Research Advances in Oil Tanning Technology: A Review

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Abstract

The conventional process of chamois leather manufacturing for industrial applications utilizes fish oil which contains substantial amounts of pentadienoic fatty acid. The applications of chamois leathers include cleaning polished surfaces, manufacture of gloves and orthopedic uses. However, due to fish oil's strong odor and high cost, considerable efforts have been made to counter these challenges. Esterification of fish oil has been used as a strategy to address the problem of odor and water absorption but this cannot solve the issue of cost. Oils from plant sources such as linseed, rubber, jatropha, castor and sunflower have been investigated as potential tanning substitutes for fish oil. Linseed has been found to produce chamois leather with mild odor and water absorption characteristics close to those of fish oil compared to other oils obtained from plant sources. Oil from goat fleshing has also been investigated in chamois leather production and has been found to produce chamois leather whose odor compares with that of linseed oil tanned leather. If these tanning oil alternatives are combined with other research advances in chamois leather production such as glutaraldehyde pre-tanning and oxidation using hydrogen peroxide or through ozonation, then the issue of cost, odor and long oxidation period can be resolved.

Introduction

Leather is one of the most widely traded commodities in the world, giving the leather industry an estimated global trade value of $414 billion per year.[1] It is generally processed in three steps, with the first step being pre-tanning. Pre-tanning involves soaking, liming and unhairing, deliming, bating, pickling and degreasing to remove unwanted components, hair, adipose tissue, fats, etc., leaving behind a network of fiber protein.[2] The next step, tanning, involves reacting the pre-tanned material with suitable agents to produce a stabilized fiber structure.[3] This can be achieved through chrome, vegetable, combined tanning or oil tannage. Finally, post-tanning - which involves neutralization, retanning, dyeing, fatliquoring and finishing in order to improve fiber characteristics and surface - is carried out. Finishing enhances color, softness, and lubrication of the leather surface.[4]

Tanning converts perishable organic material (animal skin) into a stable material (leather), which is resistant to spoilage by bacteria.[5] The skin is transformed into a non-putrescible matrix with increased hydrothermal stability via new cross-links that achieve additional dimensional stability within the matrix.[6] The most common inorganic tanning agents are chromium (III), zirconium (IV), aluminum (III), titanium and Iron.[7] Basic salts of these agents form coordinate bonds with the fiber structure and facilitates the stabilization of the protein structure.[8] Reactions take place at the carboxyl group of the collagen.[2] On the other hand, organic tanning agents are carbon and hydrogen containing compounds which have the ability to stabilize collagen within the skin. They include vegetable tannins, synthetic tanning agents (syntans), aldehydes and oils.[8] This review is focused on assessing the recent advances in oil tannage. These advances can be utilized by manufacturers to address challenges such as odor and cost faced by the chamois leather industry.

Applications of Oils and Fats in Leather Industry

Leather industry has found a wide range of applications for oils and fats. Rubber oil,[9] epoxidized oil,[10] and recovered olive oil[11] are examples of oils that have been used in oil tannage. Fatliquor-cum-filler production from fleshing waste for re-tanning has been studied.[12] The leather is fatliqoured to prevent fiber sticking when the leather is dried after completion of the wet processes.[13] Other studies on fatliquoring using seal hides oil, limed fleshing oil, synthetic, sulphited and sulphonated fatliqours have been reported.[14-17] In addition, oil has been reported as a potential formaldehyde scavenger.[18] The authors reported that use of 2% *Origanum onites* oil in the fatliquoring process substantially reduced the free formaldehyde formed in the process and offered a solution of potential importance to the leather industry.

Although the grammage of the garment leathers is directly proportional to the amount of fatliquoring agent, the drapability coefficient, stiffness and bending rigidity values also decreases with increase in amount of fatliquor.[11,19] Consequently, softness and drapability properties of garment leathers are greatly improved through fatliquoring. This indicates that leather manufacture can be influenced by the amount of oil used based on its application. Oil tannage yields low density and soft chamois leather, whereas fatliquoring is used to impart drapability in leathers tanned using other tanning technologies such as chrome tanning.

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However, it should be noted that other studies to assess the potential applications of such oils outside the leather industry have been carried out. Kolomaznik and co-workers experimented and modeled the potential of tannery fleshing waste as source of biodiesel.[20] The study proposed that oils from fleshing waste can be combined with oils from plants, such as used or waste cooking oils, for biodiesel production. Leather solid waste has also been investigated as a source of biodiesel.[21] The waste mainly originated from pre-fleshing, fleshing, shaving and trimmings, which were found to contain high amounts of fat contents. These studies point to a possibility of competition for tannery fleshing oils. Nonetheless, tannery fleshing waste is a serious challenge faced by the leather industry and any potential use for the waste will be highly welcome. Furthermore, an understanding of the various usages will spur more studies aimed at optimizing the applications of these oils.

Sources of Oils for Leather Industry

Chamois leather is popular in the leather market due to its unique properties. The process of chamois leather production conventionally relies on fish oil for its survival.[9] Attempts to come up with alternative sources of oil have been made, though their usage in the process is not adopted by the industry. Jatropha oil,[22] tannery fleshing oil,[23] linseed, castor and sunflower oils[24] have been experimented in chamois leather manufacture. Other applications such as fatliquoring that make use of seal-hide oil,[14] and oils from fleshing waste for re-tanning process[12] have been reported in literature. The modeling of fleshing oil production[120] and the practical extraction methods for these oils are also well documented.[25,26]

Chemical Characteristics of Tannage Oils

Studies on the chemical characterization of oils used in leather industry have been carried out and results are summarized in Table I below.[23,24] Iodine value is an indicator of the average degree of unsaturation (alkene group presence) within lipids while acid value shows the amount of free fatty acids present in fats or oils. Acid value is the amount of potassium hydroxide required to neutralize one gram of a fat or oil sample.[27] Saponification value indicates the average molecular weight of the triacylglycerol in a sample.

Acid values for the investigated oils average between 3 - 7 mg/g except for fish oil with a value of 15 mg/g. Despite most of the values being less than half that of fish oil, the studies have shown linseed and goat fleshing oils are promising oil tannage alternatives. However, the researchers did not provide details on animal tallow used and, therefore, the provided acidity value may not be compared to those of other oils. On the other hand, saponification values for all the oils are comparable, ranging between 174 - 188 mg/g.

Interestingly, among the characterized and investigated oils as substitutes to fish oil, linseed and goat oils are the best options for chamois leather manufacture on the basis of the organoleptic properties of the leathers produced (Table II). The organoleptic properties of the chamois leathers obtained using the two oils as tanning agents are of internationally acceptable standards.[30,31]

Characteristics of Oil Tanned Leathers

Various tests are carried out in order to grade chamois leathers. Water absorption, shrinkage temperature and organoleptic properties are some of the parameters that are used for this purpose. Table II compares the properties of leathers produced via oil tannage.[23,24]

Although not all data is available on the different oils for comparison purposes, it is useful to compare the available information. For example, linseed and goat oil produce chamois leathers with organoleptic properties that are comparable to those of fish oil except for odor. Water absorption and other features for the leathers obtained from the two oils are also within international standards.

A further comparison, based on the chemical characteristics of various oils presented in Table I and the properties of chamois leathers obtained from the oils (Table II) above, is also imperative. It is noteworthy that the top three oils (fish, linseed and goat) produce high quality chamois leathers. However, odor is rated poorly for fish oil-tanned chamois leathers. One outstanding disparity among the three oils is the acid value, which could be responsible for odor. Fish oil has acid value of 15 mg/g, way above the recommended value of less than 8.6 mg/g.[32] Conversion of fish oil into corresponding esters

Table I

Chemical characteristics of tannage oils

Parameter \ Oil	Fish	Linseed	Castor	Sunflower	Animal tallow	Goat
Iodine value (mg/100 mg)	135	160	80	120	160	71
Acid value (mg/g)	15	7	4	6	3	7
Saponification value (mg/g)	180	188	176	188	174	186

Table II
Properties of oil-tanned leathers

Parameter	Fish oil	Linseed oil	Castor oil	Sunflower oil	Animal tallow	Goat oil
Water absorption	320 ± 5	303 ± 5	195 ± 5	197 ± 5	190 ± 5	211
Tongue tear	66.7 ± 0.5	55.0 ± 0.5	41.0 ± 0.5	39.0 ± 0.5	40.0 ± 0.5	-
Stitch tear	199.8 ± 0.5	154 ± 0.5	115.3 ± 0.5	109.3 ± 0.5	120.2 ± 0.5	-
Shrinkage temperature	83 ± 1	79 ± 1	70 ± 1	67 ± 1	72 ± 1	70
Sink test	120 ± 5	130 ± 5	155 ± 5	167 ± 5	158 ± 5	-
Softness	7	6	4	3	4	8
Fullness	7	7	5	4	5	-
General appearance	8	7	6	5	6	-
Odor	2	8	8	8	3	8

has been shown to help solve this problem.[33] Generally, esters are known to have sweet smell[34] and this may explain why esterification suppresses odor in fish oil tanned chamois leather. This may lead to the inference that acid value of an oil is inversely proportional to odor of chamois leather produced. However, more research needs to be done to validate this claim; as pointed out earlier, details of animal tallow used in Table II are not available and therefore the low odor rating (given as 3) cannot be used to conclusively establish a correlation.

Oil Tannage

Oil tannage is the method of making highly porous chamois leather.[8,32] The leathers are best known for properties of absorbing water, low density, softness and flexibility.[35] Chamois leathers are also highly resistant to the action of water at ordinary temperature.[36] They have found application in the manufacture of gloves, cleaning and drying of polished surfaces and in the production of orthopedic goods.[37] Different oils with tanning properties have been investigated for chamois leather production.[9,24,33] Characteristic tanning oil contains about 18 to 20 carbon atoms and about 4 to 8 unsaturated double bonds. The oil should have high iodine value of about 75 - 120 mg/g, a low acid value below 8.6 mg/g and it should not be rancid (Hualong *et al.,* 2011).[32]

The conventional oil-tanning process is based on the oxidation of cod, fish or sardine oils, which have numerous unsaturated bonds.[38] The unsaturated free fatty acids combine with oxygen to form oxidized form of fatty acids, aldehydes and peroxides which effect the tanning action on the pelt.[39] The level of unsaturation is imperative since little unsaturation in the oil will not oxidize promptly and, therefore, will work as an ointment whereas excessive unsaturation causes the oil to crosslink with itself and solidify upon oxidation. The tannage is alluded to an aldehydic reaction since the procedure is characterised by the release of acrolein, CH_2=CHCHO, and polymerization of the oil; the availability of latter could represent the variations between the qualities of oil and aldehyde-tanned leather.[32]

Traditional oil-tanning method

The traditional method of making chamois leather involved the impregnation of the skin with cod oil in filling stock and then allow oxidation of oil to occur, with the products of the reaction having a tanning reaction.[9] This procedure is repeated until the point when pleasant leathering is accomplished. Surplus oil from the skin is expelled by pressure driven squeezing after washing in warm soluble water. The skin is then hung to dry and wrapped up.[40]

Modern oil-tanning method

The principle of modern oil tannage is to oxidize the fish oil, already introduced to the deemed pelt, with the help of atmospheric oxygen under controlled conditions. As the tanning agent, unsaturated glycerides like cod oil and fish oil are used.[41] These fatty acids having up to six double bonds in the aliphatic chain, give the necessary reaction products from oxidation and polymerization to give the characteristic chamois leathering effect under normal condition of tanning.[36]

Challenges and Advances in Oil Tannage

Time of oxidation and toxicity are some of the initial challenges that face the manufacture of chamois leather. Traditionally, tanners used to impregnate skins or splits with fish oil and then left them to hang for up to 14 days to allow for air oxidation.[39] These long periods of oxidation made many tanners to shift to other methods of leather manufacture such as chrome tanning. However, mild pre-tanning operation using formaldehyde[42] in an attempt to address this issue has since been abandoned due health reasons.[43] Glutaraldehyde has been espoused in its place. In addition, hydrogen peroxide, epoxidized oils, benzoyl peroxide or ozonation reactions have been studied as a way of accelerating the oil-tanning process.[39,42,44,45] These advances in oil tannage have reduced the time significantly to the point that oil tanning can now almost rival the other techniques of tanning.[23]

Cost and odor, especially for fish oil, are other factors that have hindered chamois leather manufacture. Concerted efforts have been made to address these problems. As already pointed out earlier, the use of linseed oil and oil from goat fleshing waste have been investigated and found to produce chamois leather that meets recommended standards. Nonetheless, linseed oil may face the challenge of cost due to its numerous uses.[46] Driven by competing demands for linseed oil, availability of the oil for tanning will not be assured. And even if it will be available, price will be driven by demand. However, oil from tannery fleshing waste can be an appealing alternative because its utilization will be a part of waste handling. Tanneries are currently grappling with fleshing waste management.[47-49] The waste produces foul smell and also piles up to the extent of minimizing available space within the tanneries or dumpsites. Thus, any alternative way of exploiting this waste will free the dumping space and yield additional income from the utilization of the extracted oil. Such value addition strategies will encourage adoption of this alternative source of oil. Countries such as India will greatly benefit from the advances in this field.[50] Availability of goat skins in India is very high and research has shown that these can be a source of fleshing waste for extraction of tanning oil.

Conclusion

With all the recent advances in oil tannage, it is possible to come up with an optimal combination of these innovations and produce chamois leather within reduced time and cost, and with desirable organoleptic and other physical features. Linseed oil and oil from tannery fleshing operations have been presented as potential alternatives to fish oil. However, linseed has competing applications and this might choke its supply. Coupled with glutaraldehyde pre-tanning and oxidizing agents such as hydrogen peroxide, these advances are likely to address the challenges in chamois leather manufacturing. More research needs to be carried out to actualize this at tannery level.

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