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Influence of Retanning Agents on Physical Characteristics of Snow-White Glove Leather from Goat Skin

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ABSTRACT

The quality of leather is subject to various elements, such as the specific raw material used, and the processing techniques implemented throughout the production process. Goatskin, renowned for its exceptional softness and durability, is frequently employed in the production of gloves owing to its desired attributes, notably its capacity to produce snow-white gloves. Retanning is an essential step in the leather manufacturing process as it significantly improves the characteristics of the leather. Through an investigation into the impacts of distinct retanning agents on goatskin leather gloves, this research endeavor seeks to offer significant knowledge regarding the most effective retanning procedures that can augment the tactile attributes of snow-white leather gloves. In this study, formaldehyde, alum, chromium, and chromium alum were employed as retanning agents. The retanned leathers were assessed by physical parameters, Principal Component Analysis (PCA), and Fourier Transform Infrared spectroscopy (FTIR), then compared to a commercially available snow-white leather glove. The physical characteristics of chrome alum retanned leather showed a high degree of softness (6.60±0.02 mm), good tear strength (1.530±117 N/cm), and tensile strength (1.500±100 N/cm^2). The principal component analysis (PCA) also supported that the properties of chrome alum retanned snow-white glove leather closely resemble commercial leather and the result from this study suggested that all factors notably impact the skin's physical quality, except for thickness. Additionally, the FTIR analysis reveals similar functional groups from the different retanned leathers indicated by comparable peaks and stretching patterns. Therefore, chrome-alum could be a well alternative retanning agent for the production of snow-white glove leather.

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Introduction

Leather is a versatile material and is widely used in various industries, including fashion, sport, automotive, and upholstery (Gatto and Parziale, 2024). The tanning industry is an ancient industry for making raw putrescible hide and skins into imputrescible, usable leather to provide notable protection of human organs especially, the feet and hands. The processing of skins to make it resistant to microbial attack covers three main stages such as beam house operation, tanning, and post-tanning (Das *et al.,* 2022). Tanning is a prehistoric protocol of leather processing where a variety of chemicals are being used. Tanning or retanning materials are selected according to the end product. In addition, the quality of leather is influenced by several factors, including the type of raw materials, the processing chemicals, and the methods employed during production (Das *et al.,* 2022; Kurnianto *et al.,* 2024).

In the leather manufacturing process, retanning plays a crucial role in enhancing the properties of the leather. The physical properties of the finished product are strongly influenced by the application of retanning agents, which are chemical substances used during this stage. Retanning agents, one of the most important post-tanning materials which are chemicals used in leather processing significantly influence the physical characteristics of the final product. These agents can alter the leather's strength, flexibility, color, and other important attributes, ultimately determining its suitability for specific applications (Guan *et al.,* 2023).

The type of leather to be produced finally is also dependent on the raw material source. Goatskin, known for its softness and durability, is commonly utilized in glove manufacturing due to its desirable characteristics, such as snow-white glove (Hossain *et al.,* 2021). Snow white leather gloves are snow white color gloves made from sheep or goat skin, that is usually used for sport glove, such as golf. Generally, snow white leather gloves are tanned with chrome or formaldehyde, then retanned with various tanning agents that can result in special characteristics, such as a high level of softness. Tanning of animal skins with chromium, formaldehyde, or other aldehydes provides the desired softness, however, high amounts of aldehydes and metals like chromium (Cr) might lead to free formaldehyde, and Cr pollution in the environment which can cause harm to human health (Mizan *et al.,* 2023).

On the other hand, the European Union has restricted the use of some hazardous chemicals in leather processing. This embargo pushed the researcher to search for alternative chemicals to achieve the physical properties of leather obtained from these banned chemicals.

Despite the embargo by the EU and the importance of retanning agents, there is a need for comprehensive analysis to understand their precise influence on the physical properties of goatskin intended for glove leather. Such analysis is essential for optimizing the retanning process, ensuring the production of high-quality leather with consistent characteristics (Ding *et al.,* 2022). Therefore, this study aims to analyze the influence of retanning agents on the physical characteristics of goatskin, specifically for leather glove production. By systematically examining the effects of different retanning agents on properties that can be gained into the optimal selection and utilization of retanning agents in leather glove manufacturing.

The selection of formaldehyde, chrome, alum, and chrome alum as the retanning agents in this study is based on their widespread use in the leather industry and their unique properties in modifying the physical characteristics of leather. Formaldehyde, a commonly used retanning agent, is known for its ability to improve the tensile strength and durability of leather (Sun *et al.,* 2018). Chrome retanning agents are favored for their superior softness and color-fast properties, making them ideal for high-quality leather products. Alum, or aluminum sulfate, is valued for its ability to enhance the dyeing and color retention properties of leather, while chrome alum combines the benefits of chrome and alum, offering a balanced combination of softness and color retention (Sancakli *et al.,* 2023). By exploring the effects of these specific retanning agents on leather glove made from goatskin, this study aims to provide valuable insights into the optimal retanning processes for enhancing the physical quality of leather gloves. Through this analysis, manufacturers can make informed decisions regarding the choice of retanning agents to achieve desired leather quality and performance characteristics, thereby enhancing the competitiveness of their products in the global market.

Materials and Methods

Materials

The primary material for this study comprised of wet blue goat leather sourced from Yogyakarta, Indonesia, with an area of 6 sqft, and one piece of commercial retanned leather from Yogyakarta as control. One pieces of wet blue leather was cut into four different parts and used for retanning with 4 different retanning materials. The commercial snow-white leather glove that was derived from goatskin wet blue retanned using chrome and chrome-alum. The retanning agents utilized in the research included formaldehyde, chrome, aluminum, and chrome alum. These chemicals were collected from the local market of Yogyakarta, Indonesia and were commercial grade. In addition, analytical grade phenolphthalein indicators, and bromocresol green indicators were used in this study.

Methods

Leather treatment. The retanning process involved the use of formaldehyde, alum, chromium, and chromium alum as retanning agents. Table 1 provides detailed formulations and stages of the retanning process. These retanning agent variations were performed in the process other than changing other chemicals. The retanned leather was evaluated by physical properties, Principal Component Analysis (PCA), and Fourier Transform Infrared Spectroscopy (FTIR), then compared with commercial snow-white leather gloves as a control.

Physical properties evaluation. The physical properties of the retanned leather were determined following the methods of international standard (ISO) by the International Union of Leather Technologists and Chemist Societies (IULTCS) (ISO, 2012), such as tensile strength (ISO 3376), tear strength (ISO 3377-2), stitch tear strength (ISO 23910), softness (ISO 17235), elongation (ISO 3376) and abrasion fastness (ISO 17076). For each of the analyses, three samples were taken into consideration and average results have been reported in this study.

Fourier Transmitted Infrared (FTIR). FTIR analysis of the retanned leather was carried out by Perkin Elmer FTIR (Chicago, USA) fitted with universal Attenuated Total Reflectance (ATR) diamond/ZnSe crystal at room temperature. The FTIR wavelength region was 4000 to 400 cm^{-1} with a resolution of 4 cm^{-1} and each of the sample was scanned for 16 times.

Data analysis. The collected data were subjected to analysis of variance in each parameter utilizing IBM SPSS Statistics 25 software. The correlations among the variables were examined through Principal Component Analysis (PCA), implemented using Minitab 21 Statistical Software. In the PCA analysis of this study, PC1 to PC8 delineates the most variations to most eighth variations consequently. There are total eight variables in terms of physical characteristics of the tanned snow white glove leather that have been considered to be the main factors for choosing a suitable retanning agent in this study.

Results and Discussion

Physical properties of the retanned snow white glove leather

Physical properties such as softness, thickness, tear strength, tensile strength, stitch tear strength, abrasion fastness, shrinkage temperature, and elongation are regarded as the key parameters to confirm the retanning performance of snow-white glove leather. Table 2 delineates the important physical properties achieved through applying different retanning agents during retanning of goat leather and a comparison was made with a control sample. It revealed that the formaldehyde retanned leather showed maximum % of elongation, higher tear strength (3263 N/cm), and tensile strength (3267 N/cm^2) compared to alum, chrome alum and control leather sample whereas minimal degree of softness (5.23 mm), abrasion fastness (3.33±0.57), and stitch tear strength (6.90±0.40 N/cm) were noticed.

This increase in tensile strength and tear strength might be attributed due to the cross-linking reaction between formaldehyde and amino acid groups of collagen present in leather, resulting in a stronger and denser fiber structure. In addition, the shrinkage temperature of the snow white glove leather retanned by formaldehyde was noticed to be 77°C, which can be considered as an indicator of the leather's thermal stability (Table 2). Other research finding is in congruence with this result as the shrinkage temperature for aldehyde-tanned leather was found around 70-80°C (Maina *et al.,* 2019).The primary indicator of the extent of cross-linking is shrinkage temperature, which means, leather will be more resistant to mechanical, biological, chemical, and physical aggressions (Plavan *et al.,* 2013).

The physical properties of the snow-white glove leather retanned with alum showed moderate degree of softness of 6.16±0.28 mm, tear strength of 2226.67±205 N/cm, tensile strength of 2300.00±100 N/cm^2, sewing strength of 9.73±0.06 N/cm, abrasion fastness of 4.67±0.57 and high elongation of 73.92±00.0 % compared to other retanned leather (Table 2). The application of alum as a retanning agent in the post-tanning process leads to enhanced physical properties of the leather, including tensile strength, elongation at break, tear strength, shrinkage temperature, and softness observed by other researchers earlier (Haroun *et al.,* 2008). In this study, shrinkage temperature of the alum tanned leather was found 76.33±1.52°C. Therefore, alum as a retanning agent could enhance the shrinkage temperature due to an increase in the number of cross-links between molecules generated by electrostatic and coordinate covalent interactions between the aluminium complex and skin collagen (Haroun *et al.,* 2008). Furthermore, utilizing a combination of alum in the retanning process and vegetable tannin in the pre-tanning process results in greater physical strength causing an increase in the shrinkage temperature value up to 125°C (Haroun *et al.,* 2008). The hydrothermal stability of the

Table 1. Retanning process details for the production of snow-white glove leather

Process	Chemicals	Weight (%)	Duration (min)
Wetting back	Water	100	30
	Surfactant	0.5	
Water drain			
Retanning	Water	150	20
	Retanning agents	2	
	HCOOH	0.5	20
Neutralizing	Water	100	20
	HCOONa	2	
	$NaHCO_3$	1.5	3 ×15 + 120
Fatliquoring	Water	10	90
	Synthetic oil	7	
	Sulpho chlorinated fish oil	5	
	Semi synthetic oil	2	
	Surfactant	1	
Water drain			
Top fatliquoring	Water	100	
	Synthetic oil	1	30
	Sulpho chlorinated fish oil	1	
	Semi synthetic oil	1	
	HCOOH	0.4	2x15
	DERMAFIK TPX	0.75	15

Table 2. Physical properties of goat snow-white glove using various retanning agents

Retanning agents	Softness (mm)	Tear strength (N/cm)	Tensile strength (N/cm^2)	Stitch tear strength (N/cm)	Abrasion fastness^{ns}	Shrinkage temperature (^{0}C)	Elongation (%)	Thickness^{ns} (mm)
Formaldehyde	5.23±0.46^{a}	3263.33±50^{d}	3266.67±57^{d}	6.90±0.40^{b}	3.33±0.57	77.00±2.00^{a}	80.15±1.15^{e}	0.60±0.02
Alum	6.16±0.28^{b}	2226.67±205^{c}	2300.00±100^{a}	9.73±0.06^{e}	4.67±0.57	76.33±1.52^{a}	73.92±0.00^{c}	0.55±0.10
Chrome	6.40±0.35^{b}	3600.00±200^{e}	3340.00±243^{d}	17.14±0.05^{c}	4.67±0.57	81.33±1.52^{b}	76.331.53^{d}	0.45±0.10
Chrome alum	6.60±0.02^{b}	1530.00±117^{b}	1500.00±100^{b}	12.40±0.20^{d}	4.33±0.57	77.67±2.08^{a}	47.50±0.57^{a}	0.60±0.02
Control	6.60±0.02^{b}	560.00±40^{a}	570±70^{a}	4.12±0.03^{a}	5.00±0.57	83.00±2.00^{b}	50.40±0.57^{b}	0.50±0.01

Different letters on the same row indicates statistical difference (P<0.05) among treatments.

resulting leathers can be ascribed to the increased formation of crosslinks and the effects of aluminum. In this study, solo alum retanning was performed to attain higher softness of the resultant leather keeping in mind the end use of the leather as a snow-white glove. However, the tensile strength of Al-Veg combination tanned leather was found 3800 N/cm^2 by Haroun *et al.* (2012) whereas this study revealed tensile strength of 2300 N/cm^2. This is because the fiber bundles are effectively segregated in Al-Veg tanned leather, whereas cemented fiber bundles are visible in Veg-Al tanned leather. This observation suggests that Veg-Al tanned leather demonstrates elevated tensile strength and tear resistance, accompanied by reduced softness. Conversely, Al-Veg tanned leather exhibits diminished strength and increased softness (Haroun *et al.,* 2012).

The chrome retanned snow white glove leather manifested the highest tear strength (3600±200 N/cm), tensile strength (3340.00±243 N/cm^2) and sewing strength (17.14±0.05 N/cm) compared to formaldehyde, alum, chrome alum and control leather of this study while the abrasion fastness (4.67±0.57), shrinkage temperature (81.33±1.52 °C), and elongation (76.331±00 %) were found to be moderate comparatively. The softness (6.40±0,35 mm) was greater than formaldehyde and alum retanned leather however, lower than chrome and control retanned leather (Table 2). As such, the application of chromium as a tanning or retanning agent is widely used globally due to its ability to produce high-quality leather that is comfortable in feel, lightness, softness, brightness, and have high hydrothermal stability (Maina *et al.,* 2019). The purpose of using chromium as a retanning agent is to increase shrinkage temperature, increase chromium levels in leather, even up color, change the reactivity of the leather, and modify the properties of leather (Covington and Wise, 2020). However, the main drawback to using chromium as a tanning agent is the Cr pollution due to the discharge of huge amounts of Cr-containing wastewater and solid waste (Das *et al.,* 2022). Still, there is a gap in treating wastewater and a lack of facilities to manage solid waste properly in most countries. Therefore, researchers still searching for alternatives to gain the same properties of leather using more sustainable, bio-based tanning materials in the leather processing.

The snow-white glove leather retanned with the combination of chromium-alumunium retanning agent exhibited a high degree of softness (6.60±0.02 mm) and lower tear (1530±117 N/cm), and tensile strength (1500.00±100 N/cm^2), compared to other retanned leather other than control sample. The sewing strength (12.40±0.20 N/cm), and abrasion fastness (4.33±0.57) were found moderate compared to the other retanning agent whereas the % of elongation (47.50±0.57 %) was found lowest compared to all other leather. According to Brown (2017), chrome-alum, with the chemical formula of $KCr(SO_4)_2$, is employed in the process of leather tanning due to its ability to enhance skin stability through the interconnection of collagen fibers. The chrome alum utilized is a dodecahydrate, and the active chromium is on the form of $[Cr(H_2O)6]^{3+}$. According to Alaysuy (2019), the retanning process involved the utilization of a combination of chrome alum and urea in a Deep Electric Solvent (DES). The process of retanning utilizing chrome alum exhibits rapid penetration into the collagen structure. The results of the retanning process show that the tanning process took place, but the final sample had an internal hole due to overtanning from the high ionic strength. The collagen areas with a positive charge may collapse as a result of the high sulfate and low water concentration (Alaysuy, 2019). However, retanning with chrome alum can successfully enhance the shrinkage temperature (Ts), as seen by the Ts value in this study reaching 77.67°C, which is acceptable for glove leather manufacturing.

Table 2 shows the physical properties results for the control, with chrome alum having been used as the retanning agent. The physical properties achieved from the control leather exhibited a softness of 6.60±0.02 mm, tear strength of 560.00±40 N/cm, tensile strength of 570±70 N/cm^2, sewing strength of 4.12±0.20 N/cm, abrasion fastness of 5.00±0.57, Shrinkage temperature of 83.00±2.00°C, and elongation of 50.40±0.57 %. The obtained results of this study indicate lower tear strength and tensile strength values of the control sample while the chrome alum combination retanned leather exhibited promising softness and other properties as well. However, chrome alum retanning should not be done for extended periods of time as this may denaturize skin proteins and damage the skin's physical condition. Nevertheless, the control group showed the highest Ts value, reaching 83.00 ± 2.00°C. According to Alaysuy (2019) study, retanning with chrome alum can successfully enhance Ts.

Principal component analysis on leather physical quality

Principal component analysis (PCA) is a statistical technique used to consolidate several indices into a single aggregated measure that enables the reduction of variables, assesses the correlation between variables, and visualizes data (Wibowo and Yuliatmo, 2020; Yuliatmo *et al.,* 2021). Thus, reducing the number of variables entails transforming the original collection of variables into a new set of principal components, which are smaller in number (Łojewski *et al.,* 2014). In this study, eight different important component of the physical properties of leather retanned by various retanning chemicals have been analyzed by PCA.

The eigenvalue of the correlation matrix (Table 3) in the first principle component (PC1) showed that 7 of 8 variables have similar values (0.357-389), and the rest of the variables have the smallest value (0.024), while the variable thickness has the highest value in the second principle component (PC2). It was supported by the loading plot in Figure 2 which showed that softness, tensile strength, tear strength, stitch tear strength, abrasion fastness, shrinkage temperature, and elongation are in areas close to each other which indicated that they have almost the same effect on the physical quality of leather gloves.

Based on the principle components from various variables, it can be seen which type of leather has the physical qualities closest to the control. In Figure 3, there are 5 different types of leather retanned by formaldehyde, alum, chrome, chrome alum and control. It revealed that the physical properties of leather retanned by formaldehyde and alum showed far difference than the control sample while the leather retanned by chrome and chrome alum exhibited very close characteristics to the control sample (Figure 3). It is evident that the chrome alum type of retanning agent could be the well alternative of conventional snow white glove leather maintaining all other parameters, especially the softness.

FT-IR Analysis of the retanned leather

FT-IR spectra provide a quick method for identifying the functional groups of organic compounds. Figure 4 shows the FT-IR results obtained from leather treated with retanning agents such as formaldehyde (Fd), alum (Al), chromium (Cr), chrome alum (Ca), and a control (Ct). The presence of distinct peaks ranges at 3309.90cm^{-1} (Ca), 3308.90 cm^{-1} (Al), 3301.56 cm^{-1} (Fd), 3300.85 cm^{-1} (Cr), and 3301 cm^{-1} (Ct). This implies that the N-H linkages to amino acid groups are present in this region which is in congruence with other studies (Rachmawati *et al.,* 2020; Wibowo *et al.,* 2018). The strong band within the range of 3200-3500 cm^{-1} indicates the presence of hydroxyl groups, particularly those with a high concentration of carboxyl groups (Qiang *et al.,* 2016). According to Hassan (2019), amino acids serve as the fundamental constituents of the skin, where they are interconnected to create a linear polymer

Table 3. Eigenvalue of correlation matrix of the physical properties of snow-white glove leather

Variable	PC1	PC2	PC3	PC4	PC5	PC6	PC7	PC8
Softness	0.383	0.050	-0.203	0.474	0.548	0.486	0.209	0.066
Thickness	0.024	0.994	0.093	-0.023	-0.031	-0.021	-0.011	-0.003
Tensile strength	0.367	-0.061	0.555	-0.036	0.050	-0.185	0.441	-0.567
Tear strength	0.374	-0.056	0.474	-0.150	-0.062	-0.012	0.071	0.775
Stitch tear strength	0.357	0.034	-0.582	-0.606	0.060	-0.167	0.363	0.038
Abrasion fastness	0.384	-0.003	-0.216	0.346	-0.808	0.159	0.068	-0.055
Temperature shrinkage	0.389	-0.002	-0.144	0.325	0.186	-0.710	-0.428	0.006
Elongation	0.389	-0.027	0.106	-0.398	0.031	0.414	-0.661	-0.262

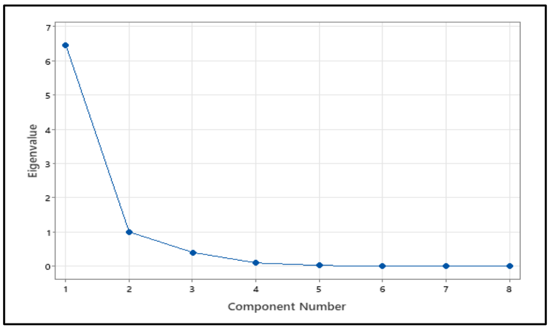

Figure 1. Scree plot for eight principal components from PCA.

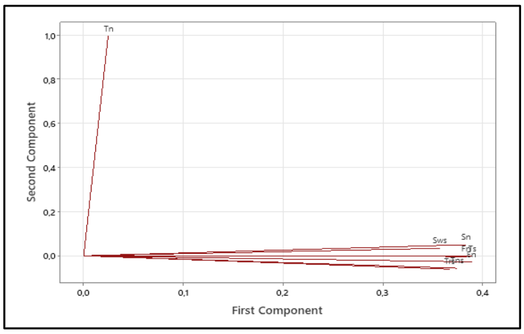

Figure 2. Loading plot of principal component analysis of Softness (Sn), Thickness (Tn), Tensile strength (Tns), Tear strength (Trs), Stitch tear strength (Sws), Abrasion fastness (Fn), Temperature shrinkage (Ts), Elongation (En).

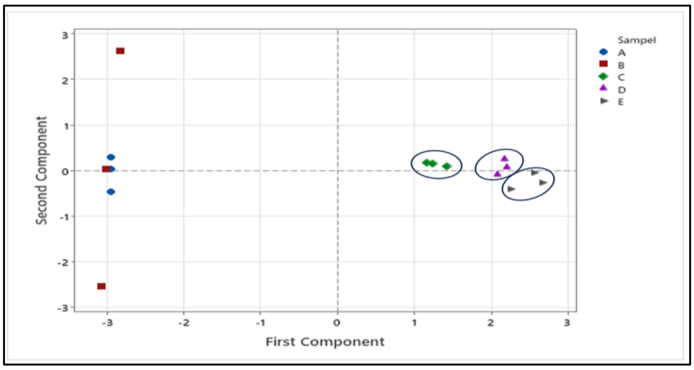

Figure 3. Principal component analysis score plot for the physical properties of leather retanned using Formaldehyde (A), Alum (B), Chrome (C), Chrome alum (D), Control (E).

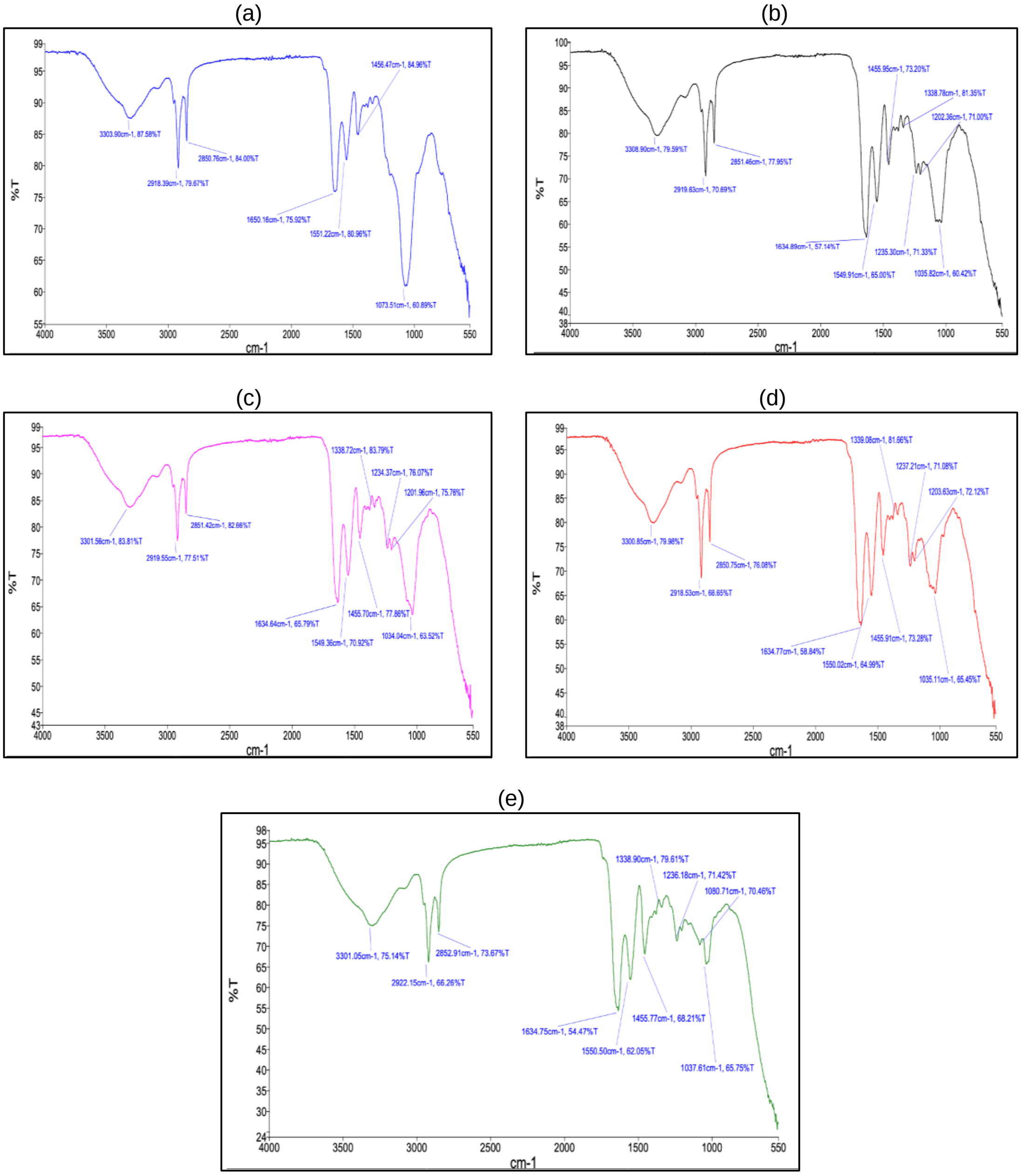

Figure 4. Infrared spectrum pattern of Chromium-Alum (a), Alum (b), Formaldehyde (c), Chromium (d), Control (e).

through the interaction between a carboxylic group on one amino acid and an a-amino group $(–NH_2)$ on another amino acid. Furthermore, Figure 4 show distinct peaks within the ranges of 2850-2918 cm^{-1} (Ca), 2851-2919 cm^{-1} (Al), 2851-2919 cm^{-1} (Fd), 2850-2918 cm^{-1} (Cr), and 2852-2922 cm^{-1} (Ct). It indicates that C-H bonds from the vibration of methyl and methylene groups are present that have been observed in this range elsewhere (Vyskočilová *et al.*, 2019; Qiang *et al.,* 2016). The chrome-tanned leathers show a significant absorption of C–H stretching, which enables them to interact with air oxygen and generate O-C-H stretch. This interaction is characterized by the presence of a diagnostic aldehyde band at 2830 cm^{-1} (Hassan, 2019).

Absorption range of 1551.22 cm^{-1} (Ca), 1549.91 cm^{-1} (Al), 1549.36 cm^{-1} (Fd), 1550.02 cm^{-1} (Cr), and 1550.50 cm^{-1} (Ct) refers to C-N-H bond (Hassan, 2019; Rachmawati *et al.,* 2020; Vyskocilova, 2019). The absorption at wave number 1553 cm^{-1} corresponds to the bending vibration of $-NH_2$ and the stretching vibration of CN in amide group II. On the other hand, the absorption bands at wave number 1547-1551 cm^{-1} indicate the presence of the C = N bond on formalin-tanned 1200 - 1350 cm^{-1}. This suggests the presence of a methylene group $(-CH_2-)$ in skin collagen, which is detected at wave numbers 1339 cm^{-1}. Furthermore, the absorption at wavenumber region 1241 cm^{-1} indicates the presence of a -NH group bond from amide III. Therefore, the FTIR analysis of the studied retanned leather confirmed the presence of individual retanning agents for producing snow white glove leather.

The resulting absorption values within the specified range of 1650.16 cm^{-1} for Ca, 1634.9 cm^{-1} for Al, 1634.64 cm^{-1} for Fd, 1634.77 cm^{-1} for Cr, and 1634.75 cm^{-1} for Ct. The spectrum of this wavenumber relates to C=O bonds, as reported by Hassan (2019), Rachmawati *et al.* (2020), and Vyskocilova (2019). Therefore, the presence of a sharp peak in the carbonyl group observed in chrome-tanned leathers might be due to the formation of additional carbonyl groups by chromium ions as they interact with collagen molecules (Hassan, 2019).

Conclusion

The physical properties highlight that using formaldehyde as a retanning agent for snow white goat leather increases tear strength and tensile strength but reduces softness and abrasion fastness significantly ($p<0.05$), whereas alum boosts physical properties such as shrinkage temperature and tensile strength due to enhanced cross-linking with skin collagens. However, retanning with chrome-alum exhibited the highest softness with moderate tear and tensile strengths. In addition, PCA analysis showed that chromium-alum retanned leather exhibit the best fit with commercial snow white glove leather. Furthermore, PCA suggested that almost all variables have a significant influence on the physical quality of the skin, except for the skin thickness variable. The FT-IR analysis of leather treated with various retanning agents reveals similar functional groups, indicated by comparable peaks and stretching patterns. Therefore, chromium-alum as a retanning agent for snow-white glove leather production could pave the way forward to achieve necessary characteristics of snow-white glove leather.

Conflict of interest

The authors declare that there are no conflicts of interest that could have influenced the outcomes or interpretations presented in this manuscript.

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Author’s contribution

Conceptualization-Yuliatmo R, Rahmawati A, Mizan A, Methodology-Maryati T, Yuliatmo R, Rahmawati A, Investigation and Resources-Maryati T, Writing original draft presentation-Yuliatmo R, Rahmawati A, Maryati T, Adamu D, and Mizan A. All authors have read and agreed to the published version of the manuscript.

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