum was dried in a blast drying oven for 12 h and then pulverized into a powder using a grinder. The powder was pre-carbonized in a tube furnace at a temperature of 400 °C with a heating rate of 5 °C/min, followed by 2 h insulation period. The carbonized material was washed 3-5 times with ethanol and deionized water, and further carbonized in a tube furnace at a temperature of 800 °C with a heating rate of 5 °C/min, followed by 2 h insulation period. The carbonized material was first washed with concentrated hydrochloric acid to remove metal elements, and then washed 3-5 times with anhydrous ethanol and deionized water. The resulting dried samples were named AGLM-1, AGLM-2, AGLM3, AGLM-4, AGLM-5, and AGLM-6 in the order of the mass ratios of KOH used.

Results and discussion

3.1 Morphology analysis

To enhance the physical and mechanical properties of original mycelial sheets (ML), glutaraldehyde tanning and glycerol fatliquoring were applied. The morphology of fiber distribution and fiber structure was observed in Fig.1. CT analysis shows that the untreated ML exhibits a network structure with irregularly distributed fibers. After tanning and fatliquoring, the fibers of mycelial leather alternative (ML-TG) become more ordered and less tangled, resulting in a smoother surface of the mycelium leather substitute (Fig.1(a)&(b)). As shown in Fig.1(c), after glutaraldehyde tanning, the fiber surface becomes rougher and the gaps between fibers decrease, leading to a more tightly woven structure. Furthermore, the addition of glycerol alone leads to an uneven aggregation between glycerol and fibers, resulting in a clumping state, while the surface of individual

ML-TG, with tightly interwoven fibers and a smooth surface. It indicates that glutaraldehyde tanning and glycerol fatliquoring have a synergistic effect .

fibers remains smooth. However, when fatliquoring is performed with glycerol after glutaraldehyde tanning, a more orderly and compact structure can be obtained in

Fig. 1. CT analysis of (a) ML and (b) ML-TG, (c) SEM analysis

3.2. Application properties evaluation

The physicochemical and sensory properties were measured in Fig.2 to determine the suitability of mycelial leather substitute for practical application. ML exhibited a tensile strength of 1.8 MPa and an elongation at break of 2.7% (Fig. 2(a)), which are insufficient for practical use due to their low strength and ductility. However, through the synergistic effect of glutaraldehyde tanning and glycerol fatliquoring, which involve the crosslinking of glutaraldehyde with mycelial fibers and hydrogen bond formation between glycerol molecules and hydroxyl groups11, the mechanical properties of ML-TG were significantly improved as shown in Fig.2(c) and 2(d). ML-TG exhibited a tensile strength of 9.1 MPa and an elongation at break of 42.1%, which are comparable to sheep nappa leather with a tensile strength of 10.9 MPa and an elongation at break of 57.9%12. In addition, the

improvement in softness (Fig.2(b)) is due to the penetration of glycerol, a hygroscopic polyol, which enhances flexibility13. The excellent thermal stability of natural mycelium results in shrinkage temperatures exceeding 100°C for all samples, signifi-

cantly higher than that of aldehyde-tanning leather, which has a shrinkage temperature of approximately 80°C14, To further evaluate the potential application ability, a wallet was made from ML-TG, and its physicochemical properties were tested (Fig.2(c)). The air permeability reached 463 ml/cm2.h and it was free of formaldehyde. These results suggest that ML-TG has the potential to be used in the production of products.

3.3. Environmental impact assessment

3.3.1 Biodegradability of ML-TG.

As shown in Fig. 3a-b, microbial enzymatic processes facilitate biodegradation of ML-TG in soil. Various extracellular hydrolases, such as chitinases and βglucanases secreted by soil microorganisms, including bacteria and fungi depolymerize the macromolecular components of the mycelial matrix into smaller, soluble compounds. Surrounding microbial communities then assimilate these low-molecular-weight molecules and metabolize through intracellular pathways which ultimately lead to their mineralization into inorganic substances such

indicates good environmental compatibility of ML-TG.

as CO, and H, O and subsequent weight loss15. Within the first 30 days, the weight reduction was very less as the microorganisms need time to attach to the surface of ML-TG and move into the stationary phase. After 40 days, there was a sharp increase in the weight loss of ML-TG from 19.77% to 51.55% (Fig. 3b). By 60th day, plant roots

Fig. 2. (a) Tensile strength and elongation at break of ML, ML-T, ML-G, (b) Softness and shrinkage temperature

of ML, ML-T, ML-G, (c) Physicochemical properties of ML-TG, (d) Stress-strain curves.

3.3.2 Evaluation of exhaust gas from incineration

Incineration is a common and effective method for the treatment of solid waste leading to the volume reduction of incinerated materials by 80%-90%. To evaluate the incineration characteristics of mycelial leather substitute, TG-FTIR was used to analyze the decomposition of ML, ML-T, ML-TG, and sheep leather which identified the presence of different functional groups as shown in Fig. 4. It is clear from the figures that the peak positions of mycelial samples including ML, ML-T, and ML-TG are similar but their peak intensities vary. NH3 was identified as the primary exhaust gas generated from the burning of

were observed to grow into the ML-TG and the weight

loss of mycelial leather alternative increased to 58%.

During the 60-day period, no adverse effects were shown

by ML-TG on the growth of radish plant (Fig. 3c). This

mycelial samples . During the incineration of ML, various gases, including CO2, NO2, SO2 and NH3 are released. However, no SO2 emissions are detected in the exhaust gas of ML-T and ML-TG due to the disappearance of elemental sulphur (Fig. 4b) and emission of CO2 also decreased gradually. This reduction should be attributed to the enhanced stability of C-containing and S-containing bonds in mycelial fibers, resulting from crosslinking and hydrogen bond formation during tanning16. In addition, due to the crosslinking of free amine groups with the tanning agent, NH3 emission was observed to reduce during the pyrolysis phase. Moreover, gases such as SO2 and CO are generated during the incineration of sheep leather.

Fig. 3. (a) Biodegradation of ML-TG; (b) Weight changes in biodegradation; (c) Effect of waste ML-TG on plant

growth (PT: Growth in natural soil. PG: Growrh in soil with ML-TG scraps).

Therefore, fewer types of gaseous waste are generated during the incineration of ML-TG and thus indicating a potential environmental advantage.

3.4 Analysis of specific surface area and pore size distribution

Based on the IUPAC classification, AGLM-1, AGLM-2, AGLM-3, AGLM-4, AGLM-5, and AGLM-6 demonstrated type I adsorption isotherms (Fig. 3(A)) 17. AGLM-1 and AGLM-2 exhibited a rapid increase in adsorption at low relative pressures ($P/P0 < 0.1$), indicating the presence of a rich microporous structure. AGLM-5

Fig. 4 TG-FTIR analyses of mycelial leather substitute and sheep leather

had a high specific surface area of 3151.9 m2/g, primarily in the form of a mesoporous structure with a small amount of micropores. The presence of micropores allowed for effective electrolyte retention for energy storage. However, excessive KOH activation leads to a decrease in SSA for sample AGLM-6 to 2727.0 m2/g due to conversion of some micropores into mesopores or graphite during high temperature pyrolysis; this process also causes pore collapse resulting in reduced total pore volume18. Therefore, employing high temperature activation with a GLK:KOH mass ratio of 1:5 enables the porous carbon material to exhibit enhanced surface area and improved pore structure.

Fig. 5: (A) N2 adsorption-desorption isotherms and (B) poresize distribution for AGLM-1, AGLM-2, AGLM-3,

AGLM-4, AGLM-5, AGLM-6.

4. Conclusion

In this study, a green and sustainable mycelial leather substitute was successfully developed through glutaraldehyde tanning, and its tanning mechanism was systematically. The obtained ML-TG showed a tensile strength of 9.1 MPa and an elongation at break of 42.1%, representing a substantial enhancement compared with untreated mycelial leather. ML-TG also demonstrated excellent thermal stability, with a shrinkage temperature above 1000 C, and high air permeability of 463 ml/cm2·h, attributed to its ordered fiber structure. Soil burial tests confirmed good biodegradability and environmental compatibility, and incineration produced fewer and less toxic gaseous emissions than sheep leather. Furthermore, waste generated during the production was valorized into a porous biochar with an ultrahigh specific surface area of 3151.9 m2/g, featuring a mesoporous-dominated structure that facilitates ion transport. Overall, this work provides a viable pathway for designing economical, biodegradable, and environmentally friendly bio-leather substitutes based on mycelium.

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