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Development of Biopolymer-Based Re-tanning Agents for

Sustainable Leather Processing

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Abstract

The leather industry faces significant environmental challenges arising from the widespread use of petroleum-derived synthetic retanning agents, which increase pollution loads and introduce free formaldehyde into finished leather. This study addresses these concerns by developing a sustainable retanning approach based on biopolymer-based systems as environmentally benign alternatives to conventional syntans. Sourced from renewable resources, these systems are engineered to minimize ecological impact while ensuring efficient leather processing. The outcomes demonstrate the capability of biopolymer retanning agents to substitute petroleumbased materials, supporting the transition toward greener chemistries in leather manufacturing. This work strengthens ongoing efforts to incorporate renewable materials into industrial practices, aligning leather processing with global sustainability and environmental compliance objectives.

Keywords: biopolymers; sustainable process; ecofriendly; re-tanning agent.

1. Introduction

The conversion of raw skin into leather provides permanent resistance to biological degradation and imparts functional properties such as strength, flexibility, and

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permeability. In post-tanning, leather characteristics including thickness, handle, grain compactness, and fullness are adjusted through the incorporation of retanning agents into the fibre network (1). These agents occupy the interfibrillar spaces and regulate fibre mobility, which directly influences softness and body. Conventional retanning systems, including phenolic and melamine-based resins, are widely used to enhance fullness and grain properties; however, they are largely petroleum-derived and frequently involve aldehyde chemistry, raising concerns regarding sustainability, restricted substances, and environmental burden (1, 2)

Driven by environmental regulations and sustainability goals, increasing attention is being given to bio-based retanning materials (3). Various natural polymers have been explored as fillers; nevertheless, their practical performance is strongly governed by physicochemical features such as particle size and surface charge, which control penetration, distribution, and retention within the skin matrix (4-6). Accordingly, the present study investigates the feasibility of carrageenan as an unmodified biopolymer retanning agent for interfibrillar filling, with the aim of replacing conventional inorganic and organic fillers.

2. Methodology

Chromium (III)-tanned goat skins (wet-blue goat leather) were used as the substrate for this study and were sourced from a commercial tannery. Carrageenan (C) was

procured from commercial market. Commercial-grade chemicals were used for leather processing. Goat wetblue leathers shaved to 0.9–1.0 mm were used for posttanning studies. The carrageenan retanning system was applied at two concentration levels (3, and 6 wt.%) to identify the optimum offer. The retanning solution was prepared by dispersing carrageenan in water (1:50) at 35°C under stirring. Before retanning, the leathers were wetted back and neutralized to facilitate penetration of the biopolymer. Retanning was carried out in a drum using the prepared carrageenan system, followed by fatliquoring and acid fixation. A commercial

melamine-based syntan (5 wt.%) was processed in parallel as a control. After chemical processing, the leathers were mechanically finished by setting, drying, and staking.

3. Results and Discussion

Characterization of Carrageenan

The particle size range and surface charge (zeta potential) of the carrageenan dispersions were measured by dynamic light scattering (DLS) using a Litesizer 500 (Anton Paar) at 25 °C, with samples prepared in aqueous medium. The dynamic light scattering results indicate that the carrageenan dispersion exhibits a particle size distribution predominantly in the nano- to submicron range, with a major population centered in the few hundred nanometer region and a broader distribution extending toward the micron scale. This particle size range is favorable for leather retanning applications, as it supports effective penetration into the micro- to mesoporous structure of wet-blue leather and promotes uniform distribution within the collagen fibre network. The presence of a broader distribution suggests the availability of both smaller particles for diffusion into finer interfibrillar spaces and relatively larger entities for effective void filling.

The zeta potential profile shows a distinct peak at approximately –17.5 mV, confirming the anionic nature of the carrageenan system. This negative surface charge contributes to colloidal stability of the dispersion and

facilitates electrostatic interaction with positively charged sites on chromium-tanned collagen. Such interactions are essential for controlled fixation of the retanning agent within the leather matrix, reducing the risk of wash-out and enabling efficient interfibrillar filling.

(a)

(b)

Fig.1: (a) DLS Result, (b) Zeta potential of Carrageenan

Fourier transform infrared spectrum (FTIR) was collected

using a JASCO-4700 model spectrometer (JASCO Co., Tokyo Japan) was used to characterize their chemical structures in the spectral range of 500 to 4000 cm-1 . The FTIR spectrum of carrageenan exhibits a broad absorption band around 3295 cm-1, corresponding to O–H stretching vibrations, indicating the abundance of hydroxyl groups and strong hydrogen-bonding capability. This feature is

significant for leather retanning, as hydroxyl groups can form hydrogen bonds with the functional groups of

Fig. 2 FTIR Spectrum of carrageenan

The band observed near 1640 cm^{-1} is attributed to bound water and associated stretching vibrations, reflecting the hydrophilic nature of carrageenan, which supports effective dispersion in the processing float and facilitates penetration into the collagen matrix.

Characteristic sulfate-related bands appear prominently at ~1032 cm^{-1} (O=S=O stretching) and in the region of 1200–1250 cm^{-1} (S=O stretching), confirming the presence of sulfated galactan units. These sulfate groups impart a strong anionic character to carrageenan, enabling electrostatic interactions with positively charged sites on chromium-tanned collagen, thereby enhancing fixation and uniform distribution within the leather structure.

The peaks around 1379 cm^{-1} and 1234 cm^{-1} are associated with C–H bending and C–O stretching vibrations, while the band at ~928 cm^{-1} corresponds to C–O–C and glycosidic linkages, confirming the polysaccharide backbone of carrageenan.

Overall, the presence of abundant hydroxyl and sulfate functional groups provides multiple interaction sites for hydrogen bonding and ionic interactions with collagen fibers. These interactions support carrageenan’s effectiveness as an eco-friendly retanning agent by facilitating interfibrillar space filling, improving fiber lubrication compatibility, and contributing to improved softness and fullness of the retanned leather. Fig. 3: (a) Load at grain crack, (b) Softness

collagens, promoting intermolecular interactions and contributing to interfibrillar filling.

(a)

(b)

The grain crack load results indicate that carrageenanretanned leathers exhibited good grain strength. At 3% offer, the carrageenan-treated sample showed a grain crack load of 26.36 kg, reflecting effective interfibrillar filling and reinforcement of the collagen matrix. However, when the offer level was increased to 6%, the grain crack load decreased to 22.53 kg, suggesting that excess biopolymer may lead to overfilling of interfibrillar

spaces, which can restrict fiber mobility and reduce resistance to grain cracking. Notably, the value at 6% was still comparable to that of the commercial syntan control (22.40 kg).

A similar trend was observed in softness. The carrageenan-treated leather exhibited higher softness at 3% (2.27), while a slight reduction was recorded at 6% (2.16). This decrease in softness with increasing offer indicates that higher carrageenan concentration may increase fiber packing density, thereby lowering flexibility. Nevertheless, the softness at 6% remained comparable to the control (2.00).

Overall, the results demonstrate that lower carrageenan offer (3%) is more favorable, providing improved grain strength and softness, while higher offers lead to marginal decline in properties, highlighting the importance of optimizing retanning concentration.

4. Conclusion

This study demonstrates the feasibility of carrageenan as a sustainable biopolymer- based retanning agent for leather processing. The nano- to submicron particle size distribution supports effective penetration into the collagen matrix, while the negative zeta potential confirms the anionic

nature of the system and its suitability for interaction with chromium- tanned collagen. FTIR analysis revealed abundant hydroxyl and sulfate functional groups, providing multiple interaction sites for hydrogen bonding and ionic interactions and enabling efficient interfibrillar filling. Performance evaluation showed that a 3 wt.% carrageenan offer provided improved grain strength and softness, whereas a higher offer (6 wt.%) resulted in a slight decline in properties, indicating the importance of concentration optimization. The carrageenan-retanned leathers exhibited performance comparable to a commercial melamine- based syntan, highlighting carrageenan as a promising eco-friendly alternative to conventional petroleum-derived retanning agents.

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