Adjustment of Mathematical Models and the Quality of Drying the Pulped Coffee at Different Air Conditions

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Abstract: The aim of the study was to describe the drying kinetics of washed coffee (Coffea arabica L.) and evaluate the best mathematical model to fit the experimental drying data conducted with different air humidity (40%, 50% and 60%), temperatures (23, 40 and 60 °C) and the quality of the coffee. The cherries coffee were separated and standardized in the processes of washing, mechanical and manual separation. Then, approx. 85 kg of coffee cherries were pulped and taken directly to the yard. The washed coffee was completed dried in a mechanical dryer and yard. The results showed that the different conditions of the ambient air significantly influenced the processes of drying. The water content of the hygroscopic equilibrium of pulped coffee is directly proportional to the water activity and relative humidity (RH), decreasing with increasing temperature, for the same value of equilibrium. The Oswin model was best represented by the hygroscopicity of the pulped coffee, while the Midilli model shows the best fit to describe the drying curves of the washed coffee. The effective diffusion coefficient increases with increasing temperature of the drying air and reducing of RH, being described by the Arrhenius equation. Electrical conductivity, potassium leaching, total titratable acidity and grease acidity increase with increasing drying temperature regardless of the type of processing. Reducing sugars, total sugars and the sensorial quality was negatively affected with increasing drying temperature regardless of the type of processing. The drying at 60 °C/40% RH negatively affected the coffee quality.

Key words: Coffea arabica L., drying, mathematical modeling, pulped coffee.

1. Introduction

There are several factors that influence the final quality of the coffee, as soil and climate characteristics, cultivars, driving and crop management, harvesting, processing, drying and storage [1, 2]. There are various forms of processing that result in major differences in the sensory attributes and there are common reports of superiority to coffee peeled and pulped and in relation to natural coffee [3, 4]. Drying is one of the most important stages in the processing of coffee, both from the standpoint of energy consumption and the influence this has on the operation quality of the final product. It is sought to control the drying parameters, such as drying air temperature, grain temperature, relative humidity (RH) and air flow, in order to minimize adverse conditions to the product and at the same time to obtain a higher yield in the process. On the other hand, if the best drying techniques are not used, the quality of the coffee can be damaged in the physical, chemical and sensorial aspects [1, 2]. The drying of agricultural products, thin layer, has the purpose of determining the rates of drying of the product using for data collection recording the mass loss occurred in a sample during water removal [3, 4].

Thus, the drying curves, thin layer, vary with species, variety, environmental conditions, methods staging post-harvest, among other factors.
Accordingly, various mathematical models have been used to describe the drying of agricultural produce, although in most cases, the semi-empirical relationships and empirical have been shown to predict the best options for the drying of grains and seeds, although its validity is restricted to the conditions under which the experimental data were obtained [4, 5]. These models generally are based on variables external to the product, such as the temperature and RH of the drying air. The semi-empirical equations are based on Newton’s law of cooling heat transfer by convection, assuming that during the drying conditions are isothermal and that the water transfer is restricted to the surface of the product.

Thus, the aim of the study was to describe the drying kinetics of washed coffee (Coffea arabica L.), evaluate the best mathematical model to fit the experimental drying data conducted with different air humidity (40%, 50% and 60%) and temperatures (23, 40 and 60 °C) and the quality of the coffee.

2. Materials and Methods

The work was conducted at the Department of Engineering and Technology Center of Post-Harvest Coffee, Federal University of Lavras. The coffee was harvested manually and selectively removing only the cherry fruit from the plant. For each repetition, 800 kg of the coffee variety Topazio were collected. All the raw materials were standardized by the washing, separation and manual selection of green coffees, green canes passes and buoy (Fig. 1). About 85 kg of coffee cherries were pulped and taken directly to the yard. The pulped coffee was divided into distinct segments in the yard, remaining for 3 d, with an average temperature of 23 °C for conditions of (40%, 50% and 60%) ± 5% RH. Then a portion of the batch of coffees were taken for mechanical drying with 40 ± 2 °C air temperature under the conditions of (40%, 50% and 60%) ± 5% RH and another portion of coffees subjected to mechanical drying with air temperature of 60 ± 2 °C for the conditions of (40%, 50% and 60%) ± 5% RH.

During the time that the coffee remained in the yard, turnings were made every half hour and monitoring the temperature and RH of the ambient air using a term hygrograph. Mechanical drying was conducted

![Fig. 1 Processing and drying of natural coffee in mechanical dryer and yard.](image-url)
**Table 1** Semi-empirical and empirical equations for mathematical modeling of the grains coffee.

| Equations Models                                      |
|-------------------------------------------------------|
| RU = exp(-kt)                                         |
| RU = exp(-kt^n)                                        |
| RU = aexp(-kt)                                         |
| RU = aexp(-kt) + c                                    |
| RU = aexp(-kt) + bexp(-kt)                            |
| RU = 1 + at + bt^2                                    |
| RU = aexp(-kt) + bexp(-kt) + cexp(-kt)                |
| RU = aexp(-kt) + (1 - a)exp(-ktb)                     |

RU: moisture ratio, dimensionless; t: drying time (h); k, k_o, k_1: drying constant (h^-1); a, b, c, n: model coefficients.

...on two prototypes of fixed layer. To determine the airflow in the dryer a graduated aperture was made at the fan inlet. The determination of the water content was performed by standard oven at 105 ± 3 °C for 24 h [6].

The drying curves were fitted to the experimental data using different semi-empirical and empirical equations discriminated in Table 1.

For determining the ratios of moisture during drying air under different conditions:

\[ RU = \frac{U - U_e}{U_i - U_e} \] (10)

where \( U \): water content of product (% d.b.); \( U_i \): initial water content of the product (% d.b.); \( U_e \): equilibrium water content of the product (% d.b.).

It is usual to consider the value of the diffusion coefficient constant or linearly dependent on the temperature of the drying air.

\[ D = A \exp \left( \frac{E}{RT_a} \right) \] (11)

where \( A \): constant (m^2/s); \( E \): activation energy (kJ/kmol); \( R \): universal gas constant (8.314 kJ/kmol/K); \( T_a \): absolute temperature (K).

The coefficients of the Arrhenius expression were linearized by applying the logarithm of the form:

\[ \text{Ln}D = \text{Ln}A - \frac{E}{R T_a} \] (12)

To obtain the water content of the hygroscopic equilibrium of coffee the dynamic-gravimetric method was used. A desorption thin layer of the product was performed for different controlled conditions of temperature (23, 40 and 60 °C) and RH of the drying air 40%, 50% and 60% until the product reached the equilibrium moisture content with air condition specified. Temperature and RH were monitored by means of a psychrometer installed next to trays containing the samples. During the drying, the trays with the product were weighed periodically and the hygroscopic equilibrium was reached when the mass change of the containers to remain unchanged for three consecutive weightings. The experimental data of the equilibrium water content was adjusted mathematical models are frequently used to represent the hygroscopic agricultural products, whose expressions are shown in Table 2.

The experimental design was a completely randomized design (CRD) with three tests for each drying air velocity and drying temperatures. To adjust the mathematical models, nonlinear regression, Quasi-Newton method and the computer program Statistica 7.0® were used. To check the degree of fit of each model, the significance of the regression coefficient by \( t \)-test, adopting the 5% level of probability, the magnitude of the coefficient of determination (\( R^2 \)), the mean relative error values (\( P \)) and the average estimated error (SE) and verified the behavior of distribution of residuals were considered. The relative average error and the average...
Table 2  Semi-empirical and empirical equations for determination equilibrium moisture of the grains coffee.

| Model designation | Models |
|-------------------|--------|
| Sigma Copace      | $U_e = \exp\{a - (bT) + [c\exp(a_u)]\}$ (13) |
| Sabbah            | $U_e = a(a_u b/T)$ (14) |
| Oswin             | $U_e = (a + bT)[(1 - a_u)/a_u]^{1/c}$ (15) |
| Henderson         | $U_e = [\ln(1 - a_u)/(-a(T) + 273.16)]^{1/b}$ (16) |
| Henderson modified| $U_e = [\ln(1 - a_u)/(-a(T) + b)]^{1/c}$ (17) |
| Halsey modified   | $U_e = \left[\exp(a - bT)/\ln(a_u)\right]^{1/c}$ (18) |
| GAB               | $U_e = (abca_u)[(1 - ca_u)(1 - ca_u + bca_u)]$ (19) |
| Copace            | $U_e = \exp(a - bT + ca)$ (20) |
| Chung Pfost       | $U_e = a - b\ln[-(T + c)\ln(a_u)]$ (21) |
| BET               | $U_e = \{1/[(1 - a_u)(1/ab + ((a - 1)/ab)])\}$ (22) |

$U_e$: equilibrium water content of the product (% d.b.); $T$: temperature of the drying air (°C); $a$, $b$, $c$: model coefficients; $a_u$: activity water (decimal).

error estimated for each model were calculated according to the following expressions, respectively:

$$P = \frac{100}{n} \sum \frac{|Y - \hat{Y}|}{Y}$$  (23)

$$SE = \sqrt{\frac{\sum (Y - \hat{Y})^2}{GLR}}$$  (24)

where $Y$: experimentally observed value; $\hat{Y}$: value calculated by the model; $n$: number of experimental observations; $GLR$: degrees of freedom of the model (the number of observations minus the number of model parameters).

The sensorial analysis of the coffee was carried out at the Monte Alegre S/A farm in Alfenas, MG. Coffee samples of 300 g were prepared, for each repetition, by removing the defective beans. The samples were coded and analyzed by three qualified tasters of the Brazil Specialty Coffee Association [7]. The methodology used was the Cup of Excellence (CoE), in which each sensorial attribute (body, aroma, acidity, sweetness, balance and characteristic flavor) received a score according to its intensity in the samples, making this methodology more objective than cup test. For each sensorial attribute the samples received a score on a scale of 0-8 [7]. The tasters evaluated the coffee in three parts: dry powder, crust and infusion and noted in a space reserved for individual observations. The attributes clean beverage, sweetness, acidity, body, flavor, remnant flavor, balance and general score were then evaluated, resulting in a final score count that indicated the higher and poorer quality coffees.

The electric conductivity [8], potassium ion leaching [9], total titrable acidity [10], total and reducing sugars [10] and fatty acidity [11] of the raw beans were determined.

The statistical design used was a split-plot in time, in a $1 \times 3 \times 3$ factorial scheme with three repetitions. The treatments correspond to one type of coffee (cherries), three drying systems (yard, temperatures of 40 °C and 60 °C) and three RH (40%, 50% and 60%). The data obtained were analyzed by the Sisvar 4.0 computer program and the means were compared through the Tukey’s test.

3. Results and Discussion

It can be observed in Table 3 that the mathematical models used to describe the hygroscopicity of fermented coffee presented, for most of their coefficients, a regression significance level of 5% of probability by the test $t$, in general, the models presented values higher than the coefficient of determination, greater than 0.90, except for the BET, GAB, Henderson, Modified Henderson, Chung and Pfost models. For further analysis,
Table 3 Parameter values estimated, mean relative error (P), standard deviation of the estimate (SE), coefficient of determination (R²) and residual distribution for mathematical models of drying relative humidity (RH) average of 40%, 50% and 60% of washed coffee (Coffea arabica L.).

| Mathematical model | Estimation of parameters* | R² (%) | P (%) | SE (decimal) | Distribution of residue |
|--------------------|---------------------------|--------|-------|--------------|------------------------|
| RH = 40%           |                           |        |       |              |                        |
| Oswin              | a = 0.010870              | 99.77  | 0.22752| 0.0011       | A                      |
|                    | b = -0.000060             |        |       |              |                        |
|                    | c = 0.063846              |        |       |              |                        |
| RH = 50%           |                           |        |       |              |                        |
| Oswin              | a = 0.008205              | 99.99  | 0.05610| 0.0002       | A                      |
|                    | b = -0.000047             |        |       |              |                        |
|                    | c = 0.081418              |        |       |              |                        |
| RH = 60%           |                           |        |       |              |                        |
| Oswin              | a = 0.004683              | 99.99  | 0.00309| 0.0008       | A                      |
|                    | b = -0.000031             |        |       |              |                        |
|                    | c = 0.078487              |        |       |              |                        |

*All estimated coefficients were significant at 5% probability by t-test; A: aleatory distribution.

Other statistical parameters to support the selection of the best model were used. Table 3 shows the summary of the mathematical models evaluated, with the parameters adjusted by nonlinear regression to the experimental data of the equilibrium moisture content of the washed coffee, obtained by desorption with the coefficients adjusted determination (R²) and average errors for P and estimated errors (SE).

It is observed in Table 3 that the equations based on the models of Oswin showed satisfactory adjustments to the experimental data of the equilibrium moisture content of the washed coffee, with better results for the Oswin model, since it had coefficients of determination set high and average relative errors and estimated very low, independent of the temperature and RH of the drying air. Therefore, when comparing the values of the equilibrium moisture content of the pulped hygroscopic coffee, it was observed that the values of equilibrium water contents were higher for lower temperatures and higher RH.

Fig. 2 shows the experimental values of equilibrium water content of the fermented coffee obtained by desorption isotherms as well as estimated by the model Oswin. The constant water activity values of equilibrium water content of hygroscopic fermented coffee decreased with increasing temperature and with decreasing RH.

The isotherms calculated by the pulped coffee Oswin model can be classified as type III as verified for the seeds of radish [12] and for crambe [13]. Nevertheless, for most agricultural products, isotherms showed a typical sigmoidal shape (type II). Fig. 3 shows the mean values of the water content of the fermented coffee beans during drying in different conditions of temperature and RH. Looking at Fig. 3, it is found that the time required to reach the fermented coffee water content 0.11 (d.b. decimal) was 12 h to 168 h, demonstrating the increased speed of withdrawal of water at 60 °C and 40% RH. As expected, the drying time is affected by air temperature, noting a greater difference between temperatures of 60 °C and 23 °C. It is also observed in Fig. 3 that with increasing temperature of the drying air, there is a higher rate of removal of product water, as noted by many researchers for many agricultural products [4, 14, 15].

Table 4 shows the coefficients of the models adjusted for the coffee that was not pulped and that was analyzed during drying at different drying air temperatures and RH conditions of the air. Among the
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models that gave good results, the Midilli model was selected to represent the phenomenon of drying coffee due to its simplicity compared to other models and selected to present a number of significant coefficients. It was observed that the magnitude of the drying constant (k) for the model Midilli, which represents the effect of external conditions drying increases linearly with the rise in temperature of the drying air (Table 4). The coefficient of determination was above 98% (Table 5), indicating a satisfactory representation of the phenomenon under study [16]. According to this research, the use of the coefficient of determination as the only evaluation criterion for the selection of nonlinear models is not a good parameter...
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Table 4  Parameters obtained for Midilli model fitted to the data for drying of washed coffee processing in the different temperatures of air drying and RH of 40%, 50% and 60%.

| RH  | T (°C) | a     | k     | n     | b     |
|-----|--------|-------|-------|-------|-------|
| 40% | 23     | 0.98137 | -0.01081 | 0.63020 | -0.00661 |
|     | 40     | 0.98716 | 0.02184 | 1.52303 | -0.00597 |
|     | 60     | 1.00236 | 0.34825 | 0.97495 | 0.00042  |
| 50% | 23     | 1.00006 | 0.87784 | 0.25203 | -0.00146 |
|     | 40     | 0.99235 | 0.10855 | 1.02298 | -0.00214 |
|     | 60     | 1.00562 | 0.35236 | 1.16389 | 0.00885  |
| 60% | 23     | 0.97188 | 0.00857 | 1.06740 | 0.00296  |
|     | 40     | 0.97144 | 0.05386 | 1.31332 | 0.00018  |
|     | 60     | 1.00690 | 0.36186 | 1.10794 | 0.00751  |

Table 5  Coefficients of determination ($R^2$), mean relative errors ($P$) and mean estimated errors ($SE$) for the Midilli model approved during drying of the washed coffee under various temperature conditions and RH of 40%, 50% and 60%.

| RH  | T (°C) | $R^2$ | $P$ (%) | $SE$ (decimal) | Distribution of residue |
|-----|--------|-------|---------|---------------|------------------------|
| 40% | 23     | 98.84 | 0.03    | 0.0535        | A                      |
|     | 40     | 99.65 | 2.55    | 0.0335        | A                      |
|     | 60     | 99.82 | 4.24    | 0.0727        | A                      |
| 50% | 23     | 96.48 | 0.01    | 0.0402        | A                      |
|     | 40     | 99.82 | 1.95    | 0.0235        | A                      |
|     | 60     | 99.82 | 3.25    | 0.0406        | A                      |
| 60% | 23     | 97.08 | 4.75    | 0.1350        | A                      |
|     | 40     | 99.45 | 6.57    | 0.0420        | A                      |
|     | 60     | 99.77 | 10.95   | 0.0727        | A                      |

A: aleatory distribution.

However, analyzing the estimated average error ($SE$), which describes the value of the standard deviation of the estimate, it was found that the models Wang and Singh, Page and logarithmic approximation of diffusion, Midilli, exponential for two terms showed lower values for drying performed in different temperatures and RH of the air. It is noteworthy that the lower the value of the average estimated error ($SE$) is the better the quality of fit of the model will be relative to the observed data. The models Page, diffusion approximation and Midilli showed a low average error estimated during the modeling of drying coffee clones of *C. canephora* [3]. It appears that most of the models presented values mean relative error less than 10%, which indicates an adequate representation of the phenomenon, except for models Thompson, Newton, Henderson and two terms and Pabis [17]. The models of Thompson and diffusion of eight terms achieved, for modeling the drying of washed coffee, the biased distribution of waste, thus resulting in poor fits to the experimental data. All the models that were evaluated attended satisfactory the $P$, $SE$ and distribution residue, but the Midilli model was better that other evaluated because presented higher $R^2$ for all.

Fig. 4 shows the values of the effective diffusion coefficient for the pulped coffee beans during different drying conditions. It appears that during the drying the effective diffusion coefficient increased significantly ($p < 0.05$), with the rise of temperature and increase in RH.

Their dependence on the temperature of the drying air is described by the Arrhenius equation as shown in Fig. 5. During the drying of adzuki effective diffusion coefficients had magnitudes between $0.51 \times 10^{-10}$ m$^2$/s and $2.23 \times 10^{-10}$ m$^2$/s for the temperature range from
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Fig. 4 Values for the effective diffusion coefficient ($D$) (m²/s), due to different air temperatures (°C) drying of pulped coffee.

Fig. 5 Representation Arrhenius relationship (ln $D$) for the effective diffusivity and air temperature drying of natural coffee.

Table 6 Physical-chemical and sensorial evaluations for pulp-processed coffee as a function of different drying air temperatures in yard and mechanical dryer.

| Drying temperatures/RH air | Electric conductivity (μS/cm/g) | Potassium leaching (ppm) | Reducing sugars (%) | Total sugars (%) | Titratable acidity (NaOH 0.1 N/100 g) | Grease acidity (KOH 0.1 N/100 g) | Sensorial analysis (%) |
|---------------------------|---------------------------------|--------------------------|---------------------|-----------------|-----------------------------------|------------------------|------------------------|
| 23 °C/60%                 | 114.00 A                        | 28.67 A                  | 0.85 C              | 8.00 A          | 179.67 A                          | 0.77 A                  | 74.3 B                 |
| 40 °C/50%                 | 130.00 B                        | 40.00 B                  | 0.64 B              | 8.33 A          | 171.33 A                          | 1.22 B                  | 72.2 AB                |
| 60 °C/40%                 | 230.00 C                        | 66.33 C                  | 0.57 A              | 8.00 A          | 216.67 B                          | 2.61 C                  | 68.1 A                 |

Capital letter for comparison of the results analyzed in the column, for each type of drying did not differ to 1% of probability.

30 °C to 70 °C [18].

The almost linear fit was obtained indicates that uniform variation of diffusivity with temperature, the value being the variation of diffusivity coefficient...
obtained at 60 °C, slightly higher than the temperatures of 23 °C and 40 °C, this may be explained due to the molecular vibration of water, because the diffusion coefficient of variation effective occurs with increasing temperature, which increases the molecular vibration of water molecules and contributes to a faster diffusion [19]. It can be said, therefore, that there was a greater diffusion at a temperature of 60 °C. It is observed that the values of \( \ln D \) as a function of the inverse absolute temperature \((1/T_a)\) show similar behavior to the ranges of temperature 23 °C to 40 °C and 40 °C to 60 °C. Therefore, it can be inferred that there was no interference of external conditions or drying temperatures.

The effects of drying air temperatures on the quality of pulp processed coffees are shown in Table 6. There was a significant increase \((p < 0.01)\) in the electrical conductivity and potassium leaching, as a function of the increase in drying temperature. As observed by other authors [2, 20, 21], the amounts of leached ions have mainly occurred due to the high drying temperatures, interfering in the integrity of the cell membranes. The same is true of electrical conductivity. It can be observed that the increase in drying temperature influenced lower values of reducing sugars. However, the total sugars were not significant with increasing drying temperature.

The higher sugar contents were observed in dry coffees dried at lower air temperatures, which also corresponded to the lower values of electrical conductivity and potassium leaching, cell membrane integrity indicators [22, 23]. The authors observed the increase of titratable acidity in the natural coffee as a function of the mucilage fermentation on the acidification of the grains. Taveira et al. [24] verified the alteration of the original composition of the grains, due to the fermentative processes that occurred with the acids. However, in the present work, it can be said that the increase in total titratable acidity may have occurred mainly due to the acid concentrations from the degradations caused by the drying temperature.

The drying temperature of 60 °C compromises the structures of the cell membranes which lead the coffee to easy deterioration. Saath et al. [2] observed a reduction in total titratable acidity indexes with the elevation of drying air temperature. These values observed in Table 6 are close to those found by Marques et al. [21] and Dias et al. [22], from 220.00 mL to 225.00 mL of 0.1 N NaOH/100 g. High drying temperatures and high water reduction rates would degrade the structure of coffee and cell membranes, causing extravasation and oxidation in oils, raising fatty acid levels [25]. Abreu et al. [20] and Coradi et al. [26] studying the acidity grease due to pre-drying periods and drying temperatures, observed high levels of acidity grease with increasing drying temperature. This explanation is similar to that made for the total titratable acidity discussed earlier. A probable explanation for such an event [26, 27], would be the fact that the oils of coffee beans under heating conditions and in the presence of acids (higher amounts in natural coffee) can be hydrolyzed to glycerol and fatty acids, the latter being partially volatilized.

4. Conclusions

The different conditions of the ambient air significantly influenced the processes of the drying pulped coffee. The water content of the hygroscopic equilibrium of pulped coffee is directly proportional to the water activity and RH, decreasing with increasing temperature, for the same value of equilibrium. The Oswin model was best represented with the hygroscopicity of the pulped coffee, while the Midilli model shows the best fit to describe the drying curves of the washed coffee. The effective diffusion coefficient increases with increasing temperature of the drying air and reducing RH as described by the Arrhenius equation.

The time for drying is affected by the different temperatures of air drying. Electrical conductivity,
potassium leaching, total titratable acidity and grease acidity increase with increasing drying temperature regardless of the type of processing. Reducing sugars, total sugars and sensory analysis decrease with increasing drying temperature regardless of the type of processing. The drying at 60 °C/40% RH negatively affected the coffee quality.

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