Evaluation of the Physical Properties of Goatskins Tanned using Banana (*Musa spp.*) Leaf Midrib Tannins

by

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Abstract

Vegetable tannins have been studied over the years with an aim to reduce the pollution load caused by chrome tanning. Although mimosa tannins have been utilized commercially, they are expensive and not readily available. The purpose of this study was to assess the physical properties of leather tanned with banana leaf midribs tannins in order to determine their suitability as vegetable tannins. Selected banana leaf midrib samples were collected from Gikondi village in Mukurweini, Nyeri County, Kenya. They were shade-dried and ground into powder. The skins were processed conventionally using banana leaf midribs tannins of *Musa sapientum Linn.* and 'Muraru' (AA genome), with mimosa as a control. The physical properties of the resultant leathers were determined following the standard IUP methods. It was found that *Musa sapenitum Linn*, 'Muraru' (AA genome) and mimosa-tanned leathers had average shrinkage temperatures of 80.33 ± 0.74°C, 78.67 ± 0.47°C and 81.67 ± 0.94°C, respectively. The properties of the tanned leathers were compared with those of the control-tanned leather. Assessment of the physical properties indicated that the leathers met the minimum recommended values safe for *Musa sapentium Linn*-tanned leather, which failed at 30,000 flexes. These results indicate that banana leaf midribs can be used as an organic tanning agent source for production of leathers from goatskins.

Introduction

The present market value of the leather industry worldwide is around US$50 billion.[1] However, majority of the hides and skins produced in Kenya, the sixth-largest raw material producer for the industry in the world, are exported. Notwithstanding, the industry plays a significant role in Kenya's economy by employing hundreds of citizens across the nation. Leather production involves the stabilization of collagen by crosslinking its fibers to prevent deterioration and increase its hydrothermal stability.[2] Vegetable-, chrome-, and aldehyde-tanning are the three most utilized tanning techniques.[3]

Vegetable tanning involves the use of plant polyphenols (also known as tannins) to stabilize the collagen against putrefaction. These tannins are categorized into condensed tannins and hydrolysable tannins.[4] Leathers produced using these polyphenols are usually firm, compact and flexible, making them suitable for production of sole leathers, upholstery and leather goods. Polyphenols have also been utilized in tanning and retannage as they offer a filling effect among other attributes as mentioned.[5] The modification is thought to be a result of the polyhydroxy groups bonding to the active chains of the polypeptides, especially the carboxylic chains, either by hydrogen bonding, ionic bonding or covalent bonding.[6]

Tannins may be acquired from galls, which are primarily present in plant branches and leaves, and form in response to insect and parasitic attacks. The tannins utilized during tanning are typically round-shaped, smooth, and contain tannin levels ranging from 40% to 70%.[7] Fruits form another source of tannins used in leather production. Valonia and dividivi trees provide fruit-derived tannins, which have been utilized in Australia, Germany, and France due to their ability to provide characteristics such as solidity and weight to leather. They are also linked to the production of leather that possesses a degree of water resistance.[8] Leather production using sumac leaves has been practiced, and these leaves reportedly contain approximately 25% tannins.[9] In the 19th century, quebracho was investigated and employed for leather production purposes.

Tree barks are the primary source of tannins, which typically contain tannin levels ranging from 6% to 17%. Oak tree barks have been conventionally utilized as vegetable tanning agents, and other types of tree barks, such as acacia, have also been used for this purpose.[10,11] Coffee grounds have equally been utilized to tan leather.[12] The use of tree barks from acacia, if not managed appropriately, could result in deforestation, which in turn could have adverse effects on global warming.[13] Therefore, there is need to explore alternative sustainable sources of vegetable tanning agents.

Banana farming is well established, ranking among the top five in the world. Production capacity is estimated at ninety million tons of this fruit among the tropical and sub-tropical regions like Africa, South and Central America and South Eastern Asia producing 13%, 28% and 47% respectively.[14] Banana cultivation occupies approximately 1.4 million hectares or 38% of agricultural land in Uganda, thus making it the most extensively grown crop.[15] It is also regarded as one of the key crops for ensuring food security in Central, Western, and Eastern Africa.[16] Bananas (*Musa spp*) are a staple food and a source

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of income to a large number of homesteads across East Africa. The region provides about half of the bananas sold in Africa.[17] In Kenya, the types of bananas grown are mainly the cooking and dessert varieties, and they are mostly grown in Central and Nyanza regions.[17] Their distribution is greatly influenced by feeding behavior, supply and demand ratio, local tastes and climatic conditions.[18]

Studies conducted on Kepok bananas (*Musa paradisiaca L.*) have shown that the tannin levels in the leaves, trunks, and peels of these bananas range from 3.7% to 5.5%, making them a potential source of vegetable tanning agents.[19] Banana leaf midrib (*Musa acuminata balbisianacolla*) has been estimated to contain tannins in the range of 12% to 16%.[20] *Musa sapentium Linn.* and 'Muraru' (AA genome) tannin contents were determined and found to be 11.71 ± 0.33% and 6.36 ± 0.19%, respectively.[21] This study is aimed at assessing the physical properties of the leathers produced using selected banana leaf midrib tannins (by comparing the results with set minimum standards) in order to determine their potential to substitute or supplement mimosa as a vegetable tanning agent. This will ensure sustainable source of plant tannins.

Experimental

Materials

Banana leaf midribs of *Musa sapientum Linn.* and 'Muraru' (AA genome) were collected from Gikondi village in Mukurweini, Nyeri County. Powders were prepared following a procedure published in the literature.[21] Wet salted goatskins were purchased from a local slaughterhouse in Nyeri, Kenya. Mimosa was purchased from Sagana Tanners Limited and all the other chemicals used during the leather processing were purchased from Priyann Industries. The chemicals used were of reagent grade.

Tanning process

The sets of skin were weighed and treated through the same beamhouse processes and separated into three groups at the tanning stage. During beamhouse treatment, soaking was carried out using 300% (weight/weight) water, 1% wetting agent, and 0.01% fungicide for 24 hours. Then paste-liming was done to unhair the goatskins using 20% water, 8% lime and 2% sodium sulfide. The paste was applied on the flesh side of the skins, folded and then left to stand overnight. The unhaired skins were then relimed with 200% water, 5% lime and 0.05% sodium sulfide for 24 hours and the pH was checked. The liquor was drained, skins washed and liquor drained again. The pelts were then fleshed. Before deliming, the pelts were weighed. Deliming was carried out with 150% water and 2% ammonium sulfate and run for one hour and then the pH was checked. 1% bating enzyme was added and the process run for another hour. Thumb test was performed to confirm the completion of the bating process.

The goatskins were treated with extracts from two different types of bananas, and with mimosa as the control tanning agent. In three separate drums, pre-tanning was done using 100% water and 1% glutaraldehyde for two hours.[19] Moreover, for the control, 30% mimosa powder was added in four batches as shown in Table I and for the experimental leathers, 40% of the selected powdered banana leaf midribs were added in five batches as shown in Table II.[22,23] Penetration was confirmed by cutting a cross-section at the neck region of the pelts. The process was continued until the tannages were complete and the tanning liquors in the three drums were exhausted. Then, formic acid (1%) diluted with water at a ratio of 1:10 (volume by volume) was added in two steps and the process was run for two hours. The pH was measured. The leathers were drained, washed, and left to age overnight. Fatliquoring was done with 100%

Table I
Process recipe for Mimosa tanning

Process	Chemical	Percentages (%)	Duration (hours)	Remarks
Pre tanning	Water	150		
	Glutaraldehyde	2	2	
Tanning	Mimosa	5	24	Check penetration
		5	24	pH 5.0
		10	24	
		10	24	
Fixing	Formic acid	1	2	pH 3.5
Drain, wash and drain. Leave Overnight for aging				
Fatliquoring	Sulfated fatliquor	3	2	
	Sulfited fatliquor	3		
Fixing	Formic acid	1	2	
Drain, wash and drain. Horse overnight. Toggle dry				

Table II
Process recipe for banana leaf midrib tanning

Process	Chemical	Percentages (%)	Duration (hours)	Remarks
Pre tanning	Water	150	2	
	Glutaraldehyde	2		
Tanning	Banana (*Musa spp*) leaf midrib powders	5	24	Check penetration
		5	24	pH 5.0
		10	24	
		10	48	
		10	48	
Fixing	Formic acid	1	2	pH 3.5
Drain, wash and drain. Leave Overnight for aging				
Fatliquoring	Sulfated fatliquor	3	2	
	Sulfited fatliquor	3		
Fixing	Formic acid	1	2	
Drain, wash and drain. Horse overnight. Toggle dry				

water at 35°C, 3% sulfated fatliquor, and 3% sulfited fatliquor for two hours, before being fixed with 1% formic acid. The leathers were horsed overnight and then toggle dried.

Physical Testing

Sampling and sample preparation

The samples were cut as per the standard official method specified under IUP 2 (2001) in triplicates, both transversely and longitudinally towards the backbone, for all the physical tests. The samples were then conditioned at a standard atmosphere of 20 ± 2°C temperature and 65 ± 4% relative humidity for more than 24 hours according to IUP 3 (2001).

Determination of shrinkage temperature

Shrinkage temperature was determined in accordance with IUP 16 (2001). The leather samples were cut into strips of 50 mm by 2 mm for parallel and perpendicular specimens and suspended over a heating media in a water jar. The temperature at which significant shrinkage was seen was recorded as the leather's shrinkage temperature.

Mechanical analysis of the tanned leathers

All tests were carried out in triplicate for both the parallel and perpendicular runs. Tensile strength was determined using an Instron machine 1026 in accordance with IUP 6 (2001). The samples were clamped on a pair of jaws separated at a height of 50 mm. The maximum force exerted to break the specimens was recorded as the tensile strength and expressed in N/mm^2. Tear strength was recorded as per IUP 8 (2001) on an Instron machine 1026. Specimen samples of 50 mm by 25 mm were clamped on a pair of holders secured on the machine. The highest force exerted to cause tear was recorded as the tear strength and expressed in Newtons (N). Ball burst extension was demonstrated using disc-shaped samples of 44.5 mm diameter. The samples were placed on a machine with the grain side up and the flesh side touching the tip of the steel rod. Bursting and cracking of the grain surface were noted and expressed in millimeters (mm). A bally flexometer machine was used to test the number of flexes the leather would endure before failure.

Results and Discussion

Tanning

This stage was preceded with beamhouse operations. Among other operations, hair, fats and flesh were removed to ready the material for tanning operation. Reliming was done to facilitate the opening up of the fibers and removal of any residual hair debris left on the skins. Glutaraldehyde was used as a pretanning agent before vegetable tanning to help aid in more firm leather as well as act as a penetrating agent to better the tanning process. On treating the pre-tanned goatskins with extracts from the two different species of bananas and mimosa powder as the control tanning agent, the leathers obtained showed differences in their physical and organoleptic properties as shown in Figure 1 (a-f). The banana leaf midrib powders required a longer time to fully penetrate the skin matrix compared to the mimosa powder. This difference in penetration time could be due to various factors, such as the varying molecular size of the tannins, tanning strength of the tannins (which were 1.61, 1.82 and 2.07 for 'Muraru' (AA genome), *Musa sapentium Linn.* and mimosa powders, respectively) and pH of the tanning solutions used, as well as differences in the chemical structure of the tannins themselves.[21]

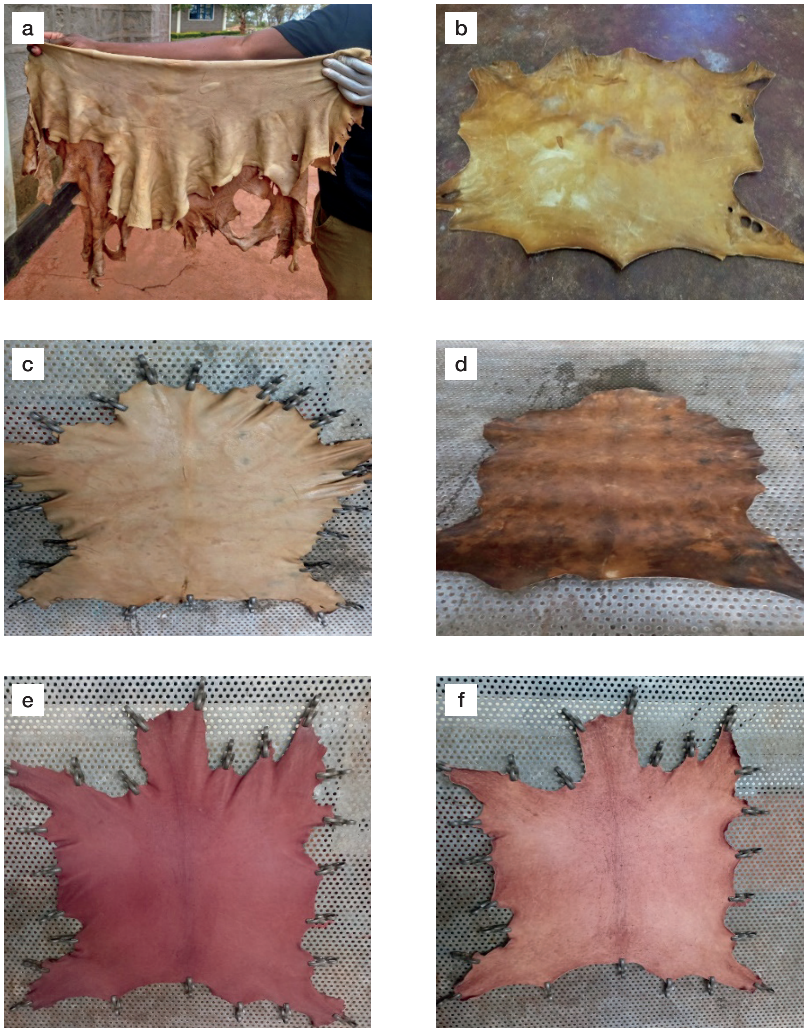

Figure 1. Leathers obtained from different tannages:
Muraru (AA genome)-tanned wet **(a)** and dry **(b)**;
Musa sapentium Linn.-tanned wet **(c)** and dry **(d)**;
mimosa-tanned wet **(e)** and dry **(f)** leathers

The wet and dried leathers showed color variations. This could be attributed to the aromatic compounds in the tannins that cause them to change color when exposed to light due to the formation of quinones on the tannins.[24]

The formation of free radicals causes rearrangement of chemical bonds and coupling of oxidized molecules, resulting in the formation of polymeric materials. If this process leads to the formation or linking of chromophoric groups, then color develops.[6] However, hydrolysable tannins have chromophores that cannot be linked together because they are located too far apart within the molecule. This results in the tannins being resistant to reddening and they are, therefore, considered to be light-fast.[25] The selected banana leaf midrib tanning agents are of condensed type,[21] and hence are affected by this effect because in the flavonoid structure, the aromatic nuclei are close together, making it easier for free radical oxidative bond rearrangements to occur.[6] This made the surfaces of the leathers quickly change their color. As a result of these tannins, reddening can be seen within Figure 1(b) and 1(d).

Physical Properties of the tanned leathers

The mean values of the physical tests of the leathers tanned with *Musa sapentium Linn.*, 'Muraru' (AA genome) and the control commercial tanning agent mimosa are as shown in Table III below.

Shrinkage Temperature

The mimosa-, *Musa sapentium Linn.*- and 'Muraru' (AA genome)-tanned leathers had values of 81.67 ± 0.94°C, 80.33 ± 0.94°C% and 78.67 ± 0.47°C, respectively. All the tanned leathers had shrinkage temperatures above the accepted value of 75°C.[26] Shrinkage temperatures above the minimum recommended value imply that the organic agent and the collagen matrix have formed strong

Table III

Physical Properties of Selected banana leaf midrib tanned leathers and Mimosa tanned leather

Physical properties		Mimosa	*Musa sapentium Linn.*	'Muraru' (AA genome)	Minimum recommended value
Shrinkage temperature (°C)		81.67 ± 0.94	80.33 ± 0.94	78.67 ± 0.47	>75
Tensile strength (N/mm2)	↑	16.54 ± 0.79	15.79 ± 0.31	20.54 ± 0.88	>12
	→	15.21 ± 0.46	13.16 ± 0.46	17.19 ± 0.41	
Tear strength (N/mm)	↑	51.61 ± 0.62	46.66 ± 0.95	40.37 ± 0.90	>20
	→	52.82 ± 0.39	59.16 ± 0.26	45.29 ± 0.74	
Percent elongation (%)	↑	43.53 ± 3.18	50.60 ± 2.95	61.10 ± 2.14	>40
	→	38.13 ± 0.74	32.67 ± 3.37	60.00 ± 2.29	
Ball burst extension (mm)	Grain crack	7.57 ± 0.22	6.86 ± 0.18	8.19 ± 0.33	6.50
	Ball Burst	8.72 ± 0.19	7.82 ± 0.46	10.22 ± 0.46	7.00
Thickness (mm)		2.10 ± 0.60	0.93 ± 0.05	0.77 ± 0.05	0.50
Flexing endurance (flexes)		No damage @50,000	Damage @30,000	No damage @50,000	50,000

crosslinks.[23] The selected banana midrib-tanned leathers had higher values in comparison with studies done by Maryati *et al*., (2020) on banana bunch tanning of rabbits skins, where the shrinkage temperature was found to be 52.47 ± 4.27°C.[27] Mimosa-tanned leather shrinkage temperature was close with studies done by Tanui *et al*., (2019), where it was reported that mimosa-tanned rabbit skins to have a shrinkage temperature of 83°C.[28]

Tensile Strength

Longitudinally, 'Muraru'-tanned leather had the highest tensile strength of 20.54 ± 0.88 N/mm^2, while *Musa sapentium Linn*.-tanned leather had the lowest value at 15.79 ± 0.31 N/mm^2. A significant difference ($p < .05$) in tensile strength was found longitudinally between 'Muraru' (AA genome)-tanned leather and mimosa-tanned leather, however no significant difference ($p > .05$) was observed between *Musa sapentium Linn*.-tanned leather and mimosa-tanned leather. Transversely, 'Muraru'-tanned leather had a tensile strength of 17.19 ± 0.41 N/mm^2, while *Musa sapentium Linn*. tanned leather had 13.16 ± 0.46 N/mm^2. No significant difference ($p > .05$) was found between 'Muraru'-tanned leather and mimosa-tanned leather, or between *Musa sapentium Linn*. and mimosa-tanned leather. All three tanning agents used exceeded the minimum recommended value of 12 N/mm^2.[26] The study findings indicate that samples cut perpendicular to the backbone had lower strength compared with those cut longitudinally and this is as a result of greater damage to the fibers resulting from increased friction, which occurs due to their alignment.[28]

The strength of leather is determined by the modification of the collagen fibrils caused by crosslinking with tanning agents. It is defined as the maximum amount of stress it can endure in the longitudinal direction without breaking.[9] Little has been done on tannin of goatskins with banana tannins. These values were lower compared to studies carried out using kepok banana bunch tannins, where the tensile strength of the tanned rabbit skins was found to be 29.61 N/mm^2.[19] The variation in tensile strength between the selected banana species tanning agents was associated with the variation in thickness which occurred as a result of an increased plumping effect in the *Musa sapentium Linn*.-tanned leather due to the low acid and salt content of its tanning liquor.[9] This swelling effect also increases the weave angle due to the increased distance between the collagen fibers reducing the load transfer hence resulting to lower strength.[29] The obtained values for mimosa-tanned leather closely matched with literature values.[2,30] It is worth noting that tensile strength is greater near the backbone compared to further away from it, which can be attributed to the way the fibers are arranged and packed.[31]

Tear strength

Tear strength values for specimens cut parallel and perpendicular to the backbone of mimosa-, *Musa sapientium Linn*.- and 'Muraru' (AA genome)-tanned leathers were found to be significantly different ($p < .05$). Parallel specimens of mimosa, *Musa sapientium L.* and 'Muraru' (AA genome) tanned leathers had values of 51.61 ± 0.62, 46.66 ± 0.95 and 40.37 ± 0.90 N/mm respectively. Perpendicular specimens had values of 52.82 ± 0.39, 59.16 ± 0.26 and 45.29 ±

0.74 N/mm respectively. Likewise, for samples cut perpendicular to the backbone, there was a statistical difference in all the tanned leathers ($p < .05$). All of the vegetable-tanned leathers examined in this investigation had tearing strengths greater than 20 N.[26] Studies done by Oliveira *et al.,* reported similar results for tear strength (58.9 N) for goatskins.[32] According to research done in Kenya using Acacia tannin extracts, goatskin samples of *Acacia nilotica* had 42 N, *Hagenia abyssinica* had 31 N, *Acacia xanthophloea* had 34.92 N, and *Acacia mearnsii* extract had 34.22 N.[9]

Grain crack and Ball burst

Grain crack values for mimosa, 'Muraru' and *Musa sapentium Linn*. tanned leathers were 7.57 ± 0.22 mm, 8.19 ± 0.33 mm and 6.86 ± 0.18 mm respectively. Ball burst values for mimosa, 'Muraru' and *Musa sapentium Linn.* -tanned leathers were 8.72 ± 0.19 mm, 10.22 ± 0.46mm and 7.82 ± 0.46 mm respectively. Grain crack comparisons between mimosa, 'Muraru' (AA genome) and *Musa sapentium Linn.* -tanned leathers revealed no statistically significant difference between mimosa and 'Muraru' ($p > .05$). However, there was a statistically significant difference between mimosa- and *Musa sapentium Linn.*-tanned leathers ($p < .05$). Ball burst comparison between mimosa-, 'Muraru'- and *Musa sapentium Linn.*-tanned leathers showed no significant difference ($p > .05$). The ball burst test is an important measure of the strength of upper leathers in shoe manufacturing, expressed in millimeters. It assesses the leather's resistance to cracking when it is lasted during shoe production. Grain crack and ball burst minimum recommended values are 6.5 mm and 7.0 mm, respectively.[26] Studies done in Kenya on *Plectranthus barbatus* indicated that mimosa-tanned leather had 7.7 mm and 8.2 mm values for grain crack and ball burst respectively, this did not differ much with our findings.[9] This was also in agreement with a study conducted using coffee pulp tannins, which reported 7.47 ± 0.09 mm and 8.25 ± 0.15 mm for grain crack and ball burst for mimosa-tanned leathers.[31] The greater the softness in the leather is attained the higher the busting height.[33]

Flexing endurance

Leathers from mimosa and 'Muraru' (AA genome) tannins had the highest flexion and bending capabilities, withstanding up to 50,000 flexes. Leather tanned from *Musa sapentium Linn*., however, was not able to withstand the same amount of flexes. It could only be able to hold up to 30,000 flexes. The failure was attributed to the firmness of the leather, hence being rigid in undergoing flexing. *Musa sapentium Linn.*-tanned leather may be used to make leathers for harness and saddlery and furniture as the leathers do not undergo much flexing during use with a minimum recommended value of 20,000 flexes.[26] Flexing endurance is carried out to tests finishes on leather surfaces as well as a fastness test in light leathers.[34] It is an assessment of leather's crack resistance during continuous flexing.[35] As shoes require materials with high and compatible flexion and bending capabilities, leather materials are subjected to this test to determine if it is suitable for this application. Research carried out on three types of Nigerian goats that were tanned using *Acacia nilotica* tannins did not reveal any significant variation in flexibility. Out of all the samples taken, only 5.83% displayed surface cracks on the leather.[36] Studies conducted on leathers that were tanned using Sunt pods, pomegranate husk, and mango leaves found that the leathers could endure up to 30,000 flexes without developing any cracks.[30]

Conclusion

The research findings from this study show that tannins derived from banana midribs can be employed as a substitute tanning agent for goatskins. Results showed statistical differences in some of the physical properties assessed, yet all the tests yielded values that surpassed the minimum set requirements for good quality leathers except for flexing endurance where *Musa sapentium Linn.*-tanned leather could only be able to hold up to 30,000 flexes. It was noted that leather samples cut along the backbone generally had superior properties as opposed to those cut across the backbone. This difference is thought to be due to the anisotropic nature of the hide and skin structure. As such, selected banana midrib tannins can be used to produce light vegetable-tanned leathers. To expand on these findings, future investigations should explore the use of banana leaf midrib tanning on other raw materials, such as sheep and exotic skins, either alone or in combined tannages.

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