2025 IULTCS Young Leather Scientist Grant

Identification: YLSG_2025_Yudan Yi

COMPLETE APPLICATION FORM (click application area)

☐ Basic Research ☐ Machinery / Testing ☑ Environmental/Sustainability

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By submitting this application, I commit to develop the project as outlined in the attached Research Project Plan and to complete a written report by 28 February 2026, with the following items:

1) Introduction
2) Materials and Methods
3) Results and Discussion
4) Conclusion
5) Suggestion for Future Work
6) References

2) **Research Project Plan outline – Maximum 3 pages**

Title: A cationic amphiphilic acrylic copolymer for metal-free eco-leather production: Integration of retanning and fatliquoring

Introduction:

Chrome tanning system, the dominant leather-making technology in the leather industry, produces leather with excellent comprehensive performance. However, a large amount of Cr(III)-containing wastewater and solid wastes are generated during leather processing, which greatly restricts the sustainable development of the leather industry (Gao et al., 2023). Therefore, considerable efforts have been devoted to the development of chrome-free tanning systems to eliminate Cr discharge from the origin (Shen et al., 2023; Zhu et al., 2022). Non-chrome metal tanning and organic tanning technologies are the two major directions, and organic tanning system exhibits vast future development potential because it can produce metal-free leather with favorable biodegradability (Inbasekar and Fathima, 2023). Nevertheless, the metal-free leather still exhibits some disadvantages compared to the traditional Cr-tanned leather from the production practices. First, the strong water absorption of metal-free tanned leather will lead to the adhesion of collagen fiber and deep penetration of the base coating agents into the leather, thereby resulting in low durability and stiff handle of the finished leather (Ngwabebhoh et al., 2023; Xu et al., 2022). Moreover, the low isoelectric point (pI) of organic tanned leather (wet white) cannot match with the charge properties of traditional anionic post-tanning chemicals. The uptake rate of anionic post-tanning chemicals by the metal-free tanned leather is low, leading to the poor performance of the finished leather and the large pollution load in post-tanning wastewater.

To address the above two issues, it is highly desired to design a multifunctional post-tanning chemical, such as a polymer retanning agent or fatliquor with proper hydrophobicity and charge regulation ability. The enhancement of hydrophobicity of metal-free leather may be addressed by the use of amphiphilic copolymers. This approach has been demonstrated to be effective in Cr tanned leather since the fiber structure can be coated with the copolymers containing hydrophobic alkyl groups (Baquero et al., 2021; Du et al., 2016). As for the second issue, amphoteric polymers can impart some electropositivity to leather to a certain extent, thereby favoring the penetration and distribution of the other post-tanning chemicals (Li et al., 2022; Wang et al., 2021). Xu et al. synthesized an amphoteric polyurethane retanning agent, which endowed vegetable-tanned leather with high thermal stability and enhanced the reactivity of leather to anionic materials (Xu et al., 2019). Some novel amphoteric acrylic polymer retanning agents reported recently showed satisfying assistant-dyeing effect on leather (Ma et al., 2021; Tian et al., 2023). Actually, the electropositivity elevation of the metal-free leather by amphoteric polymers is still inadequate. Attention should be paid to the cationic polymers which are seldom reported and used. Cationic fixing agents have been widely used in dyeing, whereas they have no effects on leather performance other than color fixation. Thus, an effort on the metal-free leather is expected to be useful through the employment of cationic amphiphilic copolymers. Moreover, the cationic amphiphilic copolymers were expected to possess dual functions of retanning and fatliquoring, which is conducive to saving leather chemicals, reducing the discharge of leather wastewater, and shortening the processing time.

Objectives:

The general objective is to develop a cationic amphiphilic polymer with proper hydrophobicity and charge regulation ability, which can be utilized in post-tanning process to produce eco-friendly metal-free leather with high performance. The specific objectives include:

- Preparing a cationic amphiphilic copolymer with retanning and fatliquoring dual function, and investigating its aggregation behavior.
- Appling the prepared copolymer in the retanning and fatliquoring processes of wet white, and assessing the environmental impact of the processes.
- Elucidating the interaction mechanism between the copolymer and wet white.

Methods:

The research content includes three aspects (Fig. 1):

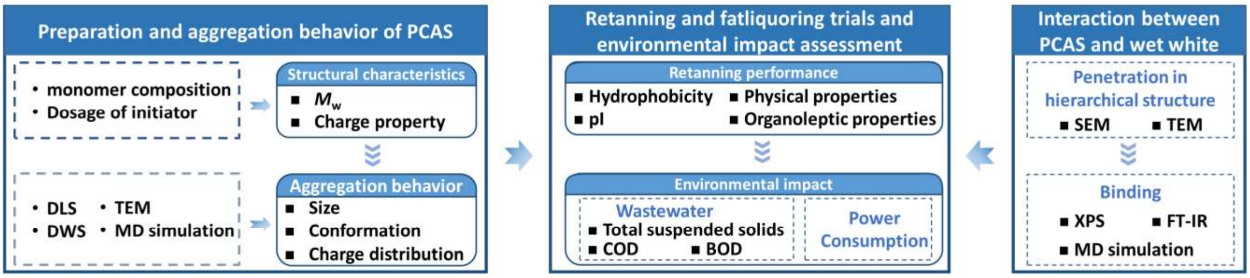

Fig. 1. Research plan

- Synthesis of PCAS

SMA (0.1 mol) and DMA (0.08 mol) were mixed with PVS (0.1-0.4 mol) to obtain the blend of the two monomers. Initiator AVBN (1-5 wt%, based on the total weight of monomers, the same below) was dissolved in 15 wt% of ethanol to prepare the initiator solution. Then the blend of monomers and the initiator solution were simultaneously added dropwise to a four-necked flask containing 10 wt% of ethanol under stirring at 90 °C. The reaction was incubated for 5 h after the dropwise addition. Then the product was protonated by acetic acid. Subsequently, deionized water at 70 °C was added to the system and stirred at high speed to adjust the solid content of the product to 30%. Finally, the cationic amphiphilic copolymers, named PCAS, were obtained (Fig. 2). The chemical structure of the PCAS was further characterized by FT-IR, ^{1}H NMR, and ^{23}Si NMR spectra. The aggregation behavior of PCAS was investigated by dynamic light scattering (DLS), diffusing wave spectroscopy (DWS), transmission electron microscope (TEM), and molecular dynamics (MD) simulation.

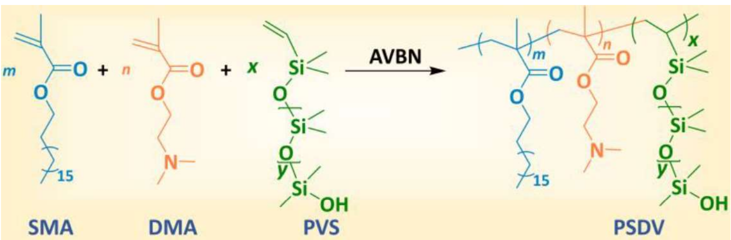

Fig. 2. Schematic of PCAS synthesis (y=1, 5, 10)

- Retanning and fatliquoring trials and environmental impact assessment

PCAS were applied in the retanning and fatliquoring processes of shaved wet white. Commercial anionic acrylic resins and fatliquoris were used for comparison. The uptake rate, hydrophobicity, pI, thickness increase rate, fiber dispersion, dyeing performance, and physical properties of the crust leathers were measured. The COD, BOD, and total suspended solids of the final bath were determined. The consumption of water, power, and time of processes were calculated.

- Interaction between PCAS and wet white

SEM and TEM were used to observe the state of PCAS aggregates in hierarchical structures of leather including fibril, fiber, and fiber bundle. The structural characteristics of PCAS processed leather were analyzed using FT-IR and XPS. Moreover, the MD simulation was also used to investigate the interaction between PCAS and wet white according to our previous work (Yi et al., 2024). A bimolecular collagen model (COL) was modified according to the structural feature of organic tanning agents used here to build model of wet white. The PCAS model was built following the method in our previous work. The hydrogen bonding, electrostatic interaction, Van der Waals interaction between PCAS and wet white can be calculated through MD simulation to illustrate the interaction mechanism.

Hypothesis/Expected Results:

A cationic amphiphilic copolymer (PCAS) with retanning and fatliquoring dual function will be successfully synthesized. PCAS will endow metal-free leather with hydrophobicity similar to chrome-tanned leather. Moreover, PCAS will lift the pI of metal-free leather, thus promoting the post-tanning materials absorption to diminish water contamination. The comprehensive properties of the metal-free leather treated by PCAS, can be improved compared to those processed by the commercial retanning agents and fatliquors. our results will give new insight for the design of post-tanning materials which can match with metal-free leather.

Research benefit for the local or global leather industry (one sentence only):

This project will hopefully provide tanneries with a practical approach to produce eco-friendly metal-free leather, which can save leather chemicals, reduce the discharge of leather wastewater, and shorten the processing time, thereby promoting the sustainability of leather industry, both for China and the world.

Literature:

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