

Heat and Mass Transfer in Leather Drying Process

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ABSTRACT: *Leather manufacturing involves a crucial energy-intensive drying stage in the finishing process to remove its residual moisture. Determining drying characteristics of leather is vitally important so as to optimize the drying stage. This paper describes an analytical way for determination of the drying characteristics of leather. The model presented, is based on fundamental heat and mass transfer equations. Variations in temperature and moisture content distribution is solved numerically using the finite difference method. The effects of operation parameters, are examined using the model. The results of the parametric study provide a better understanding of the drying mechanisms and may lead to a series of recommendations for leather drying optimization. It opens the possibility for further investigations on description of relationships between model parameters and drying conditions.*

KEY WORDS: *Convective drying, Moisture and heat transfer, Leather drying optimization.*

INTRODUCTION

Convective drying is usually encountered in many industrial fields (food industry, building industry, leather industry,...). Therefore, the study of this type of problem becomes very important and for several decades has attracted the attention of many investigators [1-14].

Leather fabrication has become an important industrial development worldwide, similar to other technologically advanced process industries. However, some of the unit operations involved in this industry, especially the drying process, are still based on empiricism and tradition, with very little use of scientific principles.

Although drying of leather is an old and well established process, it is largely controlled by the rule of thumb. This is not so much due to a lack of understanding of the basic physical principles involved as due to a lack of knowledge of the rates of various transfer mechanisms and how they interact. The type of information required for the application of modern control techniques to the drying process of leather is still largely unknown. The present study is part of a more general attempt to analyze the heat and mass transfer in leather in terms of basic mechanisms so that a mathematical description of the overall process can be established. In the meantime,

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the complexity of the problem makes it doubtful that it will ever be possible, or indeed economic, to find a complete solution. However, an analysis like the present one can contribute some useful general principles which may help to advance the leather drying technology.

Among the many processes involved in the leather industry, drying has an essential role since it determines the final texture, consistency and flexibility of the leather product. Widespread methods of leather drying all over the world are mostly convective methods requiring a lot of energy. Specific heat energy consumption increases, especially in the final period of the drying process, when the moisture content of the leather approaches the value at which the product is storable.

From among the several theories so far suggested to explain the migration of moisture in porous media, three have won general recognition: the diffusion theory, the capillary flow theory, and the evaporation-condensation theory.

It is still often assumed that under constant drying conditions, a constant drying rate only occurs when the surface of the drying medium is completely wetted, but for leathers, this, in general, is not true.

Numerous governing equations for heat and mass transfer have been derived by many researchers. Transient state diffusion in hygroscopic textile fibers was first analysed by Henry [16], who obtained an approximate analytical solution. He showed that moisture diffuses into the porous structure of fabric and the solid phase of the fabric is hygroscopic. Later, Nordon and David [17] improved Henry's model, and they were able to solve the nonlinear differential equation of moisture transfer using the finite-difference method. Farnworth [18] introduced a model to solve transient heat and mass transfer in a multilayered clothing system. Beard [19] suggested a simplified mathematical model to obtain the temperature and moisture distribution of material in a convective dryer. He assumed that a fabric consists of two layers, a dry layer and a wet one. But his analysis did not describe details of what was going on inside the fabric. Also, he used two experimental constants to fit his data to the experimental results of measured temperature variation inside the dryer.

However, energy consumption in a convective dryer can be reduced by optimizing the drying process using mathematical analysis of the temperature and moisture

distribution in the material. Thus, development of a suitable mathematical model to predict the accurate performance of the dryer is important for energy conservation in the drying process.

The aim of this paper is to proceed further on our previous published works [1-11], with the knowledge of drying mechanisms and leather characteristic under convective heating conditions.

Sample Preparation

Experiments have been carried out with wet bull nappa leather sample selected from typical pieces of leather as they come from the previous factory process. Bull nappa processing was carried out on a sample of weight category of 30 kg preserved with salt. For liming a mixture of sodium sulphide and calcium hydroxide was used. Ammonium sulphate and lactic acid was used for deliming. For pickling formic and sulphuric acid including sodium chloride were used. Tanning was carried out with basic chrome salts offering 7% on the weight of the hide (26% of Cr_2O_3 and 33% of basic) followed by 1.2% sodium bicarbonate. Neutralization was done with calcium formate and retannage followed using acrylic and other syntans. The tanned sample were fatliquored with synthetic fish oil in lanolin oils.

Present Drying Model and Solution Method

The theoretical model proposed in this article is based on fundamental equations, which we used in our previous work [9] to describe the simultaneous heat and mass transfer in porous media. The configuration is that of a flat slab constituted with a solid phase that is inert and rigid, a liquid phase (pure water) and a gaseous phase which contains both air and water vapour. The theoretical formulation of heat and mass transfer in such a media is usually obtained by a change in scale. Under constant environmental conditions the process of drying can be divided into a "constant rate" and one or two "falling rate" periods. When the initial moisture content is high enough, a considerable amount of moisture leaves the porous medium at a very high, approximately constant rate, which is roughly equal to the rate of evaporation from a continuous water surface under identical environmental conditions. During this initial period the temperature of the system also remains constant and, as a rule, equal to the wet-bulb temperature of the environment.

It is possible to assume the existence of a thermodynamic quasi equilibrium state, where the temperatures of gaseous, liquid and solid phases are equal, i.e., $T_S = T_L = T_G = T$ [Berger and Pei (20)].

Liquid Mass Balance:

$$\frac{\partial(\varepsilon_L \rho_L)}{\partial t} + \nabla(\rho_L \bar{u}_L) + \dot{m} = 0 \quad (1)$$

(Note - the terms of equation (1) are: storage mass of liquid in the control volume, flow of liquid mass through the system and rate of volumetric evaporation of liquid/vapor phase change).

Water Vapor Mass Balance:

$$\frac{\partial[(\varepsilon - \varepsilon_L)X_V \rho_G]}{\partial t} + \nabla(X_V \rho_G \bar{u}_G + \bar{J}_V) - \dot{m} = 0 \quad (2)$$

$$\bar{J}_V = -\rho_G (\varepsilon - \varepsilon_L) D_{\text{EFF}} \nabla X_V \quad (3)$$

(Note - the terms of equation (2) are: storage of vapor mass in the control volume, flow of vapor mass in the control volume, flow of vapor mass through the system by convection and diffusion and rate of volumetric evaporation of liquid/vapor phase change).

Air Mass Balance:

$$\frac{\partial[(\varepsilon - \varepsilon_L)X_A \rho_G]}{\partial t} + \nabla(X_A \rho_G \bar{u}_G - \bar{J}_V) = 0 \quad (4)$$

(Note - the terms of equation (4) are : storage of air mass in the control volume and flow of air mass through the system by convection and diffusion).

Liquid Momentum Equation (Darcy's Law):

$$\bar{u}_L = -\left(\frac{\alpha_G}{\mu_G}\right) \nabla(P_G) \quad (6)$$

Thermal Balance:

The thermal balance is governed by equation (7).

$$(7)$$

$$\frac{\partial\left\{\left[\rho_S C_{p_S} + (\varepsilon - \varepsilon_L)\rho_G(X_V C_{p_V} + X_A C_{p_A}) + \varepsilon_L \rho_L C_{p_L}\right]T\right\}}{\partial t} - \nabla(k_E \nabla T) + \nabla\left[\rho_L \bar{u}_L C_{p_L} + \rho_G \bar{u}_G (X_V C_{p_V} + X_A C_{p_A})\right]T + (\varepsilon - \varepsilon_L) \frac{\partial P_G}{\partial t} + \dot{m} \Delta H_V = 0$$

(Note - the terms of equation (7) are: energy storage

in the control volume, energy that moves through the system by diffusion, energy that moves through the system by convection, energy dissipation due to pressure change and heat necessary for liquid/vapor change).

Thermodynamic Equilibrium-Vapor mass Fraction:

In order to attain thermal equilibrium between the liquid and vapor phase, the vapor mass fraction should be such that the partial pressure of the vapor (P'_V) should be equal to its saturation pressure (P_{VC}) at the temperature of the mixture. Therefore, the concentration of the vapor in the air/vapor mixture inside the pores can be obtained by thermodynamic relations. According to Dalton's law of additive pressure applied to the air/vapor mixture, one can show that:

$$\rho_G = \rho_V + \rho_A \quad (8)$$

$$X_V = \frac{\rho_V}{\rho_G} \quad (9)$$

$$\rho_V = \frac{P \odot}{R_V T} \quad (10)$$

$$\rho_A = \frac{(P_G - P \odot_V)}{R_A T} \quad (11)$$

Combining equations (8) to (11), one can obtained:

$$X_V = \frac{1}{1 + \left(\frac{P_G R_V}{P \odot_V R_A}\right) - \left(\frac{R_V}{R_A}\right)} \quad (12)$$

Mass Rate of Evaporation:

The mass rate of evaporation was obtained in two different ways, as follows:

First of all, the mass rate of evaporation and was expressed explicitly by taking it from the water vapor mass balance (equation (2)), since vapor concentration is given by equation (12).

$$\dot{m} = \frac{\partial[(\varepsilon - \varepsilon_L)X_V \rho_G]}{\partial t} + \nabla(X_V \rho_G \bar{u}_G + \bar{J}_V) \quad (13)$$

Secondly, an equation to compute the mass rate of evaporation can be derived with a combination of the liquid mass balance (equation (1)) with a first - order-Arrhenius type equation. From the general kinetic equation:

$$\frac{\partial \alpha}{\partial t} = -kf(\alpha) \quad (14)$$

$$k = A \exp\left(-\frac{E}{RT_{SUR}}\right) \quad (15)$$

$$\alpha = 1 - \frac{\varepsilon_L(t)}{\varepsilon_0} \quad (16)$$

Drying Kinetics Mechanism Coupling:

In general, the drying of porous material occurs in two regimes: 1) an initial regime during which the drying rate is constant and evaporation occurs at the surface and in a manner similar to that of a free liquid surface and 2) a falling rate regime, where the porous body offers additional resistance to moisture movement. The model simulates the two mechanisms by coupling the mass rate of evaporation and coupled by the mass balance equations (1) to (4) and vapor mass fraction (equation (12)), with the mass rate of evaporation computed by equation (17), which represents these two regimes.

Using thermodynamic relations, according to Amagat's law of additive volumes, under the same absolute pressure,

$$m_V = \frac{V_V P_G}{R_V T} \quad (17)$$

$$m_A = \frac{V_A P_G}{R_A T} \quad (18)$$

$$m_V = X_V m_T \quad (19)$$

$$m_T = m_V + m_A \quad (20)$$

$$V_G = V_V + V_A \quad (21)$$

$$V_G = (\varepsilon - \varepsilon_L) V_S \quad (22)$$

Solving the set of algebraic equations (17) to (22), one can obtain the vapor-air mixture density:

$$\rho_G = \frac{(m_V + m_A)}{V_G} \quad (23)$$

$$\rho_V = \frac{m_V}{V_G} \quad (24)$$

$$\rho_A = \frac{m_A}{V_G} \quad (25)$$

Equivalent Thermal Conductivity:

It is necessary to determine the equivalent value of the

thermal conductivity of the material as a whole, since no phase separation was considered in the overall energy equation. The equation proposed by Brailsford and Major [21] was used to achieve the equivalent thermal conductivity of materials K_E , composed of a continued medium with a uniform disperse phase. It is expressed as follows in equation (26).

$$K_E = \frac{\left[k_S + \varepsilon_L k_L \left(\frac{3k_S}{2k_S + k_L} \right) + k_G (\varepsilon - \varepsilon_L) \left(\frac{3k_S}{2k_S + k_G} \right) \right]}{1 + \varepsilon_L \left(\frac{3k_S}{2k_S + k_L} \right) + (\varepsilon - \varepsilon_L) \left(\frac{3k_S}{2k_S + k_G} \right)} \quad (26)$$

$$k_G = X_V k_V + X_A k_A \quad (27)$$

Effective Diffusion Coefficient Equation:

The binary bulk diffusivity D_{AV} of air-water vapor mixture is given by [10,22]:

$$k_G = X_V k_V + X_A k_A \quad (27)$$

In leather, factor α_F can be used to account for closed pores resulting from different nature of the solid, which would increase gas outflow resistance, so the equation of effective diffusion coefficient D_{EFF} for leather drying is:

$$D_{AV} = (2.20)(10^{-5}) \left(\frac{P_{ATM}}{P_G} \right) \left(\frac{T_{REF}}{273.15} \right)^{1.75} \quad (28)$$

The convective heat transfer coefficient can be expressed as [23]:

$$D_{EFF} = \alpha_F D_{AV} \quad (29)$$

The convective heat transfer coefficient can be expressed as [23]:

$$h = Nu_\delta \left(\frac{k}{\delta} \right) \quad (30)$$

The convective mass transfer coefficient, is [24]:

$$h_M = \left(\frac{h}{C_{PG}} \right) \left(\frac{Pr}{Sc} \right)^{2/3} \quad (31)$$

$$Pr = \frac{C_{PG} \mu_G}{k_G} \quad (32)$$

$$Sc = \frac{\mu_G}{\rho_G D_{AV}} \quad (33)$$

The boundary conditions equations are shown in Table 1. The deriving force determining the rate of mass transfer inside the leather is the difference between the relative humidities of the air in the pores and the leather. The rate of moisture exchange is assumed to be proportional to the relative humidity difference in this study.

The rate constant in equation is an unknown empirical constant and the effect of this constant can be examined. The value of the rate constant was selected as $K=0.1$ and $K=10$. The resulting calculated leather surface temperatures are compared in Fig. 1.

When the rate constant is small, the evaporation rate is so small that the moisture content decreases very slowly. Initially, the surface temperature increases rapidly, but later it declines. When K is greater than 1.0, however, the effect of the rate constant on the surface temperature distribution is not significant. This indicates that when the rate constant is greater than 1.0, the evaporation rate is high and the drying process is mainly controlled by the moisture diffusion mechanism inside

the fabric. Thus we have assumed the rate constant to be 1.0 in the following calculations.

It is assumed that heat was transferred from the external hot air to the leather surface by forced convection, and moisture was transferred from the fabric to the external air. The heat transfer coefficient between external air and leather surface is obtained by [23,24]. $h = Nu_{\delta} (k / \delta)$, The mass transfer coefficient was calculated using the analogy between heat transfer and mass transfer as $h_M = (h / C_{PG})(Pr / Sc)^{2/3}$.

The temperature and moisture content were calculated using this model. In these calculations, the parameters used for the base condition are shown in table 2. The convective heat and mass transfer coefficients at the surface are important parameters in drying processes ; they are functions of velocity and physical properties of the drying medium. The transient fabric temperatures were calculated assuming two values of the rate constant. The resulting calculated fabric surface temperatures are compared in Fig. 1.

Table 1 : Boundary conditions equations

For $t = 0, \forall r, \forall x$	For $r = 0, \forall t$	For $r = R, x = 0, x = L, t > 0$
$T = T_{\infty}$	$\frac{\partial T}{\partial r} = 0$	$[K_E \nabla(T)] \bar{n} = h(T_{REF} - T) + \sigma \lambda (T_{REF}^4 - T^4)$
$X_V = X_{V0}$	$\frac{\partial X_V}{\partial r} = 0$	$\bar{J}_V = h_M (X_V - X_{REF})$
$X_A = 1 - X_V$	$\frac{\partial X_A}{\partial r} = 0$	$X_A = 1 - X_V$
$P_G = P_{ATM}$	$\frac{\partial P_G}{\partial r} = 0$	$P_G = P_{ATM}$
$\varepsilon_L = \frac{\rho_S U_0}{\rho_L}$	—	—

Table 2: Values of parameters for base condition

Parameter	Unit	Value
Dryer Temp.	K	450
Heat Transfer Coeff.	$W / m^2 K$	70
Mass Transfer Coeff.	m^2 / s	0.08
Leather Thickness	mm	1.98
Porosity	-	0.9
Initial Moisture	%RH	50
Drying Air Moisture	kg / m^3	0.02

DISCUSSION

Leathers are materials with much varied physical properties and despite the extensive research for simulation of drying of porous media, the complete validation of these models are very difficult. The drying mechanisms might be strongly influenced by parameters such as permeability and effective diffusion coefficients.

The unknown effective diffusion coefficient of vapor for leather under different temperatures may be determined by adjustment of the model's theoretical alpha correction factor and experimental data.

The transient temperatures of the surface and center of the leather using the data shown in table 2 were calculated. Fig. 2, shows that the surface and center temperature increase rapidly in the initial stage up to the saturation temperature, at which point the moisture in the fabric starts to evaporate. From that point on the difference between the surface temperature and the center temperature increases due to the different moisture contents of the surface and the center. In this stage, the leather starts to dry from the surface, and the moisture in the interior is transferred to the fabric surface. Then the moisture content decreases during drying of the fabric. Thereafter, the surface and center temperatures converge to reach the external air temperature.

The moisture variations of the surface and the center of the leather were also calculated and are shown in Fig.3.

Initially, the surface moisture content decreases rapidly, but later this rate declines because the moisture is transferred to the external air from the leather surface. The center moisture content remains constant for a short time, and then decreases rapidly, because the moisture content difference between the surface and the interior of the leather becomes large. After drying out, both the center and surface moisture contents converge to reach the external air moisture content.

The mathematical model is used to predict the effects of many parameters on the temperature variation of the leather. These parameters include the operation conditions of the dryer, such as the initial moisture content of the leather, heat and mass transfer coefficients, drying air moisture content, and dryer air temperature.

Fig. 4 shows the calculated results of the effect of the leather initial moisture content. When the initial moisture content is high, the temperature rise is relatively small and drying takes a long time. This may be because the

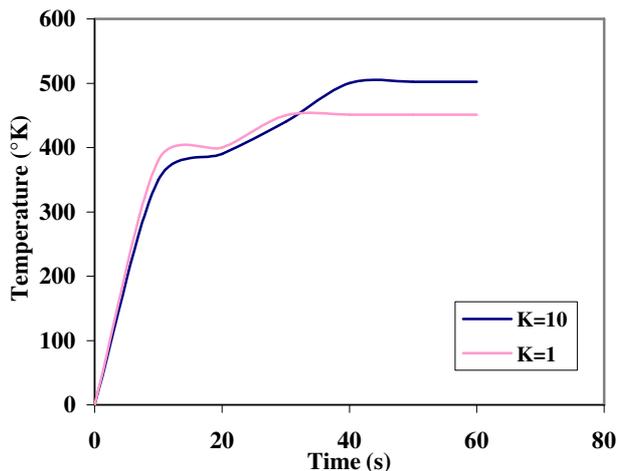


Fig. 1: Effect of rate constant on the surface temperature

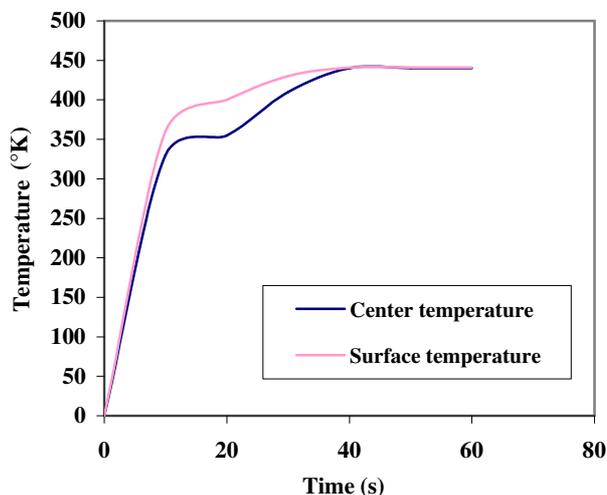


Fig. 2: Temperature variation of surface and center

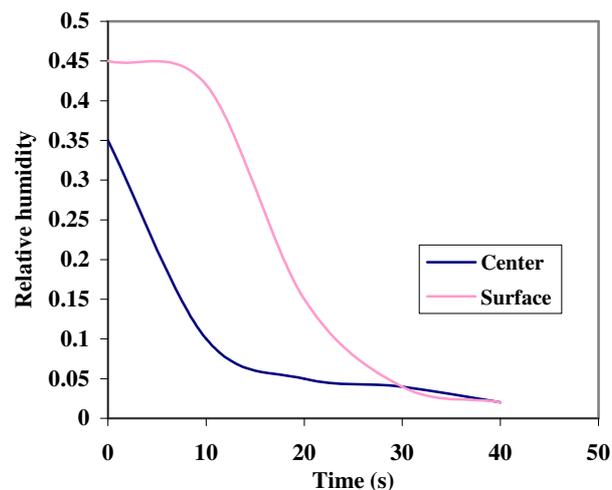


Fig. 3: Moisture content of surface and center

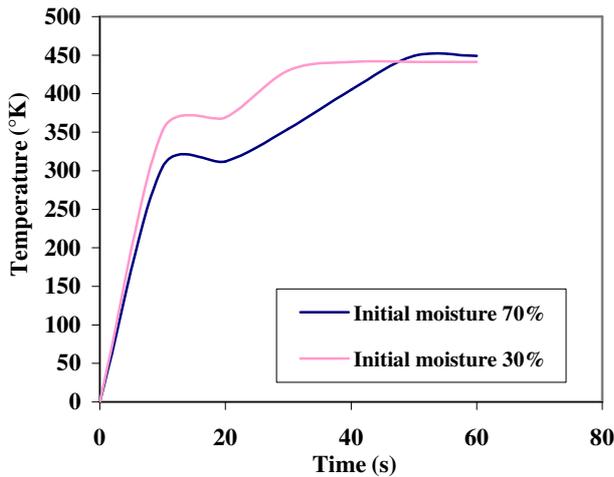


Fig. 4: Effect of initial moisture content of fabric

higher moisture content needs much more heat for evaporation from the leather. Also, the saturation temperature for higher moisture content is lower, and thus the temperature rise in the initial stage is comparatively small.

The fabric temperature was calculated to investigate the effect of heat and mass transfer coefficients in those calculation. An analogy was assumed between the heat and mass transfer, and thereby both the heat and mass transfer coefficients were determined. The calculated results are compared in Fig. 5. When the heat and mass transfer coefficients are high, the fabric temperature rise is large and the time required for drying is relatively short.

The effect of drying air moisture content, and the calculated results of the model are shown in Fig. 6. When moisture content is high, the initial temperature rise of the fabric is also high. This may be because the saturation temperature in the initial stage largely depends on the drying air moisture content. After the initial temperature rise, however, the temperature increase is relatively small, and thus the time required for complete drying is comparatively long.

The effect of dryer air temperature was also investigated and the calculation results are shown in Fig. 7. When the dryer air temperature is high, the temperature rise of the leather is large.

When such a wet material is dried by a convective medium, three drying rate periods are often observed, the constant rate period, the first falling rate period and the

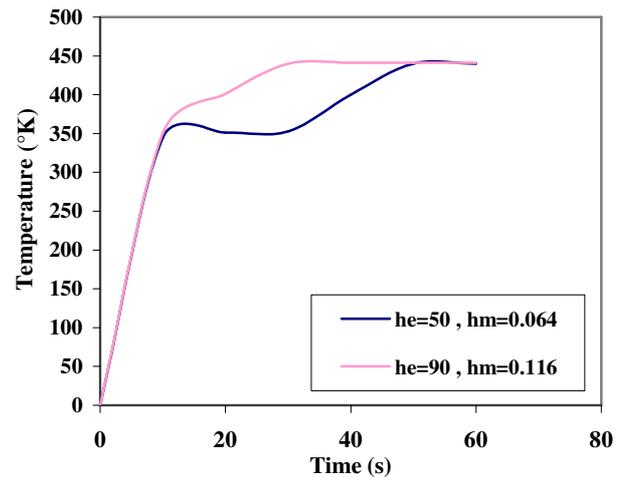


Fig. 5: Effect of heat and mass transfer coefficients

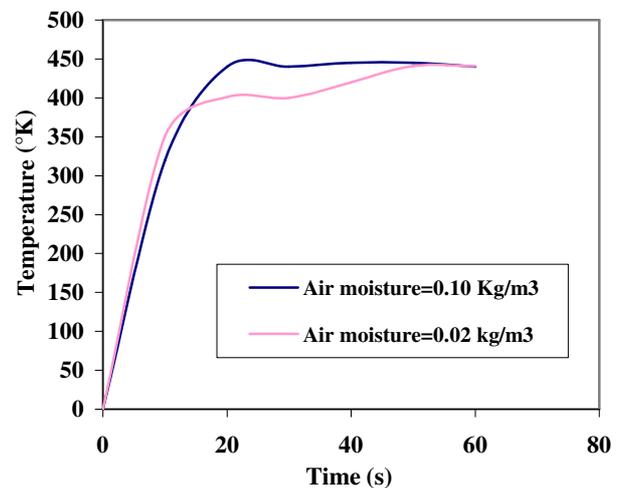


Fig. 6: Effect of drying air moisture content

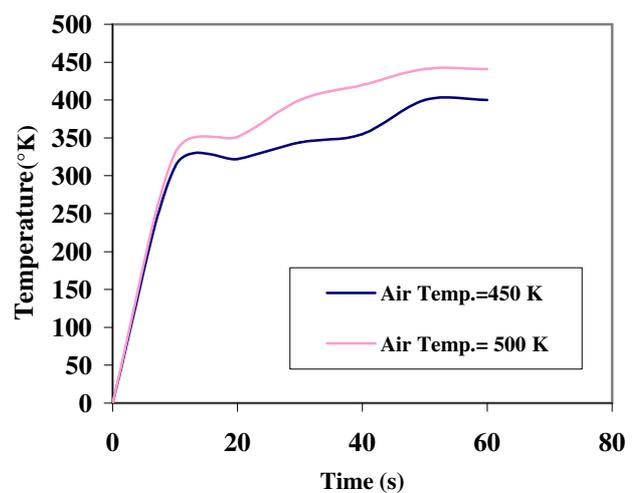


Fig. 7: Effect of dryer air temperature

second falling rate period. In the constant rate period and the first falling rate period, the material remains wet, and the model of the wet region only used. Since evaporation takes place almost entirely at the surface, the drying rate is controlled by the convective heat and mass transfer. If the temperature gradient within the material is negligible, the surface temperature is almost constant and its value is very close to wet bulb temperature of the flowing air.

The evaporation temperature is directly proportional to parameter changes in the drying agent. Thus, the greater the drying flow velocity, the greater the heat change by convection between the drying agent and the sample.

CONCLUSION

The aim of this study was to describe heat and mass transfer during drying of leather by forced convection. The mathematical model developed in this study is comprehensive and can be used to predict transient variations in the temperature and moisture content distributions in wet leather during the drying process with reasonable accuracy. Simplistic assumptions have been avoided, especially insofar as the effect of gaseous pressure is concerned. The effect of temperature and humidity of the dryer, the initial moisture content of leather and heat and mass transfer coefficients can be predicted. With such predictions, it is possible to reduce energy consumption by optimizing the drying conditions of the dryer.

On the other hand, for a fully wetted surface, the areas for the heat and mass transfer temperature is close to the wet bulb temperature ; for a partly wetted surface, the effective area for mass transfer decreases with the surface moisture content.

It was noted that the intercellular spaces, like the voids are interconnected and filled with air and a certain amount of free water. The cells themselves also contain water, which is also defined here as bound water.

Finally, when the material is exposed to a convective surface condition, three main mechanisms of internal moisture transfer are assumed to prevail: capillary flow of free water, movement of bound water and vapour transfer. If the initial moisture content of the material is high enough, the surface is covered with a continuous layer of free water and evaporation takes place mainly at the surface.

The model appears to be a simple way of predicting the efficiency of the process. It opens the possibility for further investigations on the relationships between model parameters and drying conditions.

Notifications and Notional Conventions

A	Arrhenius equation pre-exponential factor , 1/s
C_{PG}	specific heat of the gas phase, kJ/kg.K
C_{PL}	specific heat of the liquid, kJ/kg.K
C_{PS}	specific heat of the solids, kJ/kg.K
D_{EFF}	effective diffusion of gas, m ² /s
E	activation energy, kJ/kmol
f(α)	kinetic mechanism function , kg/kg
h	heat transfer coefficient , W/m ² .K
h_M	convective mass transfer coefficient, kg /m ² .s
\bar{J}_V	diffusive term of the vapor concentration, kg /m ² .s
K	drying rate , 1/s
k	thermal conductivity of the air, W/m.K
k_E	equivalent thermal conductivity, kJ/m.s.K
k_G	equivalent thermal conductivity of the gas, kW/m.K
k_L	thermal conductivity of the liquid, kW/m.K
k_S	thermal conductivity of the solids , kW/m.K
L	sample height
\dot{m}	rate of mass evaporation per unit volume, kg /m ² .s
m_A	air mass , kg
m_T	total vapor-air mixture mass , kg
m_V	water vapor mass , kg
\bar{n}	unit vector normal to surface
Nu_δ	Nusselt number
P_{ATM}	atmospheric pressure , 1×10^{-5} Pa
P_{VS}	saturation pressure of the vapor , Pa
P_V	partial pressure of the vapor , Pa
Pr	Prandtl number
R	universal gas constant, 8.314 kJ/kg.K
R_A	air equivalent constant , kJ/kg.K
R_V	vapor equivalent constant, kJ/kg.K
r	radial linear dimension , m
Sc	Schmidt number
T	temperature , K
T_{REF}	oven drying temperature , K
T_{SUR}	temperature of evaporation , K
T_∞	ambient temperature
t	time, s

U_0	initial moisture, kg liq/kg sol
\vec{u}_G	velocity vector of the gas mixture, m/s
\vec{u}_L	velocity vector of the liquid, m/s
V_A	volume filled by air, m^3
V_G	volume filled by the vapor-air mixture, m^3
V_S	total volume of the solids, m^3
V_V	volume filled by vapor, m^3
X_A	concentration of air in the air/vapor mixture, kg air/kg air-vapor mixture
X_{REF}	vapor mass fraction of drying agent
X_V	concentration of vapor in the air/vapor mixture, kg vapor/kg air-vapor mixture
X_{V0}	initial vapor concentration
x	axial linear dimension, m
α	evaporated mass fraction, kg/kg
α_G	directional permeability of the gas phase, m^2
ϵ	sample porosity, m^3 of empty pores/ m^3 solid
ϵ_0	initial liquid to total volume ratio, m^3 liq / m^3 sol
ϵ_L	liquid to total volume ratio, m^3 liq / m^3 sol
ρ_A	density of the air, kg / m^3
ρ_G	density of the gas mixture, kg / m^3
ρ_L	density of the liquid, kg / m^3
ρ_s	density of the solids, kg / m^3
ρ_V	density of the vapor, kg / m^3
μ_G	dynamic viscosity of the gas phase, N.s / m^2
ΔH_V	vaporization enthalpy of the liquid, kJ/kg
δ	annular free space between concentric cylinder, m
λ	latent heat of evaporation, kJ/kg

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