(REFEREED RESEARCH)

THERMOPHYSIOLOGICAL COMFORT PROPERTIES OF THE LEATHERS PROCESSED WITH DIFFERENT TANNING AGENTS

FARKLI TABAKLAMA MADDELERİ İLE İŞLEM GÖRMÜŞ DERİLERİN TERMOFİZYOLOJİK KONFOR ÖZELLİKLERİ

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ABSTRACT

The main purpose of the leather products is to protect from cold. So determination of the thermophysiological properties of the leathers is very important to find out the usage area. In this study thermophysiological comfort properties of the leathers tanned with different tanning agents were searched. Chrome, vegetable, zirconium, glutardialdehyde, phosphonium tanning materials were applied to the same type of leather. According to results the leathers tanned with chrome have the highest thermal conductivity whereas lowest thermal resistance. The highest thermal resistance values were obtained with glutardialdehyde tanned leathers. Vegetable tanned leathers showed highest thermal absorptivity that means cooler feeling at the first contact. Zirconium tanned leathers showed the highest water vapour permeability among the others.

Keywords: Tanning agents, Leather, Tanning process, Thermal resistance, Thermal conductivity, Water vapour permeability, Chrome, Vegetable, Zirconium, Glutardialdehyde, Phosphonium

ÖZET

Deri giysilerin kullanım amaçlarından birisi soğuktan korumasıdır. Bu nedenle mamul derinin kullanım alanının belirlenmesinde, termofizyolojik özelliklerinin bilinmesi önemli bir unsurdur. Bu çalışmada farklı tabaklama maddeleri kullanılarak tabaklama işlemi yapılmış derilerin termofizyolojik konfor özellikleri araştırılmıştır. Aynı tip deriler üzerine krom, zirkonyum, glutardialdehit, fosfonyum ve bitkisel tabaklama maddeleri uygulanmıştır. Elde edilen sonuçlara göre krom ile tabaklama yapılan deriler en yüksek ısıl iletkenlik gösterirken en düşük ısıl direnç değeri vermiştir. En yüksek ısıl direnç değeri glutardialdehit tabaklama maddesi ile tabaklanan derilerden elde edilmiştir. Bitkisel tabaklama maddesi kullanılan deriler en yüksek ısıl soğurganlık değeri göstermiş olup bu sonuç bu tip derilerin ilk temasta soğuk his vereceği anlamındadır. Zirkonyum ile tabaklanan deriler tüm deriler içerisinde en yüksek su buharı geçirgenliği göstermiştir.

Anahtar Kelimeler: Tabaklama maddeleri, deri, tabaklama işlemi, ısıl direnç, ısıl iletkenlik, su buharı geçirgenliği, krom, bitkisel, zirkonyum, glutardialdehit, fosfonyum

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1. INTRODUCTION

Natural leather is a very attractive material for applications involving industries ranging from that of technical clothes to fashion. But leather apparels are generally used for protection against cold weather conditions[1]. Designing and processing of leather plays a major role in providing thermal comfortability of that garments.

In recent years the expectation from the garments have increased because of the consciousness of the consumers and the rise of people's living standards. Not only body movement and aesthetic comfort but also thermal properties and interaction with the body also become to the forefront [2,3]. Clothing physiology is the interaction of clothing and human body in various environment [4]. It is expected from a garment to help to protect thermal balance of the body, and to maintain the body temperature. Garments work as a barrier to conserve body temperature of a human being in different atmospheric conditions. So the main function of the garments is to constitute a regulation system for keeping body thermal balance at the mean value and water vapour transfer between body and enviroment even if outer atmospheric conditions and physical activities change [5,6]. The thermophysiological comfort is a complex phonemon that contains perception of the garment comfort and coolness, warmness, wetness feelings. Thermal comfort is the sense of satisfaction in relation with the climate condition of surrounding environment. The sense of comfort and discomfort does not depend only on temperature and relative humidity of the environment, but depends also on the human metabolism that activate processes of body thermal regulation able to determining different thermo-hygrometric sensation of the skin. This means that, at different external conditions, the same temperature is not always perceived as "comfortable" . When someone wear a footwear or a garment, thermal comfort depends on their capability to isolate body or to allow the thermo regulation mechanism [7].

Another parameter showing the comfort is warm–cool feeling. When the human touches a garment that has a different temperature than the skin, heat exchange occurs between the hand and the fabric, and the warm–cool feeling is the first sensation. Which feeling is better depends on the customer; for hot summer garments a cooler feeling is demanded, whereas in winter warmer feeling is preferred [8].

Leather processing can simply be defined as, modification of hides/skins by a sequence of chemical and physical treatments. Various physical, chemical and fastness properties are required from leather products depending on their field of use. Although leather processes and the preferred chemicals have a big contribution to designate of final leather characteristics, the raw material type, tanning material have the most important effect. Tanning is the most important stage giving the hide or skin the required stability [9,10].

The tanning process is the stabilization of the collagen matrix to retain a separated fiber structure and to increase the hydrothermal stability. This is the stage at which the pelt becomes 'leather' and is then resistant to putrefaction or rotting. Organic or inorganic based materials which are able to crosslink with reactive groups of the collagen are used in the tanning process [9,11].

Opposite to the thermal properties of the fabrics there are a few studies related with leathers. Krishnaraj K. et al., investigated thermal insulation properties of the leathers and also for different designs of the garment construction. Five different types of top grade leathers were procured from different sources with uniform size and thickness. The construction types chosen were based on front opening in a garment namely "zipped" and "buttoned". It was shown that the selected leathers and garment constructions met the minimum thermal insulation value required for protection against cold as prescribed by International standards. Also it was concluded that there was a significant change in thermal insulation values when the wear temperature conditions were changed. Type of construction of the garment also has an influence on the thermal insulation values. Among all leather types (except suede), the zipped garment construction has more thermal insulation than the buttoned type. Goat suede leather has comparatively less thermal insulation value and fur (sheep nappalon) leather has better thermal insulation value compared to all the leathers selected [1].

In the study of Mascolo et. al., thermal comfort ranges were determined using thermal sensors inside for two different footwear wear. The first one was characterized by very high thermal and vapour transpiration, the second one by very low thermal and perspiration properties. The two footwears were unlined, having the same polymeric sole and constituted by uppers having different thermal conductivity, water vapour permeability and water vapour transmission rate. The first sample was an ovine chrome tanned leather (thickness 1,00 mm), the second one was a bovine chrome tanned leather (thickness 1,30 mm). Thermal conductivity, water vapour permeability and reflection IR frequencies of solar radiation were tested. Ovine leather showed higher vapour flow. It was found that samples characterized by higher IR percentage reflectance, showed a lower increasing of surface temperature and the item colour affects its temperature and thus determines the increasing of temperature of the inner surface in contact with skin, because of the thermal conductivity of the material [7].

Yumiko Tsunoda and et al. simulated the microclimate within the clothing to evaluate the leather clothing material under conditions of early and late summer in Japan, namely 25 °C and 65% RH. Cape Town hair sheep was used and full-grain leather (eco leather, chrome leather, punching leather) was prepared. Cotton, wool textile, and a synthetic material (epidermal layer: polyurethane, foundation cloth: polyester) were used for control. They used hot plate testing apparatus for simulating the clothing microclimate for standard, air blowing and sweating conditions. They also conducted wear trials of jackets and evaluated the thermal comfort level reported by the participants in the trials. A temperature and humidity sensor and a heat sensor were attached to the each participant, resting and walking trials were applied. The thermal sensation, discomfort sensation, sense of soaking, sense of sweating, and sense of stuffiness were evaluated by subjectively on the basis of a five point scale. The following results were obtained; The simulation tests of the clothing microclimate showed that the absolute humidity in the leather decreased rapidly after sweating, and of all leathers, the punching leather showed the best results. The results of the wear trials were nearly identical to the simulation tests. The sensory evaluation data for leather were better relative to the fiber material. Among all materials, punching leather had the best sensory evaluation behavior. Strong correlation was observed between the simulation of the clothing microclimate and wear trials for absolute humidity. Moreover, strong correlation was observed between the discomfort sensation and sense of soaking in the sensory evaluation of the simulation of absolute humidity [12].

Tanning materials demonstrate differences both in terms of features that are given to the leathers and color. There are numerous studies on the properties provided by the leather tanning systems and tanning agents worldwide. Fathima and et al. characterized porosity of the native, chrome treated, vegetable treated leathers using by mercury intrusion porosimetry (MIP). They found that the porosity of the leathers is % 33.17, 16.34 and 31.35 respectively [13].

Although one of the aims of leather garments is to protect from the cold, the number of research on this subject is very few. However thermal properties of the leathers for garments and shoes are as important as other quality characteristics. When previous studies have examined, the effect of the tanning materials on the leather properties were researched by many researcher but a few researches were found about the thermal properties of the leathers. Therefore in this study thermal properties of the leathers tanned with different tanning agents were searched. In addition to the chrome tanning materials which are mostly preferred all over the world vegetable, zirconium, glutardialdehyde, phosphonium tanning materials were used.

2. MATERIALS AND METHODS

2.1 Materials

Pickled Tunis origin sheep skins were employed for thermal properties of different tanned leather trials. All the chemicals were used in commercial grade.

2.2 Leather procedure

Ten skins were used in the study. The skins were divided lengthwise and applied the tanning processes. The detailed leather production processes were presented in Table 1-5.

Table 1. Tanning processes with chrome tanning material

Process	%		Time	pH
Depickle	150 1 1 1.2	Salt water (8 'Be) Sodium formate Cationic degreasing agent $NaHCO_3$ 3*15'	10' 20 15' 1.5-2h	 5
Bating	100 2	Su 38 °C Acidic bating enzyme	 30'	
Washing				
Degreasing	6 +100	Degreasing agent Water 35 °C	60' 30'	
Washing	150	Salt water 35 °C(2Be)	15'	
Pickle	80 1 2 1 1.5	Salt water (8 'Be) Sodium formate Electrolyte stabile fatliqouring agent Formic acid (1:10) (2*10') H_2SO_4 (1:10) (3*15')	10' 20' 20' 20' 90'	 3.0
Tanning/Basification	8 1 1	Basic chrome sulphate (%33 Basicity) Sodium acetate $NaHCO_3$ (2*15')	4h 20' 1h	 3.8
Neutralization	100 1 0.5	Water 40 C Coratyl NZ (1:10) (2*15) $NaHCO_3$ (1:10) (2*15)	 30' 60'	 5.0
Retanning	60 2 2 3 3 1 +150 10 1	Water 45 C Dyeing auxiliary agent Acide dye Anionic resin Synthetic tanning agent Polymer tanning agent Water 60 C Combine fatliqouring agent Formic acid	 10' 45' 30' 30' 30' 60' 20'	 3.8
Washing				

Table 2. Tanning processes with zirconium tanning material

Depickling bating, degreasing and washing procedures were performed as described in Table-I.				
	%		Time	pH
Pickle	80 1 1 2 2.5	Salt water (8 'Be) Sodium formate Formic acid (1:10) (2*10') Electrolyte stabile fatliqouring agent H_2SO_4 (1:10) (3*15')	 20' 20' 2h	 2.5
Tanning /Basification	5 5 1 2.5	Zirconium sulphate Zirconium sulphate Sodium acetate $NaHCO_3$ (3*15')	30' 1h 20' 5h	 3.8
Neutralisation and retanning procedures were performed as described in Table-1.				

Table 3. Tanning processes with vegetable tanning material

Depickling bating, degreasing and washing procedures were performed as described in Table-I				
	%		Time	pH
Tanning/ Basification	80	Salt water (2 Be)	5’	
	2	Disperse synthetic retanning agent	20’	
	6	Mimosa	1h	
	2	Electrolyte stabile fatliquoring agent	20’	
	2	Disperse synthetic retanning agent	20’	
	6	Chestnut	1h	
	2	Electrolyte stabile fatliquoring agent	20’	
	6	Mimosa	1h	
	0.5	Formic acid	45’	4.0
Neutralization	100	Water 40 C		
	2	Coratyl NZ (1:10) (2*15’)	60’	5.0
Retanning	Retanning procedures were performed as described in Table-1.			

Table 4. Tanning processes with phosphonium tanning material

Depickling bating, degreasing and washing procedures were performed as described in Table-I				
	%		Time	pH
Pickle	80	Salt water (8 ‘Be)		
	1	Sodium formate	20’	
	2	Electrolyte stabile fatliquoring agent	20’	
	3	Formic acid (3*15’)	2h	3.5
Tanning/ Basification	8	Tetrakis Hydroxymethyl Phosphonium Sulphate	4h	
	1	Sodium acetate	20’	
	0.5	$NaHCO_3$ (2*15’)	60’	5.0
Oxidation	100	Water 20 C		
	0.3	H_2O_2 (The skins wait for 1-2 day)	60’	
Retanning	Retanning procedures were performed as described in Table-1.			

Table 5. Tanning processes with glutardialdehyde tanning material

Depickling bating, degreasing and washing procedures were performed as described in Table-I				
	%		Time	pH
Pickle	80	Salt water (8 ‘Be)	10’	
	1	Sodium formate	20’	
	2	Electrolyte stabile fatliquoring agent	20’	
	3	Formic acid (1:10) (3*10’)	2h	3.5
Tanning/ Basification	2	Glutardialdehyde	30’	
	2	Glutardialdehyde	3h	
	1	Sodium acetate	20’	
	0.5	$NaHCO_3$ (2*15’)	1h	
	0.5	$NaHCO_3$ (2*15’)	30'	4.5
Neutralization	100	Water 40 C		
	2	Coratyl NZ (1:10) (2*10')	30’	5.0
Retanning	Retanning procedures were performed as described in Table-1.			

2.3 Test methods

Sem

Scanning electron micrographs of the cross section of the leathers was taken used by HITACHI TM-1000 tabletop microscope with x100 magnification.

Thermal Properties

The thermal comfort properties of the leathers were measured on Alambeta instrument constructed by Hes (Czech Republic) (Figure1). The principle of this instrument depends in the application of a direct ultra thin heat flow sensor, which is attached to a metal block with constant temperature which differs from the sample temperature. When the measurement starts, the measuring head containing the mentioned heat flow sensor drops down and touches the planar measured sample. As soon as the hot plate touches the fabric surface, the amount of heat flow from the hot surface to the cold surface through the fabric is detected by heat flux sensors. There is also a sensor, which measures the thickness of the fabric. These values are then used to calculate the thermal resistance of sample[14,5].

Thermal conductivity

Thermal conductivity (λ), thermal resistance (R), thermal absorptivity (b) values of the leathers were obtained from the instrument. Thermal conductivity (W/mK) can be thought of as a flux of heat (energy per unit area per unit time) divided by a temperature gradient (temperature difference per unit length).

Thermal resistance

Thermal resistance is an indication of how well a material insulates and under a certain climate condition if the thermal resistance of clothing is less, the heat energy will gradually reduce with a sense of coolness [15,16]. This parameter is connected with thermal conductivity and fabric thickness by the equation given below:

$R = h / \lambda$ *(m^2 K / W)*

where:

h: Thickness (m)

λ: Thermal conductivity (W/m K)

Thermal absorptivity

Thermal absorptivity value which is the objective measurement of warm-cool feeling determine the contact temperature of two materials and [17] it can be expressed as:

$b = (\lambda \rho c)^{1/2}$ $(W s^{1/2} / m^2 K)$

where:

λ: Thermal conductivity (W/m K)

ρ: Fabric density (kg / m^3)

c: Specific heat of fabric (J / kg K)

If the thermal absorptivity of clothing is low, it gives a warmer feeling at first contact [5]. The better feeling depends on customer for instance in cold regions warmer feeling is demanded, whereas in hot summer days cooler clothing is preferred [18,19]. The use of thermal absorptivity is justified just for the short initial time of thermal contact between the skin and the fabric and this initial sensation is most important for the warm-cool feeling [14].

Fig.1. Alambeta instrument

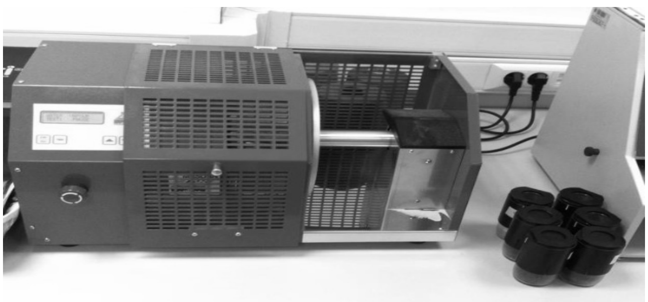


Fig.2. Water vapour permeability tester

Water vapour permeability

Water vapour permeability of the leathers was measured on Mesdan water vapour tester instrument (Fig. 2). The leather samples were tested according to ISO EN 14268. Leather sample is fixed on the neck of beaker containing a silica gel and is exposed to a quick air drought. The beakers rotate a certain time and are weighed at regular intervals to measure the quantity of water vapor absorbed by silica gel inside the beaker, hence passed through the sample [19]. The permeability to water vapour (mg/cm^2h) was calculated by the formula given below.

$P= 60m400/ (\pi d^2)t$

t: is the time between two consecutive weighing, min.

m: is the weigh variation, mg

d: is the diameter of beaker, mm

All the tests were repeated three times and average value was taken as a test result. All measurements were performed under the standard atmospheric conditions (20±2°C temperature, %65±4 relative humidity).

3. RESULTS AND DISCUSSION

Leather apparels are used for protection against cold conditions. However the total comfortability of the apparel on wearing depends on many other factors. In this study effect of different tanning materials on the leather thermal comfort properties have been investigated. Structure and connectivity of pores in leather influence the heat and mass transport processes associated with the thermoregulatory function of the material [20].

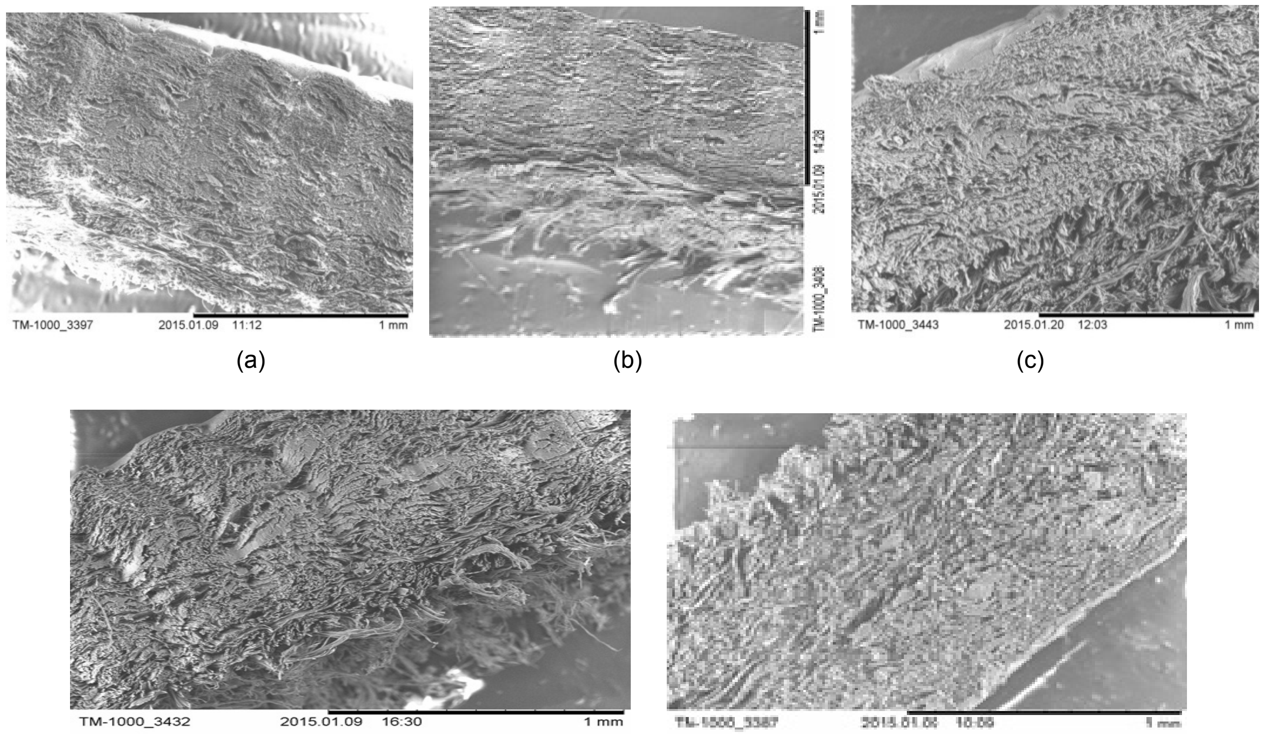
Leather derives its unique property of breathability through this porous network. The conversion of skin into leather subjects the matrix to various physical and chemical operations [21]. The SEM photos of the different tanned leathers were given in Figure 3.

Thermal resistance, absorptivity, thermal conductivity and thickness test results of the leathers which processed with different tanning materials were given in Table 6.

According to the test results the leathers tanned with chrome have the highest thermal conductivity whereas lowest thermal resistance (Fig. 4, Fig. 5). Opposite to that the leathers processed with glutardialdehyde have the highest thermal resistance values. Structural construction of the leathers has similarity with textiles. The binding structure of the tanning material are different for different tanning materials and the porosity between the fibers are changing belong to the tanning material. The difference in the % porosity between tanning treated leathers is due to the crosslinking effect brought about by these agents. Therefore it can be said that tanning material has an effect on the thermal conductivity and thermal resistance of the leathers.

Air has higher thermal resistance as compared with fiber. As explained previous studies leathers processed with vegetable tanning materials have higher porosity values than chrome tanning materials [14]. And also as the SEM photos of the leathers were examined it was found that glutardialdehyde and vegetable tanned leathers have higher porosity that causes higher thermal resistance. Opposite to the zirconium tanned leathers, chrome tanned leathers have the lowest thermal resistance whereas higher thermal conductivity.

As the thermal absorptivity values are evaluated the leathers tanned with vegetable agents have the highest thermal absorptivity that means that cooler feeling at the first contact (Fig.6). The glutardialdehyde tanned leathers have the lowest absorptivity values. Lower absorptivity gives warmer feeling, so especially this type of leathers can be preferred for the products that will be used for winter.



(a) (b) (c)

(d) (e)

Fig. 3. Scanning electron micrographs (100× magnification) showing the cross-section view of (a) glutardialdehyde (b) vegetable, (c) chrome (d) zirconium (e) phosphonium tanned leathers

Table 6. Thermal properties test results of the leathers

Tanning Materials	**Thermal conductivity** (W.m^{-1}.K^{-1})	**Thermal resistance** (m^2.K.W^{-1})	**Thermal absorptivity** (W s$^{1/2}$ m^{-2} K^{-1})	**Thickness** (mm)
Zirconium	0,0528	0,0258	188	1,347
Glutardialdehyde	0,0480	0,0275	169	1,315
Phosphonium	0,0523	0,0268	194	1,379
Chrome	0,0544	0,0255	200	1,388
Vegetable	0,0506	0,0271	219	1,367

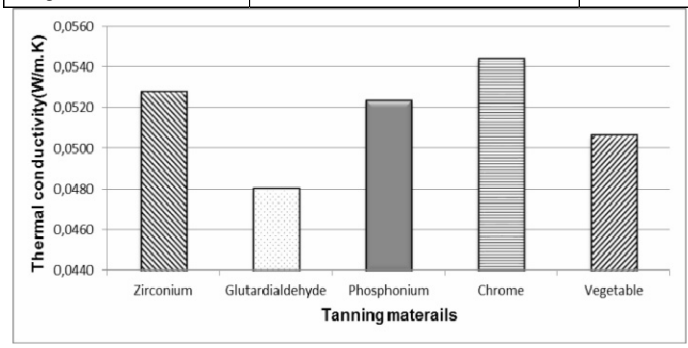


Fig. 4. Thermal conductivity values of the leathers

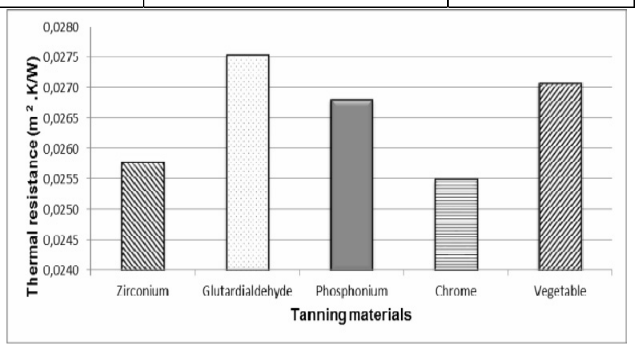


Fig. 5. Thermal resistance values of the leathers

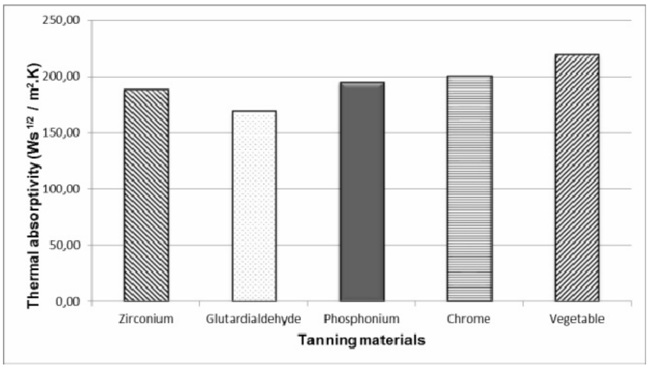


Fig. 6. Thermal absorptivity values of the leathers

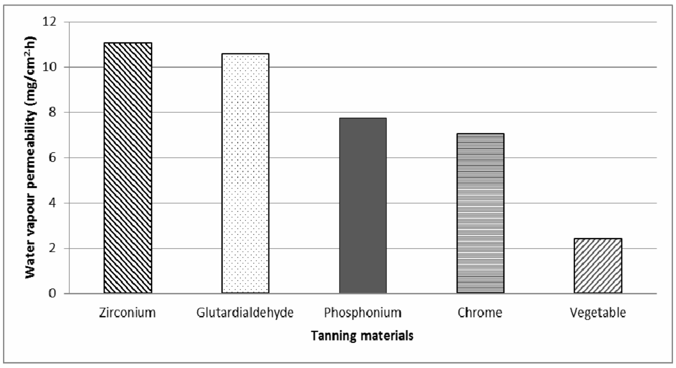


Fig. 7. Water vapour permeability values of the leathers

Water vapour permeability values of the leathers were given in Table 7 and Figure 7. Water vapour permeability values can change depending on structural properties of leather like as porosity, thickness, collagen and elastin fibre bundles, leather processing steps and presence of grain, papillar layers and density. However the tanning type affects the permeability values significantly. The previous studies showed that as the density of the leather increases the water vapor permeability decreases [22].

Table7. Water vapour permeability results of the leathers

Tanning materials	Water vapour permeability ($mg/cm^2.h$)
Zirconium	11,04
Glutardialdehyde	10,58
Phosphonium	7,74
Chrome	7,05
Vegetable	2,41

According to results leathers tanned with zirconium and glutardialdehyde have higher water vapour permeability than the others. Vegetable tanned leathers gave the lowest values. This result can be explained with the higher density of the vegetable tanned leather that causes lower vapor permeability. The garments which are worn during higher activity should allow the perspiration to pass through. Therefore zirconium tanned leathers will give better wearing comfort.

4. CONCLUSION

There are many tanning materials used in leather production. In this study thermophysiological properties of the leathers tanned with different tanning agents were investigated. Chrome, vegetable, zirconium, glutardialdehyde, phosphonium tanning materials were applied to the pickled Tunis origin sheep leather.

According to test results it was revealed that different tanning agents have significant effect on the thermal and vapour permeability properties of the leathers. The leathers processed with glutardialdehyde have the highest thermal resistance values. Opposite to that, chrome tanned leathers showed lowest thermal resistance because of the lower porosity. So glutardialdehyde tanned leathers can be recommended for winter. As the thermal absorptivity values are evaluated, the leathers tanned with vegetable agents have the highest thermal absorptivity that means that cooler felling at the first contact whereas glutardialdehyde tanned leathers have warmer feeling effect with lower absorptivity values. Water vapour permeability is also very important comfort parameter for leathers. Zirconium tanned leathers showed the highest water vapour permeability among the others. So these types of leathers can be recommended for summer suits and shoes. As a conclusion, different thermophysiological properties can be achieved by using different tanning agents for the leathers. Therefore depending on usage area of the leathers suitable tanning agents and processes should be applied.

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