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Eco-benign stabilization of skin protein—Role of *Jatropha curcas* oil as a co-tanning agent

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ABSTRACT

The research relates to use of *Jatropha curcas* seed oil as co-stabilizing agent for skins and hides in tanning process. Conventionally most of the skins and hides are stabilized through mineral tanning agents predominantly using chromium salts. However due to the ecological and safety concerns associated with chromium salts and other mineral tanning systems there arose a need for an alternative tanning system. Disposal of used leather products has also come under surveillance for their eco-compatibility posing challenges to choice and designing of novel tanning systems. Emerging criteria for such new tanning systems are environmental safety and easy biodegradability of leather products after use. Vegetable tanning agents based on poly phenols of plant materials which were in vogue prior to advent of mineral tanning are re-emerging as principal tanning materials to offset the perceived concerns of mineral tanning. This calls for overcoming the inherent shortcomings associated with the traditional vegetable tanning agents and processes. The major limitations of the vegetable tanned leathers are poor physical characteristics and their high susceptibility to fungal growth. In order to overcome short comings of vegetable tanning, use of multifunctional material(s) of plant origin has been attempted. *J. curcas* seed oil provides a possibility for its use in tanning as an adjunct along with poly phenols. This oil when used as a co-tanning agent along with vegetable tannins eliminates most of the drawbacks associated with conventional tanning. Detailed study has been made by varying the process parameters of tanning. The improvement in softness characteristics has been quantified using a softness tester. Fresh mature cultures of fungal species, *Asp. Niger*, *Asp. flavus* and *Trichoderma viridae* have been used as sources of inoculum for assessment of improvements in fungal resistance. The study indicates a novel tanning process using *J. curcus* seed oil along with vegetable tanning agents can emerge as a viable tanning system based on replenishable resources.

1. Introduction

Tanning is one of the oldest manufacturing activities of human kind. The tanning option based on vegetable tanning is the oldest in the leather sector (Bayramoglu, 2013; Covington, 1998). However the superior properties exhibited by chromium tanned leather made more than 80% of the global industry change over to chromium based tanning. Chromium tanning is the versatile tanning system known hither to. However the growing environmental, health and safety concerns are forcing tanners to adopt alternative benign tanning systems (Covington, 1997; Font et al., 1999; Sundar et al., 2011a, 2011b; Leonard and Lauwerys, 1980). Hence a global search is on for a cleaner, safe and viable tanning system. The various alternatives investigated so far include salts of aluminum, iron, zirconium, titanium, aldehyde or combinations of them. However, most of these do not meet the functional requirements of leather or do not satisfy the eco criteria adequately. Investigations on combination tanning systems predominantly based on vegetable tannins are being carried out as a possible replacement for chrome tanning system. Vegetable tannins are polyphenolic compounds present in the plant extracts. These are compounds with molecular weights in the range of 500–3000 Da. The chromatographic studies indicate that the vegetable tannin extracts are a heterogeneous polyphenolic species. Polyphenols of vegetable tannin extracts are capable of cross-linking with collagen through the formation of multiple hydrogen bonds (Covington, 1997). The leathers processed through vegetable tanning have advantages such as compatibility with human skin, comfort, and high dimensional stability. The tanning methodology adopted also affords viable treatment and disposal of spent liquors. However, the drawbacks associated with vegetable tanned leathers are that, they lack softness, fastness properties (resistance to color change) and are highly susceptible for fungal growth. *Mimosa* (*Accasia Millissima*) is the principal vegetable tanning material employed by the industry. *Mimosa* belongs to a

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group of condensed tannins, which basically contains molecules of flavanoid structure. In one of the earlier works, leather of higher stability was obtained by the treatment of skins with vegetable tannins in the presence of acrylic polymers (Madhan et al., 2001). However, this could not overcome most of the short comings of the vegetable tanning system. Efforts have also been made earlier to partially replace vegetable tannins with resins (Simon and Pizzi, 2003a, 2003b; Simon et al., 2003). Now an attempt has been made to stabilize the skin matrix through combination of vegetable tannins and oil of plant origin. Although oil is being used for conditioning of vegetable tanned leather commercially, the use of oil in this study contributes not only to this aspect of vegetable tanning but also to the tanning of the leather itself seen through its antibacterial/antifungal qualities and characteristics. *Jatropha curcas* a member of the *Euphorbiaccae* is of high commercial importance and has fungicidal properties (Gubitz et al., 1999; Kumar and Sharma, 2008; Lazzeri et al., 2006; Thanapimmetha et al., 2012). The reported calculated value of LD_{50} is 19.19 mg/kg body mass in rats and 49.87 mg/kg in mice respectively (Li et al., 2010). In the present study *J. curcas* seed oil has been used along with *Mimosa* for development of a novel, benign combination tanning system.

2. Materials and methods

2.1. Materials

Twenty wet salted goat skins of Indian origin in the weight range of one kilogram per skin and *Mimosa* tannin extract from Tanzania were chosen for the tanning studies. In order to monitor the changes effectively, the skins were cut into two halves, one half was tanned by conventional vegetable tanning system and other using vegetable–*Jatropha* seed oil tanning system, to avoid variations in results due to skin to skin changes.

Industrial grade *Jatropha* seed oil was analyzed for their iodine value by Hanus method and standard official methods were used for acid value and saponification value determinations (Cocks and Rede, 1966).

2.2. Conventional tanning methodology

The skins from which hair and flesh have been removed through conventional processing system were conditioned to pH 5.0–5.2 using 1% (*w/w*) formic acid solution prior to tanning. The skins were agitated in wooden drum for 1 h with 50% (*w/w*) water and 1% (*w/w*) Pretanning syntan (phenol condensation product). Then 20% (*w/w*) *Mimosa* tannin extract was added and agitated further for 6 h. Finally 1% (*w/w*) formic acid was added in installments and drummed for 1 h. The tanned leathers were then conditioned and dried as per conventional methods.

2.3. Experimental tanning methodology

The skins from which hair and flesh have been removed through conventional processing system were conditioned to pH ranging from 4.2 to 8.5 prior to tanning, using 1% (*w/w*) formic acid solution. The skins were agitated in the wooden drum with 50% (*w/w*) water and 1% (*w/w*) Pretanning syntan for 1 h. Then 10% (*w/w*) *Mimosa* tannin extract was added and agitated for 3 h after which *Jatropha* seed oil was added in different quantities (2, 4, 6, 8 and 10%) for optimization. Finally 1% (*w/w*) formic acid was added in installments and drummed for 1 h. The tanned leathers were then conditioned and dried as per conventional methods.

The tanning process as adopted for both conventional process and experiment is given in Annexure I.

2.4. Evaluation of leather characteristics

Since tanned leathers form resource material for consumer articles like footwear and lifestyle products, there exists certain minimum criteria for physical parameters such as strength and fastness to ensure durability and aesthetics of products. Criteria such as tensile, tear and grain crack strength are indicators of durability of the leathers. Whereas shrinkage temperature is a measure of the ability of leather to resist deformation and disintegration during fabrication of shoes. Criteria such as color fastness ensure aesthetics property of eloquence and uniformity in products during usage on exposure to various environments.

Leather samples for the physical testing were taken parallel to backbone of the leather following the official procedure of sampling and testing according to IUP 1 and 3 methods (IUP, 1958).

The shrinkage temperature, which is a measure of hydrothermal stability of leathers was determined using a Theis shrinkage meter. The tensile strength, % elongation, tear strength and grain crack strength were measured as per official procedures (SLTC Methods, 1965). The test specimens were conditioned for 48 h at $20 \pm 2\,^{\circ}C$ and $65 \pm 2\%$ RH. Strength characteristics of the leathers were tested for tensile strength and tongue tear strength tests in a Universal Instron testing machine (Instron 4501, England). A crosshead speed of 100 ± 20 mm/min was used and the distance between the supports was 40 mm. A load was applied to the center of the samples until fracture occurred and the fracture load was recorded.

The dyed crust leathers also tested for grain crack and grain burst using a lastometer (SATRA 1992). The test specimen was tightly clamped between the circular rings facing grain side upwards and the machine started by forcing the plunger at the rate of 0.2 ± 0.05 mm/s. The surface of the specimen was continuously observed at the center for initial crack on the grain and the maximum distance and force was recorded.

Light fastness characteristics were determined using grey scale ratings as per official procedure (IUP Methods, 1958). This test method is intended to determine the light fastness property of dyed leathers when exposed to artificial day light which is closely same as natural day light. The leather test specimen required to be tested for light fastness and a series of 8 reference wool fabric samples are placed in a panel, covered partly with a plate and exposed to artificial day light continuously unto a period the reference wool fabric grade 4, faded to give a contrast gray scale grade 4. The leather sample exposed simultaneously with wool fabric is to be examined for color change using gray scale. When the color change grade is worser than grey scale rate 4, then the leather is considered to have poor light fastness.

The tanned experimental and control leathers were assessed for softness, grain tightness, fullness and general appearance by standard tactile evaluation technique a subjective trade practice for evaluation of leather. Experienced tanners rated the leathers in a grade scale of 0–10 points for each of the above property. Leather fiber structure from the cross section of the tanned leather has been studied using SEM analysis. Leather samples without any pretreatment were cut into uniform thickness and then gold coated using an Edwards E 306 Sputter coater device. Analysis was performed using a Leica Cambridge Stereoscan S 150 scanning electron microscope.

2.5. Test for fungal resistance

Anti-fungal efficacy of the tanned leathers was tested following a standard procedure (Orlita, 2004). SDA media of volume 20 ml was taken in petri dishes under aseptic condition and leather specimens were placed in the middle of each inoculated nutrient agar and incubated at temperature 25 °C for *Asp. Niger*, *Asp. flavus* and *Trichoderma viridae* and growth of mold was observed for 30 days. Zone of inhibition provides information on fungicidal efficiency of