Textile & Leather Review

ISSN 2623-6281 | www.tlr-journal.com | 10.31881/TLR

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How to cite: Bekele WW, Ramaiah GB. Utilization of Tannery Flesh Waste for Production of Sustainable Leather Coating Substance. Textile & Leather Review. 2023; 6:114-131. https://doi.org/10.31881/TLR.2023.008

How to link: https://doi.org/10.31881/TLR.2023.008

Published: 30 March 2023

Utilization of Tannery Flesh Waste for Production of Sustainable Leather Coating Substance

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Article

https://doi.org/10.31881/TLR.2023.008

Received 05 February 2023; Accepted 26 March 2023; Published 30 March 2023

ABSTRACT

The leather tanning industry is one of the most polluting industries because it generates a gigantic amount of solid, liquid, and gaseous waste. Solid wastes are raw trimmings, fleshings, chrome shavings, buffing clouds of dust, and keratin wastes. Flesh wastes are generated from tanneries during leather processing, especially in sheep liming. These skin-flesh wastes from tanneries contain significant fat content, which is being wasted and dumped in open areas or at dumping sites. As a result, this flesh waste disposal has created a major environmental problem. This research aims to extract a sustainable functional substance that can be used for imparting aesthetic properties (polishing) to a leather product. In this research, a calculated amount of flesh waste was washed, de-limed, and cut into small pieces for solvent extraction of fat using the Soxhlet apparatus and hexane as solvent. Chemical analysis of the extracted fat showed a saponification value ranging from 189 to 192. These values indicate that extracted fat has a triglyceride, which can be used as a lubricant. However, when the extracted fat was applied to leather products like shoes, the surface showed an increase in their aesthetic properties (lustre, and gloss) giving excellent polishing results. The acid and saponification value, fat content, melting point, density, and viscosity of extracted fat showed stability making it a promising substance to impart functionality for leather products and usage as a polishing material. The extracted fat could replace synthetic, functional, and polishing materials overcoming the environmental challenges posed by the leather tanning process.

KEYWORDS

coating substance, flesh waste, fat extraction, saponification value, acid number

INTRODUCTION

The finishing material for leather is usually a waxy or creamy substance giving the leather surfaces functionality like a shiny look, water-repellent properties, or improving and restoring the appearance of shoes and leather products. These substances occur both in liquid and semisolid form. Most leather products require a good aesthetic appearance and a functional finish to enhance their durability and keep them lively and flexible. Leather finishes usually consist of the application of fat (wax) and solvents.

Leather products are a by-product of meat and dairy production. In leather processing, raw materials are classified as high pollutants due to their ability to generate liquid, solid, and gaseous waste [1].

Even though there are several operations during tanning, a significant number of tannery pollution loads are contributed by the beam house operation (fleshing operation).

In the leather-making process, of 1000 kg of rawhide 850 kg of solid waste is produced and only 150 kg of the basic materials are turned into leather [1]. Solid waste known as "flesh waste" is produced during the initial stages of leather production. Green flesh, lime flesh, split lime flesh, and trimmed meat are just a few of the by-products of the tanning process that are produced.

During the mechanical beam house process, the flesh layer is thrown out. This layer makes up 15% of the thickness of raw hide or skin. During the tanning process, the fleshing waste contains several substances that are used to make leather. The raw hides and skins went through different mechanical and chemical processes, so these fleshing wastes have many different qualities. These flesh scraps are produced following liming operations, and when the raw material was being prepared for soaking [3], they were not properly used or disposed of and caused many problems for the environment.

Various amounts of fatty acids, fatty tissue, blood vessels, nerves, and voluntary muscles make up the flesh.

Due to the putrefactive nature of flesh, it cannot be kept in the tanning yard [3]. Both de-limed flashings and residual hairs are important sources of protein and have many uses as bio-fertilizers in agriculture and horticulture. In recent years, researchers have developed a method to recover collagen protein and fat fractions from this material [4]. However, previous research has not addressed simple methods of recycling or technical aspects of flesh waste nor conducted tests on its acid value, saponification value, density, viscosity, and melting point.

In recent times, coating and finishing chemicals used for leather products contain hazardous substances made from ingredients such as naphtha, lanolin, wax, potassium bicarbonate, and other types of colour pigments. It also contains sodium and potassium carbonates and bicarbonates [5]. This substance, when used on footwear and applied to leather materials, for example, Bags, etc., pose a hazard to the user [6]. If the finishing substances are made from natural sources like natural wax, recycled flesh waste fat, etc., they are less harmful both to the user and the environment. So, using techniques and methods for extracting fat from flesh waste is a novel approach, and analysing its ability as a functional substance is a significant contribution to achieving sustainability and addressing environmental concerns.

The importance of this research in extracting fat using a scientific method can be seen in the enormous amount of flesh waste produced during the manufacturing of leather, as well as the potential for recycling the fat content and functional value of the waste flesh. To the authors' knowledge, no research focused on extracting fat for usage as a lubricant or a functional substance to impart aesthetic and durable properties to leather products.

The main research question in this research is to evaluate the usefulness of the extracted fat substance from flesh waste as a functional substance. The extracted substance was tested as a polishing material, and its polishing effect was evaluated.

LITERATURE REVIEW

Historically, natural materials like tallow wax have been used to prepare polish, and coat leather surfaces without using any additional chemicals. However, synthetic coating recipes were initially produced in the 20th century using chemical additives, and many of those compositions are still in use today [5]. The synthetic coating material is usually formulated by mixing different precursor chemical substances (both organic and inorganic), such as polyethylene wax, carnauba wax, microcrystalline wax, paraffin wax, beeswax, shellac wax, naphtha, turpentine, ethylene glycol, pigments, and dyes [7]. This is the existing trend in the preparation and application aspects of coating substances.

These days, paraffin wax is one of the most common synthetic coatings used to polish leather surfaces. Paraffin waxes come from crude oil and are sorted by how much oil they contain and how much they have been refined [8]. It can be blended with other types of waxes to give a variety of finishes and aesthetic appearances [9]. Paraffin coating repels other solvents or dust from the film surface and imparts elasticity and gloss to the film without destroying its hardness.

Paraffin wax is composed of hydrocarbons, saturated n-, and iso-alkenes. The general molecular formula is C_nH_{2n+2} [10]. The chemical structure of paraffin wax is shown in Figure 1. The application of a coating substance involves the formation of a microscopic film, resulting in a surface with higher reflection properties and glossiness. The particle size of the coating substance plays a prominent role in giving this effect. If the particle size is large, an uneven microsurface is created, resulting in the scattering of light and producing the Matt effect. However, when the paraffin coating substance is applied to this kind of surface, the pores are filled and a shiny surface is produced. This mechanism applies to all kinds of synthetic coating waxes. In the case of liquid-form coating materials, they do not require rubbing since they form a film very quickly as compared to wax-based ones.

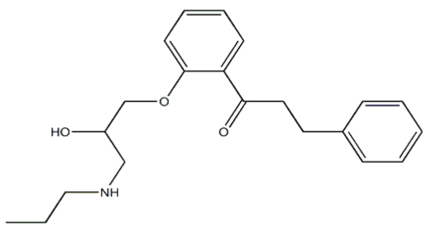

Figure 1. Paraffin wax's chemical structure

When paraffin wax is applied to the leather surface, it undergoes a combustion reaction. The chemical reaction is exothermic, involving the liberation of heat, water vapour, and carbon dioxide. The reactions of paraffin wax are highly sensitive to the skin [11], causing a heat rash and other health complications. The skin part of animals is usually composed of monounsaturated fats, ceramides, and cholesterol. The animal fat extracted from tannery waste is rich in unsaturated fatty acids [12]. The extracted fat is mainly composed of triglycerides. The chemical structure of triglyceride is shown in Figure 2. This triglyceride is mixed with solvent, pigment, and other chemicals in preparation for coating the substance. The prepared coating substance is not harmful to human skin and does not degrade the leather surface. The triglycerides used in preparation have a low melting point, undergo a hydration reaction, and increase their fluidity properties. All these properties make the leather surface flexible and create an even microporous film, enhancing the aesthetic appeal of the leather surface.

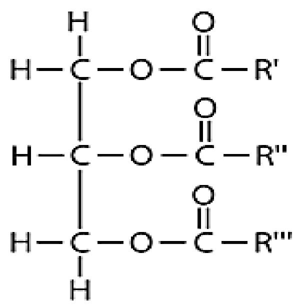

Figure 2. Structure of triglyceride

EXPERIMENTAL

Material and methods

Flesh waste: An excess amount of sheep's lime flesh waste was procured from Bahir Dar Tannery PLC. The flesh waste was de-limed, washed with cold water, and taken for fat extraction. The flesh waste is shown in Figure 3.

Figure 3. Lime Flesh waste from tannery

In addition to flesh waste, the following chemicals were also used for the extraction of fat and preparation of functional coating chemicals:

Hexane (C_6H_{14}): A non-polar dissolvable with a boiling point of 68 °C and is used as a solvent for the extraction of fat.

Hydrogen peroxide (H_2O_2): A small amount of hydrogen peroxide is used as an oxidizing and disinfecting agent for killing bacteria, viruses, and fungi present in the extracted fat and is also used as a bleaching agent.

Formic Acid (HCO^2H): Used for the neutralization of chemicals and to control extracted substances' pH.

Polyurethane binder (Pidithane AP 111): A binding agent to the substrate.

Protein binders (casein): Used to impart more lustre properties to a substance.

Naphtha (C_6H_{12}): Used as a solvent to mix fat and other ingredients.

Colouring pigment (tan A): used for producing the colouring effect of coating substances.

Paraffin wax: used as a control group during the experiment.

The formulation for preparing the coating substance uses extracted fat from flesh waste and paraffin wax. Three diverse formulations of coating substance were prepared and labelled as samples A, B, and C, as shown in Table 1.

- Sample A: This was formulated from the extracted fat as an experimental group.
- Sample B: This was formulated from extracted fat and control paraffin wax as an experimental group.
- Sample C: This was formulated from paraffin wax only as a control group for the experiment.

Table 1. Chemicals used for the preparation of coating substances

Chemical name	Purpose	Sample A (%)	Sample B (%)	Sample C (%)
Extracted fleshing fat	Main Leather coating material	98	49	-
Hydrogen peroxide	For bleaching effect and to remove bad smell	0.2	0.2	0.2
Formic acid	To adjust the pH value	0.1	0.1	0.1
Polyurethane binder	Used for waterproofing	0.1	0.1	0.1
Protein binder	To enhance the luster of coating material	0.1	0.1	0.1
Naphtha	Used as solvent	0.1	0.1	0.1
Pigment	For color effect	0.01	0.01	0.01
Paraffin wax	Synthetic coating substance	-	49	98

Test method

The fat extracted from the fleshing waste and the coating substance were tested according to the following methods: Extraction of fat by using the Soxhlet method was determined according to ISO

13944:2012. Fat content was determined according to ISO 6492:1999. The acid number of the extracted fat was determined according to ISO 660:2009. The saponification number of extracted fat was determined according to ISO 3657:2013. The density of the coating material was determined according to ISO 2811. The melting temperature of the coating substance was determined according to ISO 6321:202. The viscosity of the coating substance was determined according to ASTM D2983. The rub resistance of polished leather was determined according to ISO 24265:2020.

Methodology

Figure 4 shows the steps that were taken to get coating chemicals out of waste meat.

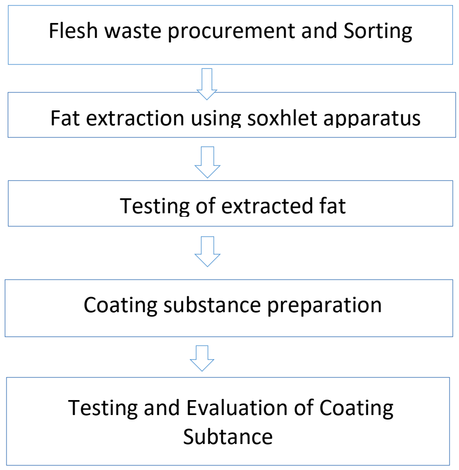

Figure 4. Methodology of research

De-liming and washing of the flesh waste were the primary steps in sample preparation. It was done to reduce the pH of the flesh waste from 12.5 to 8.00. The next step was to cut the de-limed and washed flesh waste samples into small sizes (3 x 5 mm) for extraction of fat. After cutting the samples, pH was checked using pH paper to make sure a proper pH is maintained before extraction.

Extraction of fat

A Soxhlet apparatus was used to extract the fat from cut flesh samples. A measured amount of flesh waste was covered with a filter paper bag and placed in a glass thimble. The glass thimble was then placed inside a Soxhlet glass chamber for extraction. Extraction was performed using a Soxhlet apparatus equipped with an electrical setup. One Soxhlet beaker and one condenser piece were connected to the extractor. From above, hexane solvent was injected into the condenser. The solvent was then applied to the sample. For this experiment, samples of dried flesh weighing on average 35 grams each were collected. For nearly three hours, the aggregate extraction process was completed. Following the completion of the extraction, the extra solvent remaining in the beaker was separated

https://doi.org/10.31881/TLR.2023.008

from the fat and put to use in the following batch of operations. The soxhlet extractor set-up used for the extraction of fat is shown in Figure 5.

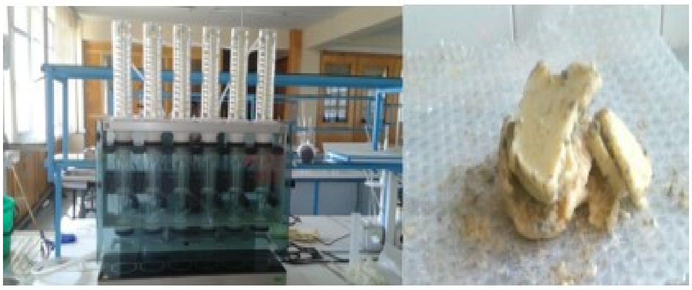

Figure 5. Soxhlet extractor and extracted fat

Determination of fat content

The extracted fat content in the samples was determined gravimetrically. The empty weight of the sample holding flask was recorded using a digital weight balance. The sample holding the bottom flask was weighed with the extract and recorded after extraction. The weight difference between the final sample holding the bottom flask and the weight of the empty bottom flask gave the amount of fat extracted. The maximum yield of flesh waste was calculated under the formula given below [13]. The formula for per cent fat content is shown in equation 1.

$$\%\text{ fat content} = \frac{W2-W1}{W}*100 \tag{1}$$

where:

W_1= weight of extraction empty bottom flask (g)

W_2 =weight of the crude fat plus extraction bottom flask (g)

W = sample weight (g)

Acid number

The acid number (AN) is the amount of potassium hydroxide, in milligrams, needed to neutralize the free acid in 1 gram of material. If other acidic substances are present in the system, the acid number may be skewed. Acid number [14] often makes the process of turning triacylglycerol into free fatty acids go much faster. The acidity of the extracted fat was obtained according to the ISO 660:2009 method. The formula for determining acid values is shown in equation 2.

$$AN = \frac{mL\ of\ KOH*N*56.1}{weight\ of\ sample} \tag{2}$$

where N = Normality of KOH

Saponification number

The saponification number is the number of milligrams of potassium hydroxide (KOH) needed per gram of material to saponify esters and neutralize free acids. The saponification number is a gauge of the sample's triacylglycerols' average molecular weight. The average molecular weight of the triacylglycerol in the sample was used to calculate the saponification number. By using an alkaline environment, neutral lipids are broken down into glycerol and fatty acids, a process known as saponification. The lower the saponification number, the higher the average molecular weight of the triacylglycerol [15]. The formula for determining saponification values is shown in equation 3.

$$SN = \frac{\text{N HCl} * 56.1(\text{B}-\text{S})}{\text{sample weight in gram}} \tag{3}$$

where:

B = millilitre (mL) of HCl required by Blank,

S = millilitre (mL) of HCl required by Sample.

Preparation of coating substance

To make the leather coating, 50 grams of extracted fat were placed in a beaker and heated to a melting point between 40 °C and 50 °C. A thermostat was in charge of regulating the temperature. Start stirring as soon as the fat has melted and turned liquid. Then, different amounts of chemicals were added for different purposes. The prepared coating material is shown in Figure 6.

Figure 6. Coating substance

Melting temperature of the coating substance

The dish with the leather-coated material inside was set on top of the electric heater. The temperature was managed by placing a thermometer on the plate. At 45 °C, the melting temperature of the extracted fat samples, the sample melted.

The density of the coating substance

Using a digital balance, the amount of extracted leather coating substance was placed in a beaker and weighed. A measuring cylinder with a known amount of water was filled with the same amount of leather coating. Changes in volume were noted and recorded. The formula for determining density is shown in equation 4.

$$\text{Density} = \frac{\text{Mass}}{\text{Volume}} \quad (4)$$

The viscosity of coating substances by the rotary viscometer method

In this test, the coating material was put in a glass tube and heated to a certain temperature in an insulating block. Then, the torque needed to turn a metal spindle is found by turning the spindle at a set speed of 20 rpm in the coating material. The absolute viscosity of the coating material can be calculated based on the internal rotation resistance that the coating substance's shear stress offers. This method is commonly known as the Brookfield method and is described in ASTM D2983. shown in Figure 7.

Figure 7. Rotary viscometer

Evaluating the Performance of a Coating Substance

Surface gloss/lustre property

The coated sample surface was checked for lustre/gloss characteristics by comparing them with each other (Samples A, B, and C).

Surface resistance to rubbing properties

The coated leather sample surface was rubbed approximately 40 times with clean, white cotton. The shade changes and degree of staining was examined.

Surface resistance to dust

Coated leather sample surfaces were exposed in an open environment where dust was easily accessible for 12 hours, and then the degree of dust adsorption was checked.

Visual Performance Evaluation

The scale was designed for visual assessment based on the pool of samples of different polished leather surfaces that were available at our laboratory [16]. A scale was designed and is shown in Table 2.

Table 2. Designed scale for visual assessment

Scale (1-10)	Grade
9 and above	Excellent
7 - 9	Very Good
5-7	Good
< 5	Bad

RESULT AND DISCUSSION

Characterization of flesh waste

pH and fat content of the extracted fat

The pH of the limed flesh used for the experiment averaged 12.5 and the fat content of the samples that were lime fleshed showed an average of 25.77%. The fat content of the flesh waste is shown in Table 3.

Table 3. Fat content values of flesh waste

Parameter	Recorded value (average)	Standard deviation
Weight of Flesh (g)	35.13	0.44
Weight of extracted fat (g)	9.05	0.45
The percentage of Fat content (%)	25.77	2.24

Lime-fleshed samples from sheep skin were observed to have a maximum fat content. From Table 3, the average sheep flesh waste was 35.13 g. On average, 9.05 g of fat with a standard deviation of 0.45 are extracted from flesh waste. Based on the experimental analysis, a 25.77% average fat content with a 2.24 standard deviation has been reported. From the data trend analysis, an inference can be made that there is no linear relationship between flesh weight and fat content percentage. The fat content percentage depends on the growth and genetic factors of the sheep breed, the individual within the breed, the degree of fatness at the time of slaughter, nutritional treatment, and age [17]. Due to this, the estimated error in the measurement values is more spread out and less reliable. Generally, in some sheep breeds, higher levels of fat content percentage can result in higher amounts of extracted fat. These breeds are useful not only for achieving a higher yield of fat but also because the extracted fat is superior for making a coating substance.

The acid number of extracted fat

Rancidity is determined by an acid number. The fat or oil will get rancid if the values are high, and vice versa. The acidity of sheep flesh samples that had been limed was measured, and the values are represented in Table 4.

Table 4. The acid number of extracted fat

Parameter	Recorded value (average)	Standard deviation
ml of KOH @ 0.1 N	4	0.16
Weight of the sample (g)	2.65	0.11
Acid number	8.47	0.092

For the preparation of a leather coating substance, the acid number should be less than 8.6. It is observed from Table 4, that the mean acid number of the fat removed from the lime flesh of sheep skin is 8.47 mg/g with a standard deviation of 0.092. The acid number of all the extracted fats is less than 8.6 mg/g, thus indicating the fat is in good condition and can be used for making a coating substance [18].

Saponification number of extracted fat

The larger the mean molecular weight of the triacylglycerol contained in the samples, the lower the saponification number. The ratio of saponification to average fatty acid molecular weight is inversely related. The values in this study were within the ranges that were predicted. The saponification number of extracted fat from lime flesh waste that is required for preparing the leather coating is shown in Table 5.

Table 5. Saponification number of extracted fat

Parameter	Recorded value (average)	Standard deviation
ML of HCL (B-S) @ 0.2 N	35.40	0.66
weight of the sample (g)	2.09	0.038
Saponification number	190.22	0.83

The average saponification number of sheep's lime flesh is 190.22, with a standard deviation of 0.83. All results fall within the standard value range, which is 180 to 233 [15]. Hence, the result shows the occurrence of triglycerides, which can be used for the extraction of coating material. The results show there is no such defect when the extracted fat is used as a coating substance. The surface properties of the extracted fat with a saponification value in the range of 180–233 do not show any surface peeling or degradation when used on leather material.

Characterization of the coating substance

Melting temperature and density

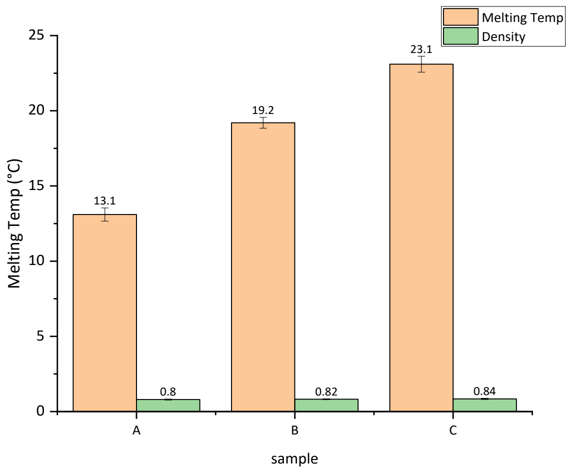

Figure 8. Melting Temperature and Density of the Coating Substance

The melting point and density of the coating substance extracted from lime sheep's flesh are shown in Figure 8. The temperature at which samples transition from the solid phase of the leather coating substance to a liquid phase is plotted by considering the change in the physical state of the coating substance when subjected to heating. All three samples (A, B, and C) show an average melting temperature of 13.1 °C, 19.2 °C, and 23.1 °C with a standard deviation of 0.44, 0.36, and 0.53, respectively. Sample A has a lower melting point than other samples. This difference in their melting points can be attributed to changes in bonding properties like the strength of bonding between the fat molecules and the molecular weight of the fat molecules [19]. Also, if a substance has a higher density, it is heavier. Likewise, a lower density means it's much lighter. The densities of samples A, B, and C show an average of 0.80 g/cm^3, 0.82 g/cm^3, and 0.84 g/cm^3 with a standard deviation of 0.04, 0.03, and 0.04 respectively. Samples that contain extracted fat have a lower density than others. So, sample A is lighter than the other samples.

The viscosity of the coating substance

The coating substance that was extracted gets thicker as the temperature goes up (the viscosity value goes down). Temperature affects the viscosity of the samples studied and their flow properties. The drop in viscosity values is shown in Figure 9. Sample A has a low viscosity compared to other samples. The main reason for the low viscosity in Sample A could be attributed to its higher fat content as compared to other samples.

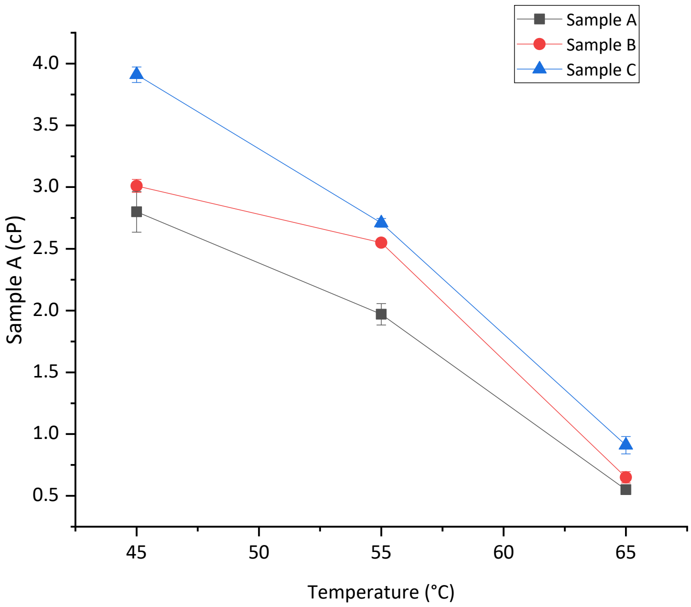

Figure 9. The viscosity of the coating substance

The viscosity of a fluid is the opposite of its fluidity, which denotes how easily it can flow. As shown in Figure 9, when the temperature increases, the fluidity of the coating substance also increases, but the viscosity is reduced. All three samples (A, B, and C) show a viscosity of (2.80, 3.01, and 3.91) at 45 °C, 1.97, 2.55, and 2.71) at 55 °C, and 0.55, 0.65, and 0.91) at 65 °C, respectively. Viscosity determines internal friction between the molecules comprising the fluid. Viscosity is an important property that affects the surface application of coating substances. If the viscosity of the coating material is lower, molecules flow more quickly, and it will be applied and penetrate the leather surface more easily. Sample A has a lower viscosity than samples B and C, which determines the ease of coating the substrate leather surface.

Application of coating substances

After making the coating and figuring out what it was made of, the coating (Sample A) was put on the surface of the leather shoe, as shown in Figure 10. Samples B and C were used as control samples in this study. The coating substance was applied by padding with a brush and then allowed to dry for 10 minutes. Then, continuous brushing of the leather surface was carried out manually. The appearance of coated and uncoated leather surfaces showed prominent differences in their appearance, water-repellent characteristics, and overall aesthetics of the leather surface.

Figure 10. Visual picture of samples A, B, and C coated on leather surfaces, respectively

As shown above, in Figure 10, the visual assessment proves the effectiveness of the prepared sample A when used as a coating substance on the leather surface. The formulations of Sample A, after being applied to the leather surface, were durable, and there was no patched appearance nor staining observed on the leather surface.

Performance evaluation of leather surfaces

The samples were evaluated visually by 30 respondents on a 1–10 subjective assessment scale for their surface properties, namely lustre or gloss, rub resistance, and dust absorption resistance. The average scores and their grades are also shown in Figure 11.

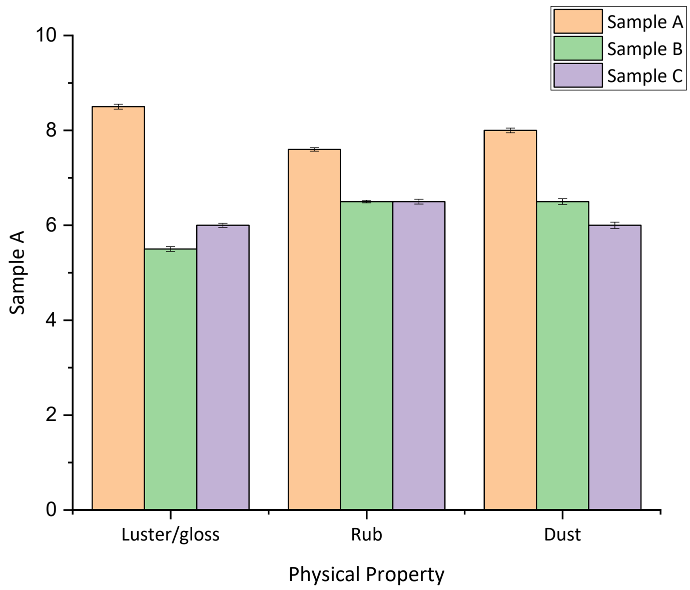

Figure 11. Coating substance performance

Figure 11 shows that Sample A has an average subjective rating of more than 7.0, which means that it has very good gloss properties, rub resistance, and resistance to dust absorption. Sample A did not only have a good subjective rating, but it also did not stain easily and did not wrinkle. However, samples B and C showed lower levels of glossiness, rub resistance, and dust absorption, as indicated in Figure 11. The lustre or gloss effect of a coating has a close relationship with the smoothness of its layer. For an extremely smooth surface, incident light will be completely reflected at a certain angle, showing a high gloss effect [20]. On a rough surface, much light is diffusely reflected instead of undergoing total reflection. Sample A showed better results than Sample B and Sample C. This may be due to sample A having a high extracted fat content, resulting in a better lustre or gloss property.

Rub resistance is an important test for most types of coating materials. It is done by rubbing them with white felt to see how well they hold up. For durable products that maintain quality of appearance over their intended lives, it is important to ensure that the surface of the coated materials used demonstrates rub resistance to surface polishing marring. Sample A has a much better rub resistance rating than samples B and C.

CONCLUSIONS

The goal of the research was to find ways to turn waste flesh from the tanning industry into useful coating materials. The fat was successfully extracted from the flesh waste using a solvent (hexane) by the soxhlet extraction method. The extracted fat contained an average of 25.77% of unsaturated fat. A sustainable material for the production of coating substances for polishing leather products has been successfully prepared using different recipes. The saponification value of fat was an average of 190.22, which proved the suitability of extracted fat for coating applications. This coating substance showed better thermophysical properties like density, viscosity, and melting temperature. Three samples showed an average density of 0.80 g/cm3, 0.82 g/cm^3, and 0.84 g/cm^3, respectively, and an average melting point of 13.1 °C, 19.2 °C, and 23.1 °C, respectively. The samples' viscosity at 45, 55, and 65 °C showed an average viscosity ranging from 3.91 to 0.55. One sample showed good subjective ratings when visually examined. High ratings indicated very good aesthetic properties when used as a polishing material on leather shoe surfaces. This extraction of coating substance requires low investment and inexpensive raw materials. Good saponification results promise the usability of recycled fat for the preparation of coating substances. This solid waste can fetch extra profit for the tanners, save money invested in disposing of it, and promote green technology.

Author Contributions

Conceptualization – Bekele WW; investigation – Bekele WW and Ramaiah GB; writing reviews and editing – Ramaiah GB and Bekele WW. All authors have read and agreed to the published version of the manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

Funding

This research received no external funding.

Acknowledgements

The authors are thankful to the Technical and Vocational Training Institute for providing the necessary facilities to carry out the research work.

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