## PERFORMANCE ASSESSMENT OF GREEN PRACTICES IN LIMING WITH REDUCTIVE POTENTI-AL CHEMICALS FOR ENVIRONMENTAL SUSTAINABILITY

#### Bahri BASARAN, Ali YORGANCIOGLU, Ersin ONEM<sup>\*</sup>, Behzat Oral BITLISLI

Ege University, Faculty of Engineering, Department of Leather Engineering, 35100, Izmir, Turkiye, e-mail:

deri@mail.ege.edu.tr

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ABSTRACT. Leather processings involved in isolation in pre-tanning stages generate large volumes of solid wastes and high strength wastewater which are major source of environmental pollution characterized by chemical oxygen demand (COD), biological oxygen demand (BOD), total dissolved solids (TDS), total suspended solids (TSS), chromium (III) and phenolics with high pH, strong odor and dark brown color. Leather industry wastewaters with high organic content and pollution load are generated from pre-tanning processes. Conventional pre-tanning process utilises chemicals which attributes to 50-60% of pollution load and alkaline effluent discharge of 70-80% toxicity. In our study, the best practices modelling and eco-compatible process shifts were the objectives for the decrement in high pollution load of large effluent portion for leather industry. Soaking, immunization and liming processes were carried out with reductive chemical additives and surfactants, fibre opening auxiliaries, enzyme based and enzyme assisted chemicals in the factories as an eco-friendly designed recipe. Studies were conducted in Bursa Leather Park, Turkey. The results revealed the advantages of time saving, cost effectivity and minimized pollution load compared to conventional process. Moreover, the designed process uses lower amount of alkaline chemicals and water, gives a very high quality pelts with open grain and yield in 50-55% lime split output with 2.2-2.4 mm thicknesses. KEY WORDS: leather, liming, wastewater, environment, sustainability

#### EVALUAREA PERFORMANȚEI PRACTICILOR ECOLOGICE ÎN OPERAȚIA DE CENUȘĂRIRE UTILIZÂND SUBSTANȚE CHIMICE CU POTENȚIAL DE REDUCERE PENTRU SUSTENABILITATEA MEDIULUI

REZUMAT. Procesele individuale de prelucrare a pielii în etapele dinaintea tăbăcirii generează volume mari de deșeuri solide și ape uzate, care reprezintă sursa majoră de poluare a mediului caracterizată prin consum chimic de oxigen (CCO), consum biologic de oxigen (CBO), total solide dizolvate (TDS), total solide în suspensie (TSS), crom(III) și compuși fenolici cu pH ridicat, miros puternic și culoare maro închis. Apele uzate din industria pielăriei cu conținut organic și poluant ridicat sunt generate din procesele anterioare tăbăcirii. În procesul convențional de pre-tăbăcire se folosesc substanțe chimice care reprezintă 50-60% din cantitatea de poluanți și de efluenți alcalini cu 70-80% toxicitate. În studiul nostru, modelarea celor mai bune practici și schimbările procesului eco-compatibile au reprezentat obiectivele pentru reducerea cantității ridicate de poluanți din efluenții care provin din industria de pielărie. Procesele de înmuiere, imunizare și cenușărire au fost realizate cu aditivi chimici reductivi și tensioactivi, auxiliari pentru deschiderea fibrelor, substanțe chimice pe bază de enzime și asistate de enzime, utilizând o rețetă ecologică. Studiile au fost efectuate în Bursa Leather Park, Turcia. Rezultatele au relevat avantajele economisirii timpului, eficiența costurilor și încărcarea minimă a poluării în comparație cu procesul convențional. Mai mult decât atât, procedeul conceput utilizează o cantitate mai mică de substanțe chimice alcaline și apă, se obțin piei gelatină de înaltă calitate, cu fibre deschise și cu un randament de 50-55% piei șpalt cenușărite cu grosimi de 2,2-2,4 mm.

CUVINTE CHEIE: piele, cenuşărire, ape uzate, mediu, durabilitate

#### ÉVALUATION DES PERFORMANCES DES PRATIQUES ÉCOLOGIQUES DANS L'OPÉRATION DE PELANAGE UTILISANT DES PRODUITS CHIMIQUES À POTENTIEL D'OXYDORÉDUCTION POUR LA DURABILITÉ DE L'ENVIRONNEMENT

RÉSUMÉ. Les processus individuels de traitement de la peau au stade du prétannage produisent de grands volumes de déchets solides et d'eaux usées, qui représentent la principale source de pollution environnementale caractérisée par la demande chimique en oxygène (DCO), la demande biologique en oxygène (DBO), les solides dissous totaux (SDT), les total des solides en suspension (TSS), le chrome (III) et les composés phénoliques à pH élevé, à forte odeur et de couleur marron foncé. Les eaux usées très polluantes contenant des matières organique, provenant de l'industrie du cuir, sont générées par les processus antérieurs au tannage. Dans le processus de prétannage classique, on utilise des produits chimiques représentant 50 à 60% de la quantité de pollution et des effluents alcalins avec une toxicité de 70 à 80%. Dans notre étude, la modélisation des meilleures pratiques et des changements de processus respectueux de l'environnement ont été les objectifs en raison de la réduction de la pollution générée par les effluents de l'industrie du cuir. Les processus de trempage, d'immunisation et de pelanage ont été réalisés avec des additifs chimiques réducteurs et tensioactifs, des auxiliaires d'ouverture de fibres, des produits chimiques à base d'enzymes et assistés par enzymes, en utilisant une recette écologique. Les études ont été menées dans le Leather Park Bursa, en processus conventionnel. De plus, le procédé conçu utilise une plus petite quantité de produits chimiques alcalins et d'eau, permet d'obtenir des peaux en tripe de haute qualité, à fleur ouverte, et donne un rendement de 50 à 55% de peaux en croûte d'une épaisseur de 2,2-2,4 mm. MOTS CLÉS : cuir, pelanage, eaux usées, environnement, durabilité

<sup>\*</sup> Correspondence to: Ersin ONEM, Ege University, Faculty of Engineering, Department of Leather Engineering, 35100, Izmir, Turkiye, Tel.: +90 232 311 2644, e-mail: ersin.onem@ege.edu.tr

#### INTRODUCTION

The tanning industry worldwide produces a significant amount of solid wastes and effluents, environmental concerns about discharge and escalating landfill costs are becoming increasingly serious problems for industry and management alternatives regarding overall consideration based on multi-spot. Due to the fact that 1 ton of wet salted hides yields in 200 kg of finished leather and requires 15-50 m<sup>3</sup> water consumption along with the usage of 500 kg process chemicals, the production emanating 800 kg wastes including tanned solid wastes of 250 kg, non-tanned wastes of 350 kg and discharged wastes of 200 kg in the process water of 15-50 m<sup>3</sup> [1].

Due to both environmental concerns caused by the industrial individuality and pollution emanating inherently from the material itself structure and also global stringencies in many places all over the world have constrained to relocate the industry to designated leather parks along with the implementation of Common Effluent Treatment Plants (CETPs) [2]. In liaison with the globalization and emerging trade unities and also their impacts, cleaner production was adopted and the industry has been obliged to stand out against the enforcement of stringent regulations.

As we know, the traditional loosening fibers process, leather liming (lime, sodium sulfide, etc.), is operated to achieve the objective of the leather loose fibers by means of decomposing the fiber in the high pH of the water. This process produces high-pH value of sewage, which is rich in lime, sulfide, biological oxygen demand, chemical oxygen demand, lipidic saponification or emulsion matter, protein, hair degradation products, organic nitrogen, ammonia nitrogen salt, and so on. Especially for the lime used in this procedure, its large dosage contributes hugely to a great deal of solid sludge.

To bring the tanning industry more in line with present environmental thinking, various methods have been devised to reduce impacts. Big efforts have been made to reduce pollution load, decrease process time and get improvement in the product quality [3, 4]. Other methods of cleaner process in leather-making are tested synchronously [5, 6]. Efforts from researchers and scholars are insistently carried out to reduce leather pollution [7-14]. However, serious betterments could not be achieved until now.

Bursa Specialized Leather and Mixed Organized Industrial Zone called as BIDOSB in abbreviation is the Pioneer Leather Park located in southern Marmara region in Turkey (Figure 1). In the region 200 tons of hides are processed daily, that is one fifth of Turkey production annually. Once in the first time of relocation it was designed just for resettlement of the tanneries in Bursa Province, but later as a consequence of high logistic prices of Tuzla Leather Park, majority of the biggest tanneries in Turkey moved their facilities in this region. However, the infrastructure and common treatment plant capacity which was designed by taking into consideration of the first facility layouts remained incapable of the treatment of newly need-based capacity. Due to full capacity running of the several biggest tanneries which process raw hides to wet-blue, the increasing flow rates and changing the regime of the effluent gave rise to decrease the efficiency of infrastructure and facilities and required to take measures for adequate and efficient treatment. For this reason management of CETP practices are of main importance now and prerequisites are to be taken into consideration. There are 5 control points before CETP entree unit and the last adjustment in equalization tank. But above all the cleaner processing attempts, best available techniques and eco compatible interventions were our main goals for now and so sustainable forth.

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Figure 1. Bursa Leather Park

#### METHODOLOGY

Nowadays, as a key sector, leather industry has a new challenge to deal with: to minimize its environmental impacts by evaluating throughout the production process under a new point of view, that is, detecting inefficiencies and discovering that an additional economic benefit can also be achieved by reducing pollution generation. Expensive end-of-pipe treatment and retrospective clean up should be overcome and complemented with pollution prevention measures in order to reduce costs and risks, and gain competitiveness.

There is one practical and reasonable approach for low level waste (LLW) in any industry; that is, waste minimization at source. Minimization is so applicable in the process inline and source reduction practices reduce or eliminate waste generation. So it comprises environmentally sound recycling practices. This minimization at source and internal recycling as priorities leads to product and process redesign, adopt new technologies and good housekeeping.

With this respect, our work was to suggest a set of recommendations to subsidize the practitioners in implementing initiatives promoting environmentally friendly measures to be adopted. It includes environmentally viable methods to practice in-line and to modify it according to the analytical results. We focused on beamhouse processes and traced back to the process parameters considering the analyses.

A process recipe was set by some preliminary trials and after some modification it was incorporated into a guideline for any private company to use their own chemicals. According to the properties of chemical used and treatments, the variables such as water, liming auxiliary, reductive surfactant, enzyme (keratinase), sodium hydrosulphide, sodium sulphide and lime were studied. The variables applied in the designed recipe, their impacts and the remarks are seen in Table 1. Process guidelines for both conventional and eco-recipe were provided in Table 2 and Table 3. Soaking, immunization and liming processes were applied in one-bath with eco-friendly way to reduce the process water.

Variables	Impacts	Cautions
Water	Continuous phase in dispersion, appropriate mechanical action	Volume of pollution load, wrinkles along with increasing mechanical action.
Liming Auxiliaries	Good dispersion of primary lime and other particles	Good and appropriate swelling, clean-up of soluble materials and epidermal deposits.
Reductive surfactants	Improves loosening of epidermis, scud and the removal of natural pigment.	Allows a significant reduction of the sulfide and reduces pollution and alkalinity.
Keratinase	Degradation of soft keratin and loosening of hair root	Low COD, less sludge, reduced nitrogen.
Sodium hydrosulphide	Milder effect on break-down of disulfide bonds (S-S) of cystine causing keratin degradation	Increases the pH, alkalinity. Total destruction of keratin and thereby increased sulphide and soluble colloids in effluent.
Sodium sulphide	Stronger break-down of disulfide bonds (S-S) of cystine causing keratin degradation	Increases the pH, alkalinity with high swelling and veinness. Increased sulphide and soluble colloids in effluent.
Calcium hydroxide	Opening-up and Collagen hydrolysis	Relaxed leathers with loosened grain, necks' and shanks' wrinkles. Collagen fibers become finer and shorter, which allows obtaining emptier leathers.

## Table 1: Variables, impacts and remarks of the process

Table 2: Basic recipe for soaking and liming process

Process	Chemicals	Percentage (%)	Temp (ºC)	Run (Min.)	Stop (Min.)	Remarks
Pre-soaking	Water	300	26			2 rpm
	Soda ash	0.2				
	Bactericide	0.1				
	Surfactant	0.2		30 30	30 40	Dirt and dung removal
Soaking	Water	200	26			1.5-2 rpm
	Surfactant	0.8		30 30 30 120	30 30 30	
	Bactericide	0.1		60	40	Drain
	Water	200	26	60		Drain
Liming	Water	200	26			
	Sodium sulphide	1.5				
	Lime	6.0		30		
	Sodium sulphide	1.5		30		2 min/60 min T: 14 hour
	Water	10	26-27	60		Hairs and epidermal residuals removed

Process	Chemicals	Percentage (%)	Temp (°C)	Run (Min.)	Stop (Min.)	Remarks
Pre-soaking	Water	100	26			1.5-2 rpm
	Soda ash	0.2				
	Bactericide	0.1				
	Surfactant	0.1		20 30	30 40	Dirt and dung removal
Soaking	Water	100	26			1.5-2 rpm
	Surfactant	0.5				
	Degreasing agent (Non-ionic emulsifier)	0.2		30 180	40	
	Soaking enzyme	0.3		20		
	Reductive auxiliary	0.3-0.4		60		
	Bactericide	0.1		60	40	Entirely soaked
Immunization	Soda ash	0.3-04		60		
	Sodium sulphide	0.1				
	Reductive auxiliary	0.15-0.20	26.5	60		Loosening of pulp. 4 Be', pH:9.5 If necessary
						drain well and add 50% water
Liming	Liming auxiliary	0.7-1	26-27			
	Enzyme (Keratinase)	0.4				
	Non-ionic emulsifier	0.3-0.4		60		
	Lime	1.0				
	Sodium hydrosulphide	1		90		
	Lime	0.5				
	Sodium sulphide	0.4		60	60	
	Lime	0.5				
	Sodium sulphide	0.4				
	Lime	0.5		40	60	5 min/30 min T: 12 hour
	Water	40		60		Hairs and epidermal residuals removed

## Table 3: Eco-friendly designed recipe

Studies were performed in the factories which are active in Bursa Leather Park. After conventional and new designed processes, analyses of electrical conductivity according to SM 2510 B standard process, total suspended solids (TSS) to TS EN 872, total dissolved solids (TDS) to SM 2540 C, chemical oxygen demand (COD) to SM 5220 B, oil & grease to SM 5220 C and total Kjeldahl nitrogen (TKN) to SM 4500 Norg B were applied in the wastewaters. Moreover, total sulphide and total chromium analyses were carried out in equalization tank with SM 4500-S-2 D and ISO 11885 methods, respectively. Thus, how pollution load changes with the new production way in the industrial zone and performance assessment of green practices in liming process was evaluated.

### **RESULTS AND DISCUSSION**

Water is crucial for life and also used in many industrial processes and the industrial sectors take up an average of 22% of the globally used water. With regard to the increasing concern with environment and sustainability, it is vital that natural sources should be inherited to next generations. Leather industry is one of the primary sector with a high water usage and such excessive usage, resulting from adoption of traditional processing methods and equipments lead to acute water availability and effluent treatment problems [15].

Leather industry worldwide is recognized as a serious environmental threat due to its own pollutive potential including salinity, organic load (chemical oxygen load or demand, biological oxygen demand), inorganic matter, dissolved, suspended solids, ammonia, total kjeldahl nitrogen (TKN), specific pollutants (sulfide, chromium, chloride, sodium and other salt residues) and heavy metals etc. [16]. A part of the leather processing, solid and gaseous wastes are also discharged into the environment. Among the numerous phases of the tanning process, the beamhouse phase represents 83% of the BOD, 73% of the COD, 60% of the suspended solids, 68% of the salinity and 76% of the total polluting charge produced during the manufacturing process of hides. Besides, in the beamhouse, the traditional unhairing process with sodium sulfide and lime is responsible for most pollution. Consequently, the development of an alternative unhairing process, characterized by a lower environmental impact than the traditional, represents a priority in the leather research field.

Liming involves the use of alkaline medium (e.g. lime) to condition raw hides and skins. The aim is to remove the hair, flesh and splitting up of the fibre bundles by chemical and physical means [17, 18]. The process not only removes epidermal structures; but also brings about the removal of non-structural protein of the pelts by a certain swelling and plumping. As the term implies leather is made or marred during liming. So, the process modifies the skin fibre for subsequent penetration of other processing chemicals such as tannins and other leather building auxiliaries. Hence; since 1880s neither the process was changed nor improved because of refraining from low quality characteristics. However, the use of this chemical combination generates large volumes of waste that constitutes major source of pollutants amounting to more than 65-70% in combined tannery effluent [19, 20]. Its effluents composed of both organic, inorganic matter and containing metals make the treatment very complicated and expensive. If this wastewater is not properly treated before discharge it may adversely affect the ecology of agricultural land.

Parameters and properties of the conventional process wastewater before the betterment studies were provided in Table 4. Table 4 also shows the analytical results after betterment studies, betterment ratios were given compared to the conventional process.

Parameters	Conventional process results	Results after betterment studies	Betterment ratio
Conductivity (mS/cm)	73	59	19.17%
TSS (mg/L)	31,520	24,660	21.76%
TDS (mg/L)	69,768	64,960	6.89%
COD (mg/L)	58,984	38,976	33.92%
Oil/Grease (mg/L)	650	565	13.07%
TKN (mg/L)	3,417	2,927	14.34%

Table 4: Parameters and analytical results from the process wastewaters and betterments

Indicated results from equalization tank before and after betterment studies were provided in Table 5. Results after betterment studies and conventional process were compared. Betterment ratios were calculated according to the differences of the values between conventional process and eco-friendly designed process.

Parameters	Conventional process results	Results after betterment studies	Reference values	Betterment
Conductivity (mS/cm)	11.71	8.16	8	30.32%
TSS (mg/L)	2,867	1,950	3,000	31.98%
COD (mg/L)	4,998	3,356	5,500	48.92%
Oil/Grease (mg/L)	320	254	668	20.63%
TKN (mg/L)	389	358	120	7.97%
Sulphide (mg/L)	110	82	95	25.45%
Total chromium (mg/L)	138	77	100	44.2%

$\mathbf{T}_{\mathbf{r}}$ is the second s		
Table 5: Indicated results from	equalization tank bet	ore and after betterment studies

Liming has utmost sensitivity. It is the key process of the leather production and directly affects the final leather quality. In case small changes in the process are not applied in a proper way, next processes are not carried out as they should be and leather can be destroyed. That is why, the industrial players never give up from the conventional ways for this process. Different applications as less water utilization, keratinase applications, minimum chemical dosages etc. make the process highly risky. In our study, this vital conditions have been taken into consideration and variables have been conscientiously selected with the sensitive works.

Results showed the considerable betterments after our eco-designed recipe. The highest enhancement in process wastewaters and equalization tank was provided on COD levels. Conventional process had 58,984 mg/L and results after eco-friendly applications were 38,976 mg/L in COD with 33.92% betterment ratio. This ratio was 48.92% in equalization tank with 4,998 mg/L and 3,356 mg/L COD levels, respectively. Table 5 indicated that all the parameters related to pollution load were reduced under the reference values. TSS, TKN, sulphide, oil/grease and total chromium results showed the satisfactory betterment grades with the acceptable distances to the limitations. Only conductivity was over the reference value, but it was also decreased from 11.71 mS/cm to 8.16 mS/cm.

In addition, not only these parameters decreased but also the wastewater treatment charge and the transport charge for tannery sludge was also reduced. Moreover, tons of wastewater and pollution load will be minimized in the industrial zones by less usage of process water and less dosage chemicals via eco-designed recipe. Filtration application was not applied in our study, but in case of this application as well, betterment attained would be much higher than the current case. Our studies are going on the filtration applications for the process and channel waters.

## CONCLUSIONS

Tanning industry produces a significant amount of solid wastes and effluents. As a well-known fact, while removing undesired substances and ingredients out of the structure and isolation in leather processing and thereby; facilitating the reactions between chemicals and skin/hide protein, its pollution load is final and highlighted agenda which needs to be overcome by introducing sustainable cleaner technologies. Novel and environmentally friendly techniques and processing ways for leather industry were highly demanded and forced by the governments after strict environmental stipulation norms. Our study provided the minimized pollution load for leather industry by creating eco-friendly designed process. Cleaner processing attempts, available techniques and eco-compatible interventions will generate the sustainable future for the industry.

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