Original Research

Characterization of Leather Industry Wastes

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Abstract

In this research samples from solid wastes coming out from various process steps during manufacturing of garment sheep skins, shoe upper goat skins, shoe upper hides and sole leathers have been collected. These samples have been classified among themselves and their characteristics have been determined by various chemical analysis. By characterization of solid wastes which state problems for the environment and costs for treatment; data have been obtained which can be used for new management and evaluation methods.

Keywords: leather, hide, skin, solid waste, environment

Introduction

The principal aim of the leather industry, which plays a significant role in today's global economy, is to transform animal hides/skins into a physically and chemically stable material by subjecting them to chemical and mechanical sequential processes, and therefore to obtain products for meeting various needs of people. The leather industry generally uses hides and skins as raw materials, which are the by-products of meat and meat products industry. In this respect, the leather industry could have easily been distinguished as an environmentally friendly industry, since it processes waste products from meat production [1]. However, the leather industry has commonly been associated with high pollution due to the bad smell, organic wastes and high water consumption caused during traditional manufacturing processes [2]. Different forms of waste in quality and quantity, which emerge during the transformation of hides and skins into leathers in thousands of leather factories, from primitive to modern all around the world, have negative impacts on the environment.

Although the characterization of solid wastes from the tanning industry is well documented, our study investigates the properties of the solid wastes generated from different types of leather – producing tanneries, i.e. garment, shoe upper, sole, etc. As it is known, producing different types of leather requires different types of processes and chemical usage. Consequently, leather wastes generated from each type of leather and process have different characteristics. According to us, for the utilization of these wastes in various fields, having more specific information about their characteristics has great importance. In this direction, in this research an overview of leather processing is mentioned and wastes coming out of various factories and process steps are characterized with the aim of obtaining data for evaluating them.

According to the data received from the studies of several researchers, approximately 200 kg of leather is manufactured from 1 tone of wet-salted hide [1-3]. This amount constitutes about 20% of rawhide weight. More than 600 kg of solid waste is generated during the transformation of rawhide into leather. That is to say, solid wastes containing protein and fat that constitute more than 60% of rawhide weight are disposed to the environment by leather factories without turning them to good use (Table 1).

In other words, besides the 30-35m^3 waste water disposed to environment during the processing of every 1 ton of rawhide in world leather industry, the data from FAO reveals that approximately 8.5 million tons of solid waste is generated during the production of 11 million tons of rawhide processed in the world [4].

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Table 1: Estimated amount of solid (protein, tanned and untanned) waste during the processing of 1 ton of salted hides according to various authors

	Püntener	Alexander	Buljan
Untanned waste:			
shavings			
subepidermal Tissue	530 kg	120 kg	100 kg
trimmings	135 kg	70-230 kg	300kg
Tanned Waste:			
shavings	145 kg	100 kg	99 kg
split		115	107
Dyed and finished waste:			
shavings	10 kg	32 kg	10 kg
fluff		2 kg	1 kg
Total	870 kg	439-599 kg	637 kg

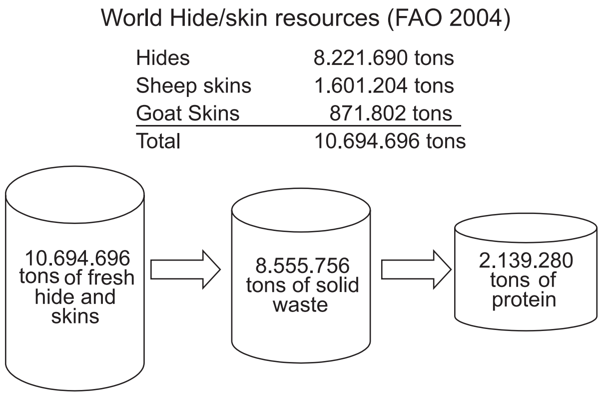

Fig. 1. Amounts of leather industry raw material input and solid waste output.

It is possible to divide leather industry processes of transforming rawhide into leather into four main stages:

i. Beamhouse processes:
The conserved hides are first subjected to a *trimming* process for removing the unwanted parts, and then they are *soaked* to restore the lost water and to remove substances like dirt, blood and conservation salt. After the wetted hides are *fleshed* to remove the excess flesh and fat adhering to the hide (hypodermis), they are treated with an intense alkali solution of lime ($Ca(OH)_2$) and sodium sulphide (Na_2S) to ensure hair and wool removal (*unhairing process*). Later the hides are swelled up in liming process by immersing them in a strong alkaline bath so as to open up the collagen structure. The hides may be treated with a second fleshing process after liming in order to clean the flesh. At this stage, hides are treated with *splitting process* and split into two or three layers. *Deliming* is then performed to decrease the pH level in order to remove the lime and to make the hide more receptive to the chemicals that will be used in further stages. Through *bating process*, hides are exposed to an enzymatic effect for both opening up the structures, and the removal of unwanted proteins from the hide. Following the bating process, a *degreasing process* is applied to hides for removing the excess natural fat in their structure and providing a homogeneous distribution of the fat in it.

ii. Tanning Processes:
The hides at this stage are first treated with *pickle process* in a solution composed of salt and acids so as to obtain a homogeneous distribution of tanning materials through the cut. After the hides are conditioned as above, the *tanning process* is applied with various tanning materials (materials able to form stable bonds with collagen) in order to provide the leather with a stable form and high thermal stability. Tanning materials such as vegetable tannins, mineral tanning materials and syntans (synthetic organic tanning materials) are used in tannage. Among mineral tanning materials, chrome is the most widely used in leather production due to its unique features that it gives to the leather. Aluminium and vegetable tanning materials are also widely used in leather production.
Before the leathers are treated with further processes, the *setting out and samming process* is applied, and *shaving* is done to obtain the desired thickness of the leather.

iii. Post-Tanning Processes:
The next step for the leathers, which are tanned and standardized to a desired thickness, is *retannage process* with various retanning agents improving the requested characteristics of products. In this process, structural differences within leathers are compensated to obtain uniform structure. The *Fatliquoring process* is applied by using a combination of various fatliquoring agents in order to allow the leather to be more supple and softer. In the *dyeing process* leathers are dyed to the desired color. After this stage, leathers are hanged and dried, and they are prepared for the finishing process through certain mechanical operations. The unwanted parts are trimmed and removed.

iv. Finishing Processes:
After the leathers are fatliquored and dyed following the tanning process, they are processed with a series of coatings on the surface in order to improve their resistance and produce appealing and uniform surface effects. After this process, leathers are *trimmed* for a final form and sent to confection.

Solid wastes generated by the leather industry in these stages of processes may be classified as follows:

i. wastes from untanned hides/skins (trimmings, fleshing wastes)
ii. wastes from tanned leather (shaving wastes, buffing dust)
iii. wastes from dyed and finished leather (trimmings from leather)

Data obtained from research reveals that 80% of solid wastes are generated during pre-tanning processes, while 20% of the wastes are caused by post-tanning processes

[5]. Due to the bad smell they produce during their putrefaction and their harmful chemical content, untanned hide/skin wastes have negative effects on the soil and/or water resources of the environment where they are discharged, in other words on the local plant flora and animal fauna. Therefore, uncontrolled discharge of such wastes should be prevented without taking adequate precautions. Legal arrangements gradually gaining speed all over the world enforce the leather industry to apply innovations in terms of reusing solid wastes generated during leather production processes such as fleshing, shaving, trimming and splits.

The most significant approach in preventing environmental pollution is the idea that prevention is better than reuse, reuse is better than recycling, and recycling is better than disposing of the wastes [6]. In other words, cleaner production. On this account, in order to provide cleaner production, the producers are supposed to prevent or reduce waste formation by using clean technology during production processes, and transform the inevitable small amounts of waste into environmentally friendly materials.

Solid wastes create a major problem for leather industry in terms of both their variety and quantity. A high amount of reusable waste is generated in the leather industry. It is possible to recycle these products and even use them as raw materials for different industries [7]. The variety and quantity of solid wastes depends on animal species, breeding conditions, slaughterhouse practices, conservation conditions, leather process stages, mechanical operations, qualification of the personnel, and chemicals used in processes. Yet this fact causes uncertainties in reusing the generated wastes.

First of all, solid wastes should be characterized so that they can be reused. In this study, wastes from different companies have been analyzed with various chemical and instrumental analysis methods, and their characteristics have been defined. This data is thought to be useful in terms of preventing both environmental pollution and waste of resources by putting solid wastes into good use as secondary raw material in different industries rather than transferring them to disposal areas.

Materials and Methods

Materials

Samples of wastes from pre-fleshing, lime fleshing, shaving, buffing and trimming, which were examined within the scope of the study, had been obtained from various factories producing different leather types (clothing sheep skin, shoe upper goat skin, shoe upper hide and sole cattle hide). Samples are coded as follows:

A: Sheep skin
B: Goat skin
C: Hide (shoe upper)
D: Hide (sole)

1: Pre-fleshing waste
2: Lime fleshing waste
3: Shaving waste
4: Buffing waste
5: Leather trimmings

Example: A1: Sheep skin pre-fleshing waste.

Method

Solid waste samples examined for the study were randomly selected among piles in 2 different leather factories manufacturing products for each leather type. Pre-fleshing and lime fleshing wastes, which are defined as untanned wastes, were scrapped after the water in their structure was removed (to a certain extent) in airy conditions, and then they were equally mixed and dried at 50-55°C. Shaving, buffing and leather trimmings from tanned leather wastes were ground and mixed before they were dried at 102°C ± 2°C and prepared for chemical analyses. All other analyses (excluding water content measurement) were done by taking sufficient amounts from these samples.

The analysis were carried out using the following standard methods: The amount of water content (volatile components) according to IUC 5 [8], pH according to SLC 13 [8], Substances (fats and other substances) soluble in dichloromethane according to SLC 4 [8]**,** Nitrogen content according to Kjeldahl Method, Sodium chloride content according to SLC 402 [8], Sulphide content according to Monier-Williams Method.

Thermal value of the samples were measured in Bertholet calorimeter under 23-30 atmosphere pressure by burning with electric current in an aerobic environment and monitoring the temperature increase of a certain amount of water by the generated heat.

In order to determine Cr, Fe, Na, and Ca in waste samples using an atomic absorption spectrophotometer, the samples were first treated with an acid digestion process. Sample solutions were filled up to 100 ml with pure water after they reached room temperature (it may be filtered through filter paper if necessary). Desired measurements were later done in these obtained solutions by using atomic absorption.

Cr, Fe, Na and Ca amounts of wastes were detected with Perkin-Elmer 2380 Atomic Absorption Spectrophotometer. Cr was measured at 357.9nm with 0.7 slit intervals by using air+C_2H_2 mixture, Fe was measured at 248.3nm with 0.2 slit intervals by using air+C_2H_2 mixture, Na was measured at 589.6nm with 1.4 slit intervals by using air+propane mixture and Ca was measured at 422.7nm with 1.4 slit intervals by using air+C_2H_2 mixture.

Results and Discussion

Water Content

According to the results of water content determination in solid waste, water content was found to be

Table 2. Mean values of some characteristics of leather industry waste.

Type of Waste	Water Content %	pH	Fat&Oils %	Nitrogen %	Salt %	Suphur SO_2 (ppm)	Calorific Value kcal/kg
A_1	58.99	7.29	58.5	14.7	1.41	n.d.	7953
B_1	44.40	7.23	53.5	5.9	1.71	n.d.	7375
C_1	45.77	7.4	56	34.6	2.56	n.d.	5345
D_1	45.77	7.44	56	17.1	2.52	n.d.	5642
A_2	61.65	12.37	53	13.1	1.77	439	4852
B_2	83.72	10.06	25	25	0.87	243	4916
C_2	57.13	12.43	6	31.3	2.82	65	5753
D_2	57.13	12.49	7	18.2	2.86	n.d.	5146
A_3	66.01	3.93	2.4	60.3	4.65	n.d.	3884
B_3	63.99	4.28	0.85	61.2	3.95	n.d.	3912
C_3	46.61	4.17	2.15	64.1	4.68	n.d.	4354
D_3	45.86	5.24	2.1	31.7	1.72	n.d.	4552
A_4	14.58	4.3	4.05	35.4	2.00	n.d.	4820
B_4	8.57	5.81	3.5	46.5	1.60	n.d.	4902
C_4	6.97	6.01	3.9	47.7	1.66	n.d.	4056
D_4	9.26	4.26	4.1	26.4	1.37	n.d.	4544
A_5	7.24	5.47	6.95	49.4	1.49	n.d.	4685
B_5	6.39	4.46	4.35	48.6	1.48	n.d.	4929
C_5	8.83	4.75	7.05	46.7	2.21	n.d.	4744
D_5	8.83	4	4.25	28.7	2.08	n.d.	4541

45-59% in pre-fleshing waste, 57-83% in lime fleshing waste, 45-66% in shaving waste, 7-14% in buffing waste, and 6-9% in leather wastes. Sempere et al. [9] have observed that water content of leather waste (18-25%) is lower than the water content of shaving waste (45-57%).

The obtained data reveal that fleshing and shaving wastes include higher amounts of water in comparison to other waste types. This result is due to the fact that fleshing and shaving wastes are generated during leather process steps called beamhouse processes, in which leathers are processed in watery environments. Water content of solid wastes like buffing and trimming are lower, since they are generated during the stages of dry processes.

Water content of solid wastes differs to a great extent in accordance with how and where they are collected, storage conditions, and climate. Yet there is always an obvious water content difference between the wastes generated during wet and dry processes. Water content of solid wastes is of great significance at the time of storage and use.

Due to the high amount of water in leather wastes, or natural factors like rain or snow, infusion of the chemicals in leather waste into soil and surface/ground waters causes the deterioration of the natural structures of these receiver environments. Especially in the case of wastes including chrome, researchers have reported the infusion of chrome into plants and ground waters [9].

pH

It has been observed that solid wastes show similarities in parallel with the process stages disclosing the pH values of solid wastes, and the pH values at which these processes are carried out; and it was found that liming fleshing waste has the highest pH value, since the liming process is done at around pH 11-12.

Acid-base reactions are among the most important matters regarding the environment. Most of the living organisms are very sensitive to environmental pH. Another situation is that, when pH is forced to extreme limits, chemical balance reactions slide to the left or right, causing the occurrence of undesired sediments or gases. Hence, in order to protect the current ecosystem, wastes should be subjected to a neutralization process before they are disposed to the environment.

The Amount of Substances (Fats and Other Solubles) Soluble in Dichloromethane

Unless the fat in rawhide structure is removed, it affects other processes in a negative way by prohibiting the penetration of chemicals into the hide in further processes, and may also cause a strong smell, an increase in micro-organism activity and defects like spew on leathers. For this reason, vast amounts of fat in hides are removed before tanning by applying a degreasing process. Bienkiewicz [10] noted that fat content varies between 0.5% and 4% in cattle and horse hides, 3% and 30% in sheep skin, 4% and 40% in pig skin, and 3% and 10% in goat skin over dry weights.

Another fat source in solid wastes is the natural or synthetic fats used in the fatliquoring process in order to provide the leather with features like softness, elasticity and resistance, and to allow the leather fibers to slide easily over one another according to the leather type and purpose of use. The fat content of leathers consists of the remaining natural fat after degreasing process, and the fat used in fatliquoring process. Due to this reason, different fat proportions may be detected in leather wastes.

In the research, it has been observed that pre-fleshing and lime fleshing wastes from the processes before degreasing include higher amounts of fat compared to other waste samples (40-70%). In addition, it should be kept in mind that wastes were generated at the end of the fleshing process that was applied to remove the hypodermis layer of the hide, which is the layer including the highest amount of fat. It should also be taken into consideration that flesh and fat layers left on hides may differ greatly, depending on slaughter and flaying techniques, which in turn directly affects the fat amount in waste. This high amount of fat in fleshing waste may be evaluated in different fields such as the chemical industry, soap production, biodiesel production, and the production of fatliquors [7]. The fat obtained from lime fleshing waste is generally regarded to be of low quality due to its dirty colour and bad smell resulting from sulphide contact.

Nitrogen Amount in Wastes

Determination of nitrogen in solid wastes gives an idea about proteins as the main component of leather, and therefore about the effect of leather processes on proteins.

The results obtained from nitrogen determination analysis conducted with the Kjeldahl method in waste samples have revealed that fleshing wastes with 6-34% nitrogen have the lowest amount of nitrogen, and therefore the minimum protein content. As mentioned before, this result stems from the fact that fleshing wastes are the ones generated by the removal of a hypodermis layer, which is rich in fat and poor in protein.

Shaving wastes include the highest amount of nitrogen (64%). Chrome in these shaving wastes may be removed to a great extent by treating the samples with alkali or with the help of enzymes, and thus protein hydrolysates without chrome may be obtained (<5ppm). Therefore, it is possible to put shaving wastes into commercial use due to its wide area of use such as leatherboard, fertilizer or hydrolysate production [11, 12].

When other data were examined, nitrogen content of the wastes from sole leather measuring 26-31% was found to be lower than the nitrogen contents of other waste types. Since the rate of vegetable tanning substance in the waste (polyphenolic substance) increased, the nitrogen amount detected in leather decreased accordingly.

Sodium Chloride Content

When the salt contents of solid wastes were examined, it was found that salt proportions in most samples vary between 1% and 2.5%. As a matter of fact, it is remarkable that salt contents of shaving wastes from all skin types are higher than the average values of other samples, and this difference is thought to stem from the high rate of alkali salts used especially in basification at the end of the chrome tanning process; since this parallelism detected in shaving wastes is not observed in the shaving wastes from sole leather treated with vegetable tanning process.

The high content of salt in solid wastes causes an increase in the salt amount of the soil where these wastes are stored. Soluble salts have negative effects on plant growth, because the intake of nutritional elements by the plant is prohibited due to ion competition when the soil contains high amounts of cation and anions. This salt damage emerges because of the insufficiency of nutritional elements. Additionally, high salinity or total dissolved salts in water causes high osmotic pressure, which results in reduced water availability to plants and retarded plant growth of crops which are not salt tolerant [13].

Sulphide Content

As mentioned before, in beamhouse processes, according to the type of desired leather products, hides/skins are treated with an intense alkali solution and sodium sulphide for hair and wool removal and opening up the structure. In this step, disulphide bonds are destroyed by reduction under alkali conditions. Therefore, as is also seen from the results, the wastes coming out from these stages are distinguished from others by their high sulphide content. The highest amount of sulphide was detected in sheepskins (439 ppm), because these raw materials are generally used in production of garment-like soft leather types and in order to obtain required properties, intensive liming and unhairing is applied. Goat skins and hides used for production of shoe upper leathers are generally treated with less lime and sulphide in order to obtain firmer leathers, and so wastes occurring at these stages have less sulphide content.

Thermal Values

As presented in the graphics, due to their high content of fat, fleshing wastes with an average of 5872 kcal/kg thermal value have higher thermal values than that of other wastes with an average of 4493 kcal/kg.

The thermal values belonging to some urban wastes were determined to be 2770 and 4450 kcal/kg in paper, 6660-8885 kcal/kg in plastic, 3600-4440 kcal/kg in textile, 4990-6660 kcal/kg in rubber, and 3600-4730 kcal/kg in leather [14]. In some cases, leather wastes are exterminated by incineration due to their high calorific values (4500-5000 kcal/kg) [15]. Thermal treatment of leather scrap must be justified not only by its high calorific value, but by taking into account as much as possible of the consequences of it, namely (i) pollutants and their levels in the released gasses; (ii) characteristics of the ashes and factors that influence changes in the chromium oxidation state during the burning process as well as in some of the further ash treatment options. These considerations must always be directed to useful ends [16].

Determination of Cr, Fe, Na, Ca Ions in Leather Wastes

Some Ion amounts detected in solid wastes generated by the leather industry are introduced in the table below.

As displayed in the table, chrome was detected to be high in waste samples which are treated with chrome in the tanning process. The indicated Cr^{+3} content is the result of chrome used in tanning.

Chromium traces found in sole leather shaving and buffing wastes were associated with the contamination of wastes, since chrome is not used as the main tanning substance in sole leather production. An increase was observed in chrome contents of sole leather wastes, and therefore it was considered that waste samples treated with chrome retannage in the final stage for a specific purpose during sole leather production were evaluated among the samples. Naturally, Cr ion was not detected in fleshing wastes.

Chromium has two important oxidation states: trivalent (Cr^{+3}) and hexavalent (Cr^{+6}). Trivalent chromium is of low toxicity and is an essential trace ion necessary for several biological activities, whereas hexavalent chromium is of high toxicity [17, 18].Tannery wastes normally contain only trivalent chromium [10, 19, 20]. The production of chromium-containing solid wastes (including chrome shavings) in a tannery has been recognized as a problem for many years but recently pressure from environmental authorities has given the problem increasing urgency. As a result, many scientific groups have oriented their research to find a process to recycle these wastes. The chemical composition of chrome shavings make them suitable for processing to recover their constituents but the economics of the process is very important for industrial implementation [21]

On the other hand, about 90% of hides/skins in the world are still tanned with chrome, because other tanning materials fail to give the leather a high hydrothermal stability and other use properties that chrome provides.

Iron amounts in waste are generally not very high, and vary according to the iron content of the water used in processing, and the process after which the waste is generated. For example, the iron dust appearing during the sharpening of knives used in the fleshing process to remove the flesh side called hypodermis layer, or the iron dust generating from buffing paper during the buffing process may cause changes in determined values.

In the results concerning calcium (Ca) content, the high amount of calcium determined especially in fleshing wastes is caused by the lime used in the liming process applied for opening up the structure and conveying additional functional groups.

Sodium content in the wastes can be formed from many sources. It can either originated from sodium chloride or sodium sulphate. The amount of salts used in the production of various kinds of leather types and various processes differs in a wide range. For example, tanning processess carried out at low pH values or strong acidic

Table 3. Mean Values of Cr, Fe, Na and Ca Contents of Leather Industry Wastes (ppm).

Type of Waste	Cr	Fe	Na	Ca
A_1	n.d.	1206	27496	6972
B_1	n.d.	667	4014	19310
C_1	n.d.	915	14612	12185
D_1	n.d.	1407	27217	16424
A_2	n.d.	894	9000	76000
B_2	n.d.	1835	15000	87000
C_2	n.d.	4272	45384	11799
D_2	n.d.	626	29679	42399
A_3	14500	11	15000	700
B_3	14493	27	10243	745
C_3	17943	282	14234	711
D_3	43	378	5919	1566
A_4	11783	1467	49350	4632
B_4	23747	77	707	1516
C_4	15000	521	11000	6200
D_4	28	992	14894	2780
A_5	14989	34	2100	900
B_5	18360	123	1504	701
C_5	13530	29	997	897
D_5	1492	452	19083	1094

conditions, require pickling and more buffer salts. Similarly, in neutralization and basification procceses various Na-based buffing salts and alkali salts are also used. These salts cannot be completely washed out and remain in leather. High concentrations of sodium ions in irrigation water adversely affect soil structure by causing dispersion of clay. Problems with soil permeability can occur depending on sodium, calcium and magnesium ion concentrations in soil [13].

Conclusion

Leather products are manufactured using various processes and chemicals which may show variations depending on the desired the leather characteristics. Considering this particular fact about the leather industry, we have carried out this study on the solid wastes of different tanneries that produce leather for different purposes and therefore use various processes and chemicals. The results of this study have revealed that the leather industry generates waste with different characteristics according to the process step at which it is generated. The obtained data clearly reveals that the contents (both protein and fat) of almost all the fleshing, shaving and trimming wastes, etc., which constitute a great amount of the generated wastes in general, may be reused in other different fields for economic purposes. For example, pre-fleshing wastes can be used in the production of fatliquoring oils and biodiesel, limed fleshings can be used in production of methane gas, grease and protein recovery and fertilizer after composting, shaving wastes can be used in production of leatherboard or retanning agent and gelatine after enzyme/alkali digestion and also shavings and trimmings can be used in the production of activated carbon, etc.

Consequently, in order to determine the most appropriate method for reusing and disposing of these wastes, it is highly important to acquire the information concerning the process steps, during which these wastes are generated, the target product desired to be produced through these processes, and the characteristics of wastes dependent on these factors.

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