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The effect of pH and tanning agents on the quality of sheep leather using the ecoprinting dyeing method

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Abstract. Leather eco-printing is an eco-friendly dyeing technique performed by transferring colors from plants. Therefore, this study aims to investigate the effect of pH and tanning agents on the quality of sheep leather using eco-printing dyeing method to produce an eco-friendly animal product. A total of 30 sheep crust leather were randomly assigned using a 2 × 5 factorial arrangement, with three replications. The first factor was the leather tanning agent namely vegetable and mineral, while the second was pH of 5, 6, 7, 8, and 9. The plants used in eco-printing were Kenikir (*Cosmos caudatus*) leaves, Kenikir flowers, Jatropha (*Jatropha curcas*) leaves, Ganitri (*Elaeocarpus ganitrus*) leaves, Ferns (*Pteridophyta*) leaves, Lanang (*Oroxylum indicium*) leaves and Teak (*Tectona grandis*) leaves. Color sharpness and fastness data were collected and then analyzed using a two-way analysis of variance followed by Duncan's Multiple Range Test. The results showed interactions between the tanning method and pH ($P = 0.01$) on color sharpness, with the highest values produced in mineral leather processed under pH 9. Mineral leather had higher color sharpness and fastness values with $P < 0.05$ compared to vegetables. Furthermore, the pH significantly affected sharpness ($P < 0.01$) but had no effect on colorfastness. Based on the results, the combination of mineral tanning with a pH of 9 can be applied for eco-printing to produce the best sheep leather product.

1. Introduction

The animal leather tanning industry is known as a source of considerable environmental pollution due to a high concentration of chemical materials, both solid and liquid. Tanning wastes produce widespread odors, toxic gas, as well as pollution to air, water, and land [1]. Consequently, the leather industry has now focused on eco-friendly tanning chemicals with less environmental impact and sustainability [2]. This is implemented by using vegetable tanning agents rather than minerals [3].

Meanwhile, the dyeing process usually carried out in the finishing stage of leather tanning is famous for using acid dyes containing azo groups, hence, this process generates an extremely complex effluent that is stable and hard to destroy [4]. The toxicity of dyeing agents in the long term potentially affects the environment and human beings. Therefore, alternative dyeing agents are needed to mitigate or even eliminate the harmful consequences [5].

Natural dyes can be made into a solution to produce environmentally friendly types, also, several studies have been conducted to determine the best method for natural dye [4–6]. One of the methods developed is eco-printing [6] which is a coloring technique performed using natural dyes by transferring colors and shapes to the media through direct contact. The ecoprint technique utilizes plant parts in the form of leaves and flowers as a source of dye. This method has been widely used in the textile industry but rarely used in leather media [7]. Meanwhile, eco-printing is not only environmentally friendly because it is biodegradable, less toxic, and less allergenic but also produces unique and beautiful dyeing product [8].

Despite the several advantages over the synthetics and conventional methods, the use of eco-printing in the leather industry is still limited due to the non-availability of standard application procedures. Given that there are no standards, leather products produced from eco-printing have weaknesses such as lack of desirable fastness properties and color sharpness [9]. The process of color transfer from plants can be influenced by pH because each plant has different coloring characteristics. In synthetic dye products, standard pH already exists, hence, the coloring process needs to follow the procedure for each dye. Moreover, various kinds of tanning materials produce different leather characteristics, thereby allowing for differences in the color produced. Therefore, this study aims to investigate the effect of pH and tanning agents on the quality of sheep leather under eco-printing dyeing method to produce an eco-friendly animal product.

2. Materials and methods

2.1. Materials and experimental design
A total of 30 sheep crust leather were randomly assigned using a 2×5 factorial arrangement, with three replicates. The first factor was the leather tanning method namely vegetable and mineral, while the second was pH of 5, 6, 7, 8 and 9. The plants used in eco-printing were Kenikir (*Cosmos caudatus*) leaves, Kenikir flowers, Jatropha (*Jatropha curcas*) leaves, Ganitri (*Elaeocarpus ganitrus*) leaves, Ferns (*Pteridophyta*) leaves, Lanang (*Oroxylum indicium*) leaves and Teak (*Tectona grandis*) leaves

2.2. Ecoprinting process
The leaves and flowers were selected with uniform color, size, and received treatment before entering the eco-printing stage. Subsequently, the leaves were soaked in a solution of 15 ml of vinegar in 1.5 l of water for 15 minutes, while the flowers were immersed in 15 ml of a tunjung solution in 1 liter of cold water for 5 minutes. Tunjung solution was made from 100 grams of Iron (Fe) powder and 1 liter of water, left overnight, and then a clear liquid was taken.

The sheep crust leathers were soaked in 1% surfactant solution for 2 hours for re-wetting, then, the skins were soaked in a mordant solution and soda ash (Na_2CO_3) for 60 minutes. The mordant solution was made by dissolving 30 grams of alum, 15 ml of vinegar, and 1.5 l of warm water (35 °C). Soda ash was given according to pH 5, 6, 7, 8, and 9, then, the skins were air-dried and spread on a plastic sheet.

The leaves and flowers were affixed to the skins and then covered with a blanket with plastic wrap. Furthermore, the skins were rolled up and covered with plastic that was tied, then, they were steamed for 90 minutes at 80 °C. Eventually, they were dried in a room before being proceeded to the laboratory testing.

2.3. Variables and data analysis
The variables observed in this study were color sharpness and fastness, the color sharpness was tested using the 5 scale CMYK color step, with 1 being the worst and 5 being the best. Meanwhile, colorfastness was tested using a crought meter with wet and dry cloth media according to the Indonesian National Standard (SNI) 06-0996-1989. The data were analyzed by two-way analysis of variance (ANOVA) followed by Duncan's Multiple Range Test.

3. Results and discussion
The results showed that there was an interaction demonstrated by $P < 0.05$ between the tanning type and pH on color sharpness, but there was no interaction as shown by $P = 0.66$ on colorfastness (Table 1). Color sharpness of the tanned mineral crust improved following the increase of pH from 5 to 9,

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while the vegetable-tanned leather only increased up to pH 6 as shown in Figure 1. The mineral-tanned leather has a light color [10], and higher pH is known to increase color transfer from plant to leather [11], hence, an increase in the pH improved color sharpness. Meanwhile, vegetable tanning materials produced from plant parts with color pigment tends to provide secondary dyeing effect as well as dark or colored leather products [12], hence, the increase in pH was only optimal up to 6.

Table 1. The interaction of tanning agents and pH on the quality of sheep leather.

Tanning agents	pH	Color sharpness	Color fastness
Mineral	5	62.00 ± 2.00^{b}	5.00 ± 0.00
Mineral	6	63.33 ± 2.31^{b}	5.00 ± 0.00
Mineral	7	65.33 ± 2.31^{b}	5.00 ± 0.00
Mineral	8	68.67 ± 7.57^{ab}	5.00 ± 0.00
Mineral	9	75.33 ± 1.15^{a}	4.66 ± 0.39
Vegetable	5	12.67 ± 1.15^{e}	4.58 ± 0.63
Vegetable	6	31.27 ± 1.10^{c}	4.58 ± 0.63
Vegetable	7	29.20 ± 9.85^{cd}	4.58 ± 0.63
Vegetable	8	20.67 ± 6.43^{de}	4.75 ± 0.32
Vegetable	9	24.67 ± 6.43^{cd}	4.75 ± 0.32
Significance		0.01	0.66

$^{a-e}$ Different superscript letters following mean values in the same column indicate significant differences ($P < 0.05$).

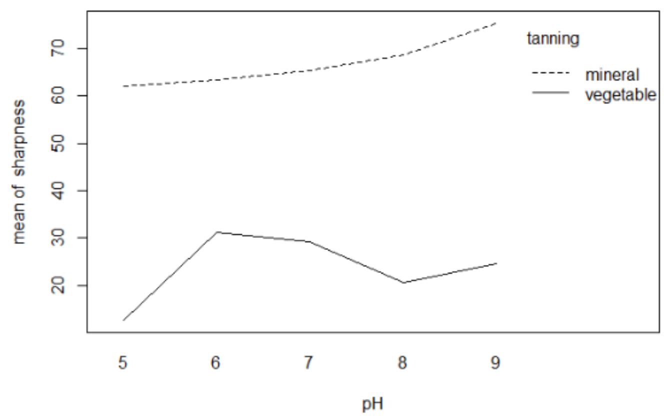

Figure 1. The interaction plot between tanning agents and pH on color sharpness of sheep leather.

The mineral-tanned sheep crust leather showed better color sharpness with $P < 0.05$ and colorfastness $P = 0.03$, compared to the vegetable-tanned (Table 2). This result is in agreement with a previous study on Iranian Sheep leather [13]. Mineral tanning agent produces bright or white skin because it does not contain color, while vegetable tanning agents produce dark leather colors according to plant sources because the color pigments of tanners provide a secondary effect as a dyeing agent [10,12]. Ecoprinting uses the original colors of plants with different other types. Similar to when coloring on white paper, eco-printing on mineral-tanned leather produces high contrast colors. The shape of each leaf and flower is clearly imprinted on the leather and is prominent. Meanwhile, eco-printing on vegetable-tanned leather tends to mix color pigments with others from various plants leading to dark leather and the shape along with the color of the leaves are not printed properly.

Ecoprinting on mineral-tanned leather has a high colorfastness because it has a strong bond between the dye molecules and leather fibers. The addition of a mordant derived from the mineral aluminum leads to a strong bond with the tanning agent. Mordant can increase the adhesion of various natural dyes, aluminum compounds in the mordant form a chemical bridge between dye and leather fiber molecules [14]. In contrast, vegetable-tanned leather does not have mineral bonds in the fiber, hence, the addition of mordant does not provide maximum effect to resist fading [15,16].

Table 2. The effect of tanning agent and pH on eco-printing quality of sheep leather.

Treatment	Color sharpness	Colorfastness
Effect of tanning agent		
Mineral	66.93 ± 5.90^a	4.93 ± 0.21^a
Vegetable	23.69 ± 8.55^b	4.65 ± 0.48^b
Significance	<0.01	0.03
Effect of pH		
5	37.33 ± 27.06^b	4.79 ± 0.47
6	47.30 ± 17.64^a	4.79 ± 0.47
7	47.27 ± 20.80^a	4.79 ± 0.47
8	44.67 ± 27.03^a	4.87 ± 0.25
9	50.00 ± 28.06^a	4.71 ± 0.33
Significance	<0.01	0.95

$^{a\text{-}b}$ Different superscript letters following mean values in the same column indicate significant differences ($P < 0.05$).

The pH affected the color sharpness as demonstrated by $P < 0.05$ but not the colorfastness with $P = 0.95$. The pH 5 produced the lowest color acuity compared to 6, 7, 8, and 9 as shown in Table 1. Alkaline levels in the mordant increase the release of color from plants, hence, it is more easily transferred to bind the skin molecules. Therefore, a rise in the pH can increase the sharpness of the color. Each part of the plant at different age conditions contains different phytochemicals especially tannin or phenol [17]. Under acidic conditions, phenol is in the form of a molecule, while phenol is in the form of a phenolic ion under alkaline conditions. When the hydrogen atom in the –OH group of phenol is cut off and turns into a phenolic ion, the solubility in polar solvents increases. This causes the number of dissolved flavonoids to increase with the addition of a base. The higher the total dissolved flavonoid and tannin compounds, the higher the compounds are absorbed by the fabric during the dyeing process, thereby increasing the color intensity [11,18].

4. Conclusions

Based on the results, it was concluded that the combination of mineral tanning agent with pH 9, can be applied for tannery eco-printing to produce the best sheep leather product. However, this study does not fully support the environmental friendliness of the leather industry because the best results were obtained using mineral tanning agents.

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