Moisture, acidity and temperature evolution during cacao drying

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Abstract

Theoretical moisture and temperature evolution during cacao fixed bed drying were estimated with non-steady heat and mass transfer equation both in particle and bed. The model takes into account the cacao internal profiles and the air macroscopic balance of moisture, acidity and temperature. The results showed that the theoretical model predicts the experimental evolution of these state variables during cacao drying at three different conditions with average % of error lower than 18%.

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1. Introduction

The post-harvest process of cocoa beans consists of gathering, opening, fermentation, drying, selection and storage. Fermentation and drying are the principal operations for the curing of cocoa beans.

Volatile acidity formed during cocoa beans fermentation reaches approximately 2% of the dry basis. This acidity is the result of some components such as acetic, propionic, butric, isobutric, and isovaleric acids. 90% of these components is acetic acid, which has an important role in the catalysis of enzymatic reactions for producing components of desirable sensorial characteristics (Barel, 1997; Biehl, Passern, & Wilfried, 1982). A not fermented cocoa does not develop the aroma characteristics and it is more bitter and astringent. After fermentation, a drying process is adopted with the purpose of reducing moisture content (from 55–60 to 8%) and volatile acidity content for stopping undesirable reactions and the oxidation of polyphenol. The aromatic components are hidden for volatile acidity excess.

(Faborode, Favier, & Ajayi, 1995; Jaquet, Vincent, Hahn, & Lotode, 1980; Jinap, Thien, & Yap, 1994). Studies on the kinetic behavior during the artificial drying of cocoa beans are scarce. Jinap et al. (1994) studied different types of drying conditions evaluating the acidity and volatile fatty acids and concluded that beans dried in an oven at 60 °C retain a high content of acetic, propionic, butric, isobutric, and isovaleric acids, which helps in the making of causes a low quality chocolate.

In order to solve the problem of acidity at the end of cacao drying, it is necessary to study the acidity mass transfer properties during drying. Augier, Nganhoun, Benet, Berthomieu, and Barel (1999) reported an experimental study of mass transfer of acetic acid and showed the complexity of elimination under different operational conditions in convective drying, stating the differences that exist in concentration levels in testa-nib. Wan Daud, Meor Talib, and Hakimi (1998), Carrère-Gée, Lecomte, Fotso, and Fudym (1996), Fotso, Lecomte, Pomathios, and Nganhoun (1994), Nganhoun, Lecomte, and Dumargue (1992) studied the heat and mass transfer during cocoa beans drying, but they did not take into account the volatile acidity as state variable.
On the other hand, cacao drying is a type of grain drying. Grain drying theory was reported since 40 years ago (Barre, Baughman, & Hamdy, 1971; Henderson & Pabis, 1961, 1962; Spencer, 1969). The models proposed by these first authors have been used practically without changes by the modern researcher (Barrozo, Murata, & Costa, 1998; Bruce & Giner, 1993; Torrez, Gustafsson, Schreil, & Martínez, 1998). All of them are based in four differential equations (for product moisture, product temperature, air moisture and air temperature), in which the water mass transfer is represented by an empirical expression. Some authors presented a theoretical analysis over the relation of this empirical expression with respect to water diffusivity in the grain (Barre et al., 1971; Barrozo et al., 1998; Henderson & Pabis, 1961, 1962; Spencer, 1969), while others reduce the analysis to the determination of an empirical parameter (Bruce & Giner, 1993; Torrez et al., 1998). A mechanistic approach of deep-bed drying (not specific for grains) has been suggested by Herman-Lara, Salgado-Cervantes, and García-Alvarado (2005), in which each one of the differential equation is a function of specific heat or mass transfer coefficient, and each coefficient depends upon the transfer mechanism.

Resuming, there is not reported a general model that takes into account water, acidity and heat transfer during cacao drying. Such model would be useful in the search of process conditions that produce the desired contents of moisture and volatile acidity in the dried cacao with minimal energy resources. The model must be mechanistic, similar to that of Herman-Lara et al. (2005), so that it need not depend of an empirical parameter and therefore it can be generalized at different conditions, but with the inclusion of the cacao skin. Therefore, the objective of this work was to build the model described, and validate it experimentally.

2. Model building and analysis

Cacao artificial drying is done in convective dryers with hot air as the drying medium. In this situation, one of the
most general models for the description of air state was proposed by Herman-Lara et al. (2005). This model, extended for acidity mass balance, is
\[
dX_{\text{aig}} \frac{dt}{\varepsilon} = k_{\text{aig}}a(X_{\text{aig}} - X_{\text{aig}}) - \frac{G_{i}(X_{\text{aig}} - X_{\text{aig-1}})}{V_{\rho_{\text{aig}}}^{2}} \tag{1}
\]
\[
dX_{\text{aig}} \frac{dt}{\varepsilon} = k_{\text{aig}}a(X_{\text{aig}} - X_{\text{aig}}) - \frac{G_{i}(X_{\text{aig}} - X_{\text{aig-1}})}{V_{\rho_{\text{aig}}}^{2}} \tag{2}
\]
\[
dT_{ij} \frac{dr}{\varepsilon} = h_{i}(T_{ij} - T_{ij}) + \frac{k_{i}a(X_{\text{aig}} - X_{\text{aig}})\Delta H_{\text{as}}}{(C_{p_{i}} + C_{\rho_{\text{aig}}}X_{\text{aig}})\varepsilon} + \frac{k_{i}a(X_{\text{aig}} - X_{\text{aig}})\Delta H_{\text{as}}}{(C_{p_{i}} + C_{\rho_{\text{aig}}}X_{\text{aig}})\varepsilon}
- \frac{H_{\text{in}}^{0} + C_{\rho_{\text{aig}}}T_{ij}dX_{\text{aig}}}{C_{p_{i}} + C_{\rho_{\text{aig}}}X_{\text{aig}}} \left[ \frac{h_{\text{out}}A_{\text{out}}(T_{ij} - T_{\text{out}})}{(C_{p_{i}} + C_{\rho_{\text{aig}}}X_{\text{aig}})\varepsilon p_{j}V_{j}} - \frac{G_{i}}{(C_{p_{i}} + C_{\rho_{\text{aig}}}X_{\text{aig}})\varepsilon p_{j}V_{j}} \right]
- \frac{C_{p_{i}}T_{ij-1} + (H_{\text{in}}^{0} + C_{\rho_{\text{aig}}}T_{ij-1})X_{\text{aig-1}}}{(C_{p_{i}} + C_{\rho_{\text{aig}}}X_{\text{aig}})\varepsilon} \right] \tag{3}
\]
Eqs. (1) and (2) are the mass balances for water and acidity in a complete mixed unit j of drying air. Eq. (3) is the energy balance in a complete mixed unit j of drying air. In Eq. (3) the evaporation of latent heat of water and acidity (second and third terms of right side), and energy losses to the surroundings (fifth term of right side) are considered.

Herman-Lara et al. (2005) model represents the product state variables with other macroscopic heat and mass balances equations. However, we need to describe the cacao skin and the internal acidity and moisture. Therefore, the cacao state variables must be represented with microscopic heat and mass balances. Ruiz-Lopez, Cordova, Rodriguez, and Garcia (2004), and Maroulis, Kiranoudis, and Marinou-Kouris (1995) proposed the most general form for the representation of microscopic heat and mass balances during food drying of a single particle. However, they considered only water and heat flux, and constant air conditions. Generalizing their concepts in order to take into account acidity flux, a mass transfer resistance in cocoa skin, and the general balance with the finite air flow, the following results are obtained:
\[
(1 - \varepsilon) \frac{\partial(p_{\text{aig}}X_{\text{aig}})}{\partial t} = \frac{\partial}{\partial z} \left[ D_{\text{aig}} \frac{\partial(p_{\text{aig}}X_{\text{aig}})}{\partial z} \right] aL \quad \text{at } t > 0 \text{ in } 0 \leq z \leq L \tag{4}
\]
\[
- D_{\text{aig}} \frac{\partial(p_{\text{aig}}X_{\text{aig}})}{\partial z} = k_{\cos}p_{\text{aig}}[X_{\text{aig}} - X_{\text{aig}}] \quad \text{at } t > 0 \text{ in } z = L \tag{5}
\]
\[
k_{\cos}p_{\text{aig}}[X_{\text{aig}} - X_{\text{aig}}] = k_{\text{aig}}p_{\text{aig}}[X_{\text{aig}} - X_{\text{aig}}] \quad \text{at } t > 0 \text{ in } z = L + l \tag{6}
\]
\[
X_{\text{aig}} = X_{\text{aig}} \quad \text{at } t > 0 \text{ in } z = L \tag{7}
\]
\[
X_{\text{aig}} = \frac{a_{i}p_{i}}{1 - a_{i}p_{i}P_{\text{aig}}} \quad \text{at } t > 0 \text{ in } z = L + l \tag{8}
\]
\[
D_{\text{aig}} \frac{\partial(p_{\text{aig}}X_{\text{aig}})}{\partial z} = 0 \quad \text{at } t > 0 \text{ in } z = 0 \tag{9}
\]
in all equations. \( \omega = w \) and \( a \)

Eq. (4) represents the moisture or acidity movement in the interior of cocoa during drying. Eq. (5) represents the moisture or acidity transportation at the interface product-skin. Eq. (6) implies that in skin the accumulation is negligible, and therefore the water flux between product-skin interface is equal to skin-air interface. Eq. (7) states that distribution constant between \( \beta \) and \( \delta \) phases is one. Eq. (8) represent the sorption isotherms for water and acidity between cocoa and air. Finally, Eq. (9) states the symmetry of mass transfer with respect to cocoa particle center. For heat transfer the equations are
\[
(1 - \varepsilon) \frac{\partial(p_{\text{aig}}C_{p_{\text{aig}}}T_{\text{aig}})}{\partial t} = \frac{\partial}{\partial z} \left[ k_{\text{aig}} \frac{\partial T_{\text{aig}}}{\partial z} \right] aL \quad \text{at } t > 0 \text{ in } 0 \leq z \leq L \tag{10}
\]
\[
-k_{\text{aig}} \frac{\partial T_{\text{aig}}}{\partial z} = h_{b}(T_{\text{aig}} - T_{\text{air}}) \quad \text{at } t > 0 \text{ in } z = L \tag{11}
\]
\[
h_{b}(T_{\text{aig}} - T_{\text{air}}) = h_{b}(T_{\text{aig}} - T_{\text{air}}) \quad \text{at } t > 0 \text{ in } z = L + l \tag{12}
\]
\[
T_{\text{aig}} = T_{\text{air}} \quad \text{at } t > 0 \text{ in } z = L \tag{13}
\]
\[
k_{\text{aig}} \frac{\partial T_{\text{aig}}}{\partial z} = 0 \quad \text{at } t > 0 \text{ in } z = 0 \tag{15}
\]

Eq. (10) represents the temperature evolution in the interior of the dried product. Eq. (11) represents the interfacial heat exchange in product-skin contact surface. Eq. (12) expresses that the heat transferred by conduction inside the food is the difference between the heat transferred by convection and the heat required for water and acetic acid phase change. Eqs. (13) and (14) imply that the skin heat transfer resistance is considered negligible. Finally, Eq. (15) states that heat transfer is symmetric with respect to cocoa particle center.

The terms \((1 - \varepsilon)\) and \(aL\) included in the left and right hand side of Eqs. (4) and (10) are necessary to satisfy the mass and heat balance of the process. If the average concentrations of water, acidity, and temperature are defined as
\[
\langle X \rangle_{\text{aig}} = \int_{0}^{L} X_{\text{aig}} dz/L, \quad \langle T \rangle_{\text{aig}} = \int_{0}^{L} T_{\text{aig}} dz/L \tag{16}
\]
then, Eq. (4) may be expressed as
\[
(1 - \varepsilon) \frac{\partial}{\partial t} \left[ \langle X \rangle_{\text{aig}} \right] = \int_{0}^{L} \left[ D_{\text{aig}} \frac{\partial}{\partial z} \langle X \rangle_{\text{aig}} \right] aL \tag{17}
\]
and by Eqs. (5), (6) and (9),
\[
(1 - \varepsilon) \frac{\partial}{\partial t} \left( \langle X \rangle_{\text{aig}} \right) = -k_{\text{aig}}p_{\text{aig}}X_{\text{aig}} \quad \text{at } t > 0 \text{ in } z = L + l \tag{18}
\]
substituting in Eq. (1) or Eq. (2),
\[
V_{\text{j}}\rho_{\text{aig}}c_{\text{aig}} \frac{dx_{\text{aig}}}{dt} = -V_{\text{j}}(1 - \varepsilon) \frac{\partial}{\partial t} \left( \langle X \rangle_{\text{aig}} \right) - G_{\text{j}}(X_{\text{aig}} - X_{\text{aig-1}}) \tag{19}
\]
that represents the two phase mass balances for water or acidity. Similarly, for heat transfer,
\( (C_p + C_{pw,x}X_{w_{ij}}) \epsilon_p \rho_j \frac{dT_{ij}}{dt} = -V_j (1 - \varepsilon) \frac{d}{dt}(\rho_j C_p(T_{ij}^0)) - (H_{wv}^0 + C_{pw,x} T_{ij}^0) \epsilon_p \rho_j \frac{dX_{w_{ij}}}{dt} \)

\( = -V_j (1 - \varepsilon) \frac{d}{dt}(\rho_j C_p(T_{ij}^0)) - (H_{wv}^0 + C_{pw,x} T_{ij}^0) \epsilon_p \rho_j \frac{dX_{w_{ij}}}{dt} \)

\( - h_{out} A_{out} (T_{ij} - T_{out}) - G_r \left\{ \left[ C_p, T_{ij}^0 + (H_{wv}^0 + C_{pw,x} T_{ij}^0) X_{w_{ij}} \right] \right\} \)

\( - C_p, T_{ij-1}^0 + (H_{wv}^0 + C_{pw,x} T_{ij-1}^0) X_{w_{ij-1}} \) \}

Eqs. (1)–(16) represent a general and mechanistic model for water, acidity and energy transfer during cocoa convective drying in a series of complete mixed units. This model takes into account the main mechanism of mass and heat transfer and assures consistency of heat and mass balances.

### 3. Experimental

The drying experiments were performed at Instituto Tecnológico de Villahermosa at Tabasco Mexico during the October 1996–May 1997 cocoa season. Some of these results were presented in a previous work (Augier et al., 1999). Fermented cocoa beans were dried in a convective dryer equipped with automatic regulation for air velocity, temperature and relative humidity. The samples to be dried were placed in a 19 cm diameter and 26 cm high cylindrical dried chamber. The air drying variables (velocity, relative humidity and temperature) were measured with the electronic sensors of the equipment. The evolution of product humidity and temperature were measured with the electronic sensors of the equipment. The beans moisture was evaluated by weight loss measured with an automatic balance of the dryer. The volatilic acidity was extracted in agreement with Mothe–Efter method. More details of the experimental procedure may be in the previous work (Augier et al., 1999). The process variables of these experiments were listed in Table 3 as conditions 1–3.

### 4. Model solving

The model (Eqs. (1)–(15)) was solved by the method of lines using a longitudinal scheme with central finite differences for space derivatives as suggested by Ruiz-Lopez et al. (2004). This scheme produces an ordinary differential equation (ODE) system with \( t \) as independent variable. Time derivatives were treated as,

\[
\frac{\partial(\rho X)}{\partial t} = \frac{\partial X}{\partial t} + \frac{\partial \rho_j \frac{\partial X}{\partial X}}{\partial t} \quad \text{(17)}
\]

The water and acidity diffusivities inside cocoa bean were estimated from experimental profiles reported by Augier (1999) and Augier et al. (1999). The models obtained were

\[
D_{wb} = e^{(-10.24 - 4176.024/T_e)c_1 - 0.00034985 X_{w_{ij}} \epsilon} \quad \text{for } T \text{ in K} \quad \text{(18)}
\]

\[
D_{ab} = D_{wb}[1 - e^{-5 X_{w_{ij}}}] \quad \text{(19)}
\]

The thermophysical properties of cacao were considered as common for food products (Ruiz-Lopez et al., 2004),

\[
k_p = 0.49 - 0.443 e^{0.206 X_{w_{ij}}}, \quad C_p = 1.755 + 2.345 X_{w_{ij}}/[1 - X_{w_{ij}}] \quad \text{(20)}
\]

The sorption isotherms were obtained in the laboratory. The results were

\[
a_w = 1 - e^{-100.16784 X_{w_{ij}}^{0.9936}}, \quad a_a = 1 - e^{-1.177673 \times 10^{-4} X_{w_{ij}}^{0.4521}} \quad \text{(21)}
\]

Air and water properties were taken from Geankoplis (1978),

\[
H_{wv}^0 = 2501.4 \, J \, g^{-1}, \quad C_{pw,x} = 1.60892 \, J \, g^{-1} \, C^{-1},
\]

\[
C_{pw,x} = 4.185 \, J \, g^{-1} \, C^{-1}, \quad C_p = 1.0 \, J \, g^{-1} \, C^{-1}
\]

The mass transfer coefficient \( k_{ev} \) was calculated with the following correlation suggested by Geankoplis (1978) for fixed beds:

\[
J_D = J_H = 0.4548 Re^{-0.469} / \varepsilon, \quad \text{(22)}
\]

\[
k_{cm} = J_D G_f/(\rho_a d_A/\mu) S c^{-2/3}, \quad h_f = J_H C_p G_f/\mu_a S c^{-2/3} \quad \text{(23)}
\]

where

\[
Re = \frac{D p (G_f/A_{tra})}{\mu_f}, \quad Sc = \frac{\mu_f}{D p \gamma f}, \quad \frac{D_p}{a} = 6 (1 - \varepsilon)
\]

The skin mass transfer coefficients were estimated from,

\[
k_{m} = G_f/\sqrt{4l} \quad \text{for } \omega = w \text{ or } a \quad l = 1 \times 10^{-5} \, m
\]

The moisture content and volatile acidity were reported as dimensionless form,

\[
\Psi = (X_e)_{w_{ij}} - X_{w_{ij}} \quad \text{for } \omega = w \text{ and } a
\]

<table>
<thead>
<tr>
<th>Condition</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
</tr>
</thead>
<tbody>
<tr>
<td>( X_{w_{ij}} ) (g g dm)</td>
<td>1.22</td>
<td>1.12</td>
<td>1.03</td>
<td>1.12</td>
<td>1.12</td>
</tr>
<tr>
<td>( T_{ij}^0 ) (C)</td>
<td>36</td>
<td>38</td>
<td>38</td>
<td>36</td>
<td>36</td>
</tr>
<tr>
<td>( X_{w_{ij}} ) (g g dm)</td>
<td>0.023</td>
<td>0.0394</td>
<td>0.075</td>
<td>0.027</td>
<td>0.027</td>
</tr>
<tr>
<td>( X_{w_{ij}} ) (g g dm)</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
<td>0.0</td>
</tr>
</tbody>
</table>
The equilibrium concentrations were calculated with Eq. (21) at air input conditions. In the case of acidity the equilibrium concentration is zero.

5. Results and discussion

In Fig. 1 is plotted the dimensionless moisture simulated at conditions described in Table 1 together with experimental ones. The equilibrium concentrations of moisture and acidity are listed in Table 2. It is important to note that the simulated kinetics are not fit. The properties were obtained from independent data. The moisture and acidity diffusivities (Eqs. (18) and (19)) were estimated from concentration profiles (Augier et al., 1999) in the interior of cacao beans during drying runs at different conditions that are listed in Table 1. The rest of the heat and mass transfer were calculated from published data. Therefore, in Fig. 1 the capacity of the model to predict the cacao drying kinetic is shown. The quantification of this capacity was calculated with the average % or error,

\[ \%E = \frac{\sum_{i=1}^{n} |\phi_{\text{sim}} - \phi_{\text{exp}}|}{\phi_{\exp}} \times 100/n \]  

(24)

and its 95% confidence interval,

Confidence interval = \( \%E \pm \frac{s}{\sqrt{n}} t_{1-0.05/2(n-1)} \)  

(25)

The average % of error obtained for moisture at conditions 1–3 (Table 1) is listed in Table 3.

The temperature evolution, both simulated and experimental, is plotted in Fig. 2. The prediction is acceptable, but some deviations may be appreciated at the beginning of the process. These deviations may be explained because the model was solved under complete mixed assumption, and the temperature evolution has the shape of plug flow (Herman-Lara et al., 2005). The plug flow assumption in the proposed model (Eqs. (1)–(23)) would require \( j = N \) ideal mixed steps, which represent a considerable numerical effort. In such case, simplification of microscopic equations (Eqs. (4)–(15)) to average equations like the ones used by Herman-Lara et al. (2005) is more convenient. This analysis will be reported in another manuscript. However, the average % of error obtained until 5 h; (Fig. 2) for the temperature was lower than 5% (Table 3).

With respect to acidity evolution during drying, Augier et al. (1999) and Augier (1999) report inconsistencies. That is, in some experiments can be appreciated acidity losses and in others (at similar conditions) cannot be observe acidity losses. These results may be explained in terms of slow rate of acidity transport. In order to take in account this effect, the acidity diffusivity described by Eq. (19) was proposed. This diffusivity decreased to one fifth of the water, when the moisture content is lower than 0.1. Therefore, the acidity transport rate decrease. As it can be observed in Fig. 3, the skin moisture reaches a value of 0.1 in 1 h of drying and therefore the skin acts as a barrier for acidity. However, as shown in Fig. 4 the model predicts enough acidity losses to produce an acidity profile in the interior of cacao beans. Therefore, an acidity kinetic exists during drying (Fig. 5). Summarizing, the model predicts that even with a small diffusivity, the acidity can evaporate from the interface of cacao skin-air, but only 40% of the initial acidity can be removed after 22 h; at 80 °C.
However, these results are not conclusive because the acidity drying kinetics reported by Augier et al. (1999) and Augier (1999) are not enough. More experimental data of acidity evolution are necessary during cacao drying. On the other hand, the objective of the model is the simulation of the process at different conditions in order to increase the efficiency of process. The proposed model was used for the simulation of cacao beans drying at conditions 4 and 5 of Table 1. These conditions were stated closer in a manner that saves energy and removes acidity. From Fig. 5, it is evident that the better state with respect to the removal of acidity is condition 3. However, the air input variables (80 °C and 0.075 g water per g dry air) have an enthalpy of 280.24 J g⁻¹. Then, if the environmental air was assumed as typical in a tropical zone (35 °C and 75% of relative humidity), then it has an enthalpy of 105.5 J g⁻¹. Therefore, condition 3 (at 150 g s⁻¹) requires at least 26,211 J s⁻¹. Conditions 4 and 5 were stated closer...
than condition 3 but with slower energy consumption. In condition 4, the air mass flow and temperature are the same as that in condition 3, but the air moisture content is environmental (in 3 the air must be humidified). The air enthalpy at condition 4 is 152.68 J g⁻¹ and the energy consumption is 7077 J s⁻¹. In condition 5, the air moisture is environmental, the temperature is decreased (70 °C) and the air mass flow is increased (170 g s⁻¹). At this state, the air enthalpy is 142 J g⁻¹ and the energy consumption is 6205 J s⁻¹. The simulation results of conditions 4 and 5 are plotted in Figs. 6–8. Fig. 6 shows that conditions 4 and 5 dry the cacao at a greater rate than condition 3, and Fig. 8 shows that condition 4 removes more acidity than condition 3. However, condition 4 produces greater temperature in the cacao beans during drying (Fig. 7), which can produce quality losses in the dry cacao. Therefore, in these examples condition 5 is better because it keeps the cacao temperature lower than 70 °C, removes acidity at a similar rate to condition 3, but has an energy saving of 77% with respect to condition 3, and does not require prior air humidification.

6. Conclusions

It was shown that the mechanistic model proposed in Eqs. (1)–(23) can reproduce experimental evolution of moisture and temperature during cacao drying with a maximum average error of 17.3% for moisture and 4.15% for temperature. This proposed model can be used for cacao drying optimization with respect to energy consumption subject to moisture and acidity contrictions. However, more experimental data of acidity kinetic during cacao drying are required in order to obtain a better representation of acidity diffusivity.

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