Mass transfer phenomena for erythrosine and red gardenia dyes in cherries (Prunus avium): influence of temperature on effective diffusion coefficients

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ABSTRACT

The study focuses on the quantification of the diffusion phenomenon of the artificial and natural colourant, erythrosine, and red gardenia, respectively, at 238 ppm and different temperatures (40, 50 and 60°C) in cherries, by means of the calculation of diffusion coefficients, using a mathematical model based on Fick's second law. CIELAB colour space parameters were measured in triplicate, with a Konica Minolta CR-400 colourimeter device, D65 illuminant, in approximately 5 kilograms of calibrated, pitted, and desulfited cherries for 24 h, through immersion in water baths, prior to the candying process. The sampling of cherries was carried out at different frequencies, whilst as the phenomenon of colouration progressed, two cherries were randomly extracted from different parts of the container to dispose cross section and measure the colour space parameters, thus detecting colour changes during pigment diffusion. Additionally, effective diffusion coefficients in skin and flesh were calculated, adjusting a hollow sphere diffusion model to experimental values. A significant difference was observed in red gardenia effective diffusion coefficients ($p<0.05$), contrary to those calculated for erythrosine ($p>0.05$), this might indicate that diffusion of red gardenia between temperatures is differential. Superior coefficients were obtained according as the temperature increased, indicating that this could accelerate the diffusion of dyes. Erythrosine and red gardenia's effective diffusion coefficients in cherry skin were between 1 and up to 2 orders of magnitude lower compared to those in cherry flesh, probably due to the barrier effect provided by the cherry's epidermis.

Keywords: *natural colourant, erythrosine, space colour, mathematical modelling, osmotic dehydration.*

1. INTRODUCTION

Normally, during the food transformation process these have a proper colour in their natural state, which depending on the operations or processes it is subjected can vary enormously. Concerning food, a parameter that determines primarily judgment on the flavour and conservation state is the colour, as well as product quality and food's functional characteristics (Mohamed, 2014; da Silva *et al.*, 2019). Colours can have multiple origins, e.g., the presence of substances through chemical reactions generating new compounds responsible of the appearance of novel colours in the food, for instance, the Maillard reaction or, in the case of this work, the presence of colourants incorporated as additives (Martins *et al.*, 2016; Faustino *et al.*, 2019; Oladzadabbasabadi *et al.*, 2022). Additionally, there is an enormous interest in food market regarding organic and natural products, further consumers tend to select natural additives with health promoting functional properties (Maldonado, 2021).
Concerning the elaboration of candied cherries through impregnation processes in sweetener solutions, the use of dyes has become an essential requirement given the brief seasonality of the raw material, therefore, its processing requires the placement of cherries in a solution of sulphur dioxide, eliminating the natural red colour by transforming it into leucobases, making it necessary to restore colour completely. Furthermore, Giusti and Wrolstad (1996, 2008) as well as Sigurdson, Tang and Giusti (2017) obtained maraschino cherries with an attractive stable bright red colour using radish extract, being the latter mentioned one of the few precedents in the use of natural colours with respect to the elaboration of maraschino type candied cherries.

In regard to the elaboration of candied or candied cherries through impregnation processes in sweetener solutions (Maldonado *et al.*, 2014; Maldonado and González Pacheco, 2020), the use of dyes has become a necessity given the brief seasonality of the raw material, therefore, its processing requires the placement of cherries in a solution of sulphur dioxide, eliminating the natural red colour by transforming it into leucobases, making it necessary to restore the colour completely. Furthermore, as well as obtained maraschino cherries with an attractive, very stable bright red colour, with the addition of radish extract, this being one of the few precedents in the use of natural colours in the elaboration of maraschino type candied cherries.

On the other hand, although synthetic dyes are highly versatile in the food industry, several studies have found a correlation between their consumption and various health disorders, e.g., hyperactivity (Oplatowska-Stachowiak and Elliott, 2015), DNA defects (Khan *et al.*, 2020), dermatological allergic reactions (Panachiyil *et al.*, 2019), among others.

Fick's models (Abraão *et al.*, 2013; Rubio-Arraez *et al.*, 2015; Gusmão *et al.*, 2016; Gagliardi, 2019; González-Pérez, Ramírez-Corona and López-Malo, 2021; Sayago, 2021) as well as non Fickian (Berkowitz, Emmanuel and Scher, 2008; Simpson *et al.*, 2013; Ferreira *et al.*, 2015;El Aissaoui and El Afif, 2017; Hasan *et al.*, 2019), multicomponent diffusion (Bordin *et al.*, 2019; Jamali *et al.*, 2020), numerical simulation (Deen *et al.*, 2014), irreversible thermodynamics (Seguí, Fito and Fito, 2012), hydrodynamic flow models (Shi and Le Maguer, 2002) have been used in the modelling of mass diffusion phenomena.

In order to obtain effective diffusion coefficients several studies were carried out, e.g., studies concerning mathematical modelling of concomitant diffusion of salts in champignon mushrooms (Clemente *et al.*, 2020), infrared drying process in banana slices using Fick's law (Baptestini *et al.*, 2017), pre-treated by pulsed electric field and osmotically dehydrated tissues of apple, carrot and banana (Amami *et al.*, 2014), sodium, sodium chloride and glucose diffusion during the processing of green table olives (Maldonado, Zuritz and Assof, 2008; Maldonado *et al.*, 2011).

According to the above mentioned, the objective of this work is to quantify, by means of a mathematical model, the mass transfer phenomenon of erythrosine and red gardenia dyes in cherries subjected to different temperatures, through the calculation of effective diffusion coefficients, based on Fick's second law (Rezagah *et al.*, 2011; Simpson *et al.*, 2015; González-Pérez *et al.*, 2022).

2. MATERIALS AND METHODS

Parameters L* (Lightness), a* (redness and greenness) and b* (blueness and yellowness) of the CIELAB colour space were measured in triplicate by means of Konica Minolta CR-400 colourimeter device, D65 illuminant, in approximately 5 kilograms of sized, pitted and desulfited cherries for 24 h, through immersions in water baths, former to the candied process. A multiple impregnation method named "Slow or French Method" employed by Maldonado and González Pacheco (2020) was adopted to confit the fruit, based on submerging the food matrix in hypertonic solutions of slight initial concentration, then gradually increasing and leaving them at rest for a period of 24 h between each impregnation, until reached the desired.

The formulation used to sweeten was sucrose 50% - xylitol 50%, since their mixture in these proportions did not precipitate or form crystals, during six months of storage. On the other hand, the process commenced with a sweetener solution concentration of 25 Bx, to avert the formation of wrinkles in the matrix, previously boiled and cooled to approximately 50°C.

Thereafter, the cherries were submerged in 35 Bx syrup, under the same conditions as the previous day, 24 hours after the first impregnation at 25 Bx. Erythrosine as well as red gardenia dye were used both at 238 ppm to stain the fruits during the second impregnation, moreover, these experiments were performed under permanent agitation and a constant temperature of 40, 50 and 60°C, using a PIOWAY 78 HW-1 thermostatic magnetic heating stirrer device.

Additionally, 0.9 mL of 10% (m/V) citric acid or 0.3 mL of 10% (m/V) $NaHCO_3$ were incorporated to the sweetening solution to provide a pH between 4.2 and 4.8, aiming to generate a slight precipitation of the pigment inside the cherry's cellular tissue (Mohd-Adnan *et al.*, 2011; Eggleston *et al.*, 2013).

2.1 Sampling

As the colouring phenomenon progressed, 2 cherries were randomly sampled from various zones of the container, then, by using a knife, they were cut in half, in order to dispose their cross section and proceed with the measurement of the CIELAB colour space parameters, thus distinguishing colour progress during pigments diffusion (de Freitas *et al.*, 2018; Gutiérrez-Macías *et al.*, 2019). Measurements were performed in triplicate at each sampling time, additionally, during candying process, effective diffusion coefficients for both colourants, in skin and flesh, were calculated.

2.2 Mathematical Modelling

Firstly, an equation that governs the molecular diffusion phenomenon through a solid porous hollow sphere matrix was adjusted to the experimental values (equation 1).

$$\frac{\partial a_i}{\partial t}+\left(v_r\frac{\partial a_i}{\partial r}+v_\Theta\frac{1}{r}\frac{\partial a_i}{\partial \Theta}+v_\phi\frac{1}{r\sin\Theta}\frac{\partial a_i}{\partial \phi}\right)$$
$$=D_{ij}\left[\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial a_i}{\partial r}\right)+\frac{1}{r^2\sin\Theta}\frac{\partial}{\partial\Theta}\left(\sin\Theta\frac{\partial a_i}{\partial\Theta}\right)+\frac{1}{r^2\sin^2\Theta}\frac{\partial^2 a_i}{\partial\phi^2}\right]+R_i \qquad (1)$$

2.2.1 Calculation

The non-dimensional diffusion equation with constant effective coefficients in flesh (DF) and skin (DS), considering molecular diffusion of dyes the only transport mechanism within the fruit, without generation of substances by reaction, and further assuming negligible convective transport, can be treated as a partial differential equation presented as (Carslaw and Jaeger, 1959; Crank, 1979):

$$\frac{\partial a}{\partial\Theta}=\frac{2}{R}\frac{\partial a}{\partial R}+\frac{\partial^2 a}{\partial R^2} \qquad (3)$$

Subjected to the following initial and boundary conditions:

$$\text{I.C.: at } \Theta=0 \qquad a=a_i \qquad \text{at } A\le R\le 1 \qquad (4)$$

$$\text{B.C. 1: at } \Theta>0 \qquad a=\frac{a_s-a_s}{a_i-a_s}=0 \qquad \text{at } R=A \qquad (5)$$

$$\text{B.C. 2: at } \Theta>0 \qquad \frac{\partial a}{\partial R}=-\left[\frac{\frac{D_S}{r_S-r_0}}{\frac{D_L}{r_0}}\right](a-a_s) \qquad \text{at } R=1 \qquad (6)$$

Where:

$\Theta=\frac{\mathcal{D}_{ij}\,t}{r_0{}^2}$; $R=\frac{r}{r_0}$; $A=\frac{r_a}{r_0}$; $a=\frac{a_{(t)}-a_S}{a_i-a_S}$

Then, an expression as infinite series that represents the evolution of the dimensionless $< a_\Theta >$ parameter in relation to dimensionless time Θ, modified to that presented by Maldonado and González Pacheco (2022), is expressed:

$$< a_\Theta > = \frac{3}{(1-A^3)}\sum_1^\infty\left\{\frac{a_i}{b}\left[\int_A^1 R^2\,\phi_{m(R)}dR\right]^2-\frac{EA_0}{\lambda_n{}^2-A_1}\left[e^{(\lambda_n{}^2-A_1)\Theta}-1\right]\int_A^1 R^2\,\phi_{m(R)}dR\right\}e^{-\lambda_n{}^2\Theta} \qquad (7)$$

With the aim of calculate the effective diffusion coefficients and eigenvalues λ_n, an iterative nonlinear regression method by means least squares method was implemented using Microsoft Excel® software.

2.3 Statistical analysis

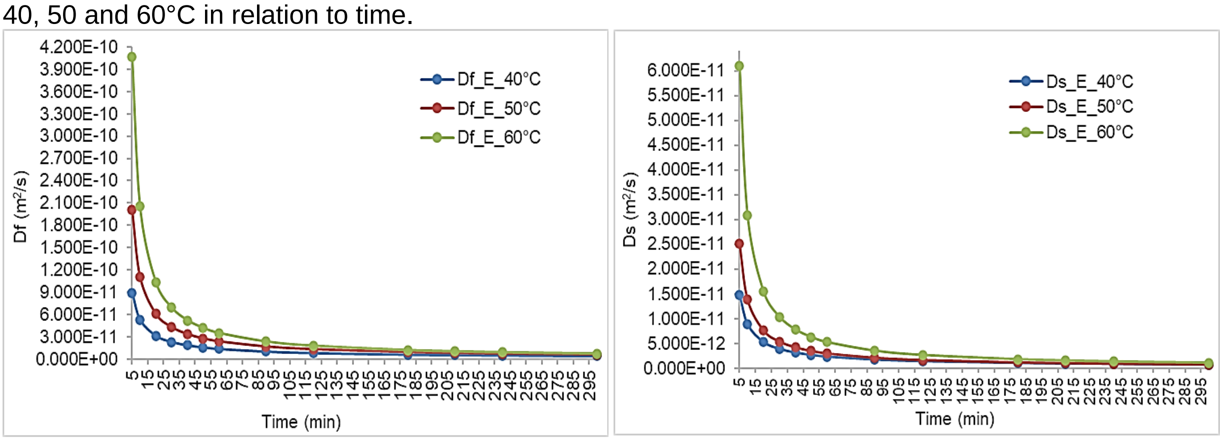
Tukey-Kramer HSD test was used in analysis of variance (ANOVA), as well as Scheffe test, with a significance level of 0.05, using the IBM® SPSS® software (V22.0, IBM Corp., NY, USA), in order to determine the statistical difference in the mean of the coefficients. The results are presented as mean ± standard deviation (SD).

3. RESULTS

Regarding Figure 1, it can be observed that both in cherry skin and flesh, the major diffusion coefficients were obtained when impregnations performed at 60°C with a difference of 1 order of magnitude (Fig. 1), then the phenomenon lessened at 50°C, whilst at 40°C the lowest coefficients were obtained, this could indicate that temperature, generates an accelerated effect of the erythrosine diffusion phenomenon through the flesh and cherries' skin.

Additionally, the diffusion coefficients in cherry's flesh (Df) were 1 and up to 2 orders of magnitude higher than those calculated in cherry skin (Ds) (Maldonado, Zuritz and Miras, 2008; Maldonado *et al.*, 2011; da Conceição Silva *et al.*, 2012; Herman *et al.*, 2018), possibly due to the barrier effect provided by the fruit's epidermis.

Fig. 1. Evolution of effective diffusion coefficients of erythrosine in flesh (Df) and cherry skin (Ds), at 40, 50 and 60°C in relation to time.



Concerning Figure 2, the effective coefficients for red gardenia dye, demonstrates the same behaviour explained above, however, these coefficients obtained were higher than those obtained for erythrosine. This difference is possibly due to its smaller size and molecular weight, compared to those for erythrosine. Moreover, it is observed that during the first 2 hours of started the process, the main diffusion phenomenon of dyes occurs to a greater extent, when the major difference concentration is presented (Della Rocca, Roche and Mascheroni, 2013).

On the other hand, the effective diffusion coefficients, both for erythrosine and red gardenia, decreased as the phenomenon progressed, approaching zero, for advanced impregnation times. This could be due to reduction of the porous solid's permeability, therefore, the solution that impregnated the wall of the canaliculi could increase its tortuosity, increasing the difficulty of transportation through them.

Fig. 2. Evolution of effective diffusion coefficients of red gardenia in flesh (Df) and cherry skin (Ds), at 40, 50 and 60°C in relation to time.

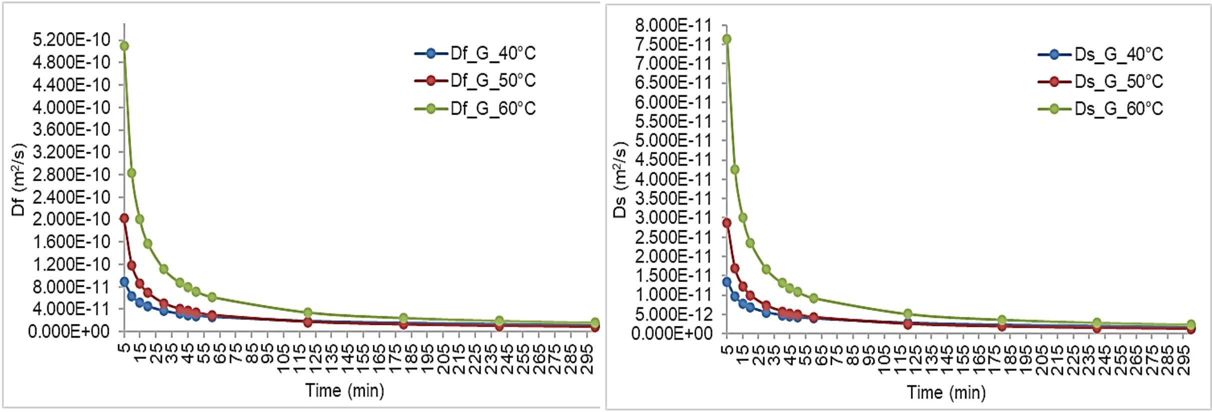


Table 1. Effective diffusion coefficients of erythrosine and red gardenia in skin (Ds) and cherry flesh (Df) for each temperature.

		Temperature (°C)		
		40	50	60
Erythrosine	Df (m^2/s) †	2.175E-11±2.442E-11[a]	4.304E-11±5.550E-11[a]	7.633E-11±1.135E-10[a]
	Ds (m^2/s) †	3.626E-12±4.070E-12[a]	5.380E-12±6.937E-12[a]	1.145E-11±1.702E-11[a]
Red Gardenia	Df (m^2/s) †	3.562E-11±2.248E-11[a]	5.501E-11±5.456E-11[ab]	1.274E-10±1.390E-10[b]
	Ds (m^2/s) †	5.343E-12±3.372E-12[a]	7.859E-12±7.795E-12[ab]	1.911E-11±2.086E-11[b]

†Results are presented as means ± SD. Mean coefficients with different letters in the same row are significantly different ($p < 0.05$).

Table 1 indicates the average values of effective diffusion coefficients of erythrosine and red gardenia in skin and cherry flesh, for each temperature. Considering flesh and skin diffusion, at 60°C, the effective diffusion coefficients were superior than those obtained at 40 and 50°C, further, significant differences ($p>0.05$) were observed between red gardenia diffusion coefficients, indicating the diffusion between temperatures could be differential (Maldonado and González Pacheco, 2022), nevertheless erythrosine diffusion coefficients in both skin and flesh were not significantly different ($p>0.05$).

4. CONCLUSIONS

By means of mathematical modelling the diffusion phenomenon of erythrosine and red gardenia at 238 ppm in cherries, subjected at 40, 50 and 60°C was quantified. Superior diffusion coefficients were obtained when impregnations performed at 60°C, reporting a difference of 1 order of magnitude greater compared to those calculated at 40 and 50°C, this could be due to the temperature accelerates the colourants diffusion phenomenon in the food matrix. Moreover, a difference of 1 and up to 2 orders of magnitude higher was obtained for coefficients in flesh in contrast with skin, in both dyes, which might be due to the barrier effect provided by the cherry's epidermis.

During the first 2 h of trials, all experiments revealed the main dyes transport when the maximum difference concentration is presented. No significant difference was obtained as regards the erythrosine diffusion coefficients ($p>0.05$), whereas concerning red gardenia coefficients in skin and flesh presented differences between the experiments ($p<0.05$), possibly due to the diffusion amongst temperatures was differential. Additionally, at advanced process times, all effective coefficients, decreased as the impregnation phenomenon progressed.

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