



Modeling Super-Diffusive Drying Behavior of Laser-Perforated Grapes Using Fractional Calculus: A Comparative Study with Alkaline Dipping

Berkay Berk¹ · Ebru Köroğlu¹ · Sevcan Ünlütürk¹

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Abstract

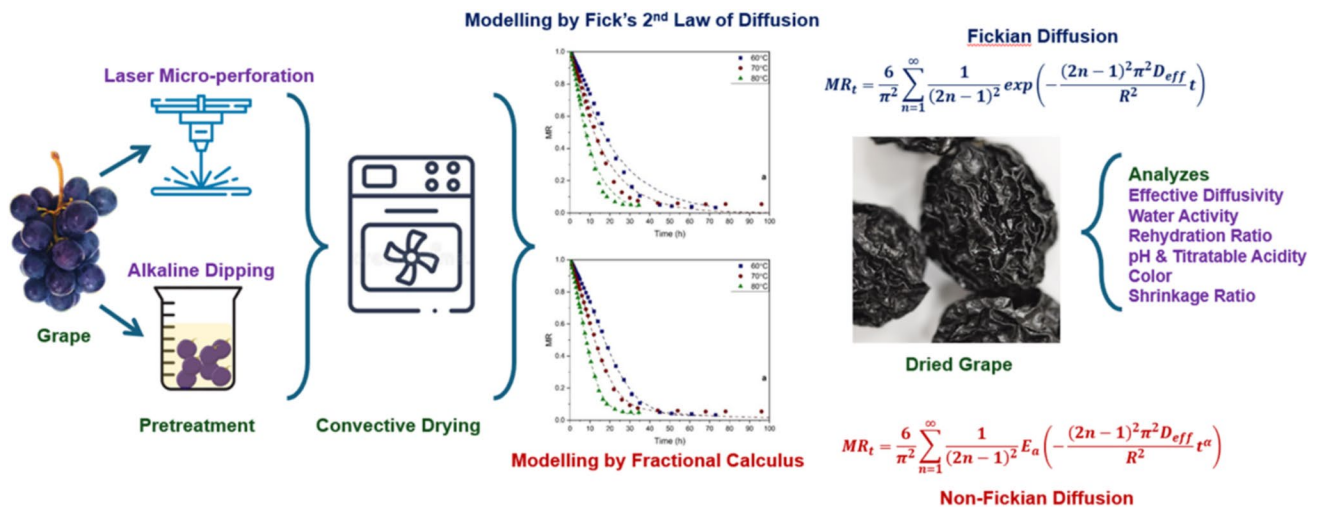
Drying large grape varieties is limited by low mass transfer rates, resulting in long drying times. This study evaluated blue laser micro-perforation as a chemical-free pretreatment alternative to alkaline dipping for drying large Kavacık grapes (*Vitis vinifera* L. cv. Alphonse Lavallée), focusing on mass transfer behavior. Drying was conducted at 60, 70, and 80 °C with an air velocity of 2.5 m/s. The effects of pretreatment on drying behavior, drying mechanism, and selected physicochemical properties were examined. Moisture diffusion was modeled using both Fick's second law and an anomalous (non-Fickian) diffusion model based on fractional calculus. The non-Fickian model provided a superior fit to the experimental data ($R^2 > 0.99$; $\chi^2 < 0.008$; RMSE < 0.01) under all drying conditions. The effective moisture diffusivity (D_{eff}) of laser-treated samples ranged between 1.42×10^{-10} and 7.79×10^{-10} m²/s and was lower than that of untreated samples (3.22×10^{-10} – 12.80×10^{-10} m²/s), indicating super-diffusive mass transport behavior ($\alpha > 1$), where α represents the anomalous diffusion exponent. Drying temperature significantly enhanced D_{eff} in both pretreatments, such that higher drying temperatures resulted in increased effective moisture diffusivity due to enhanced molecular diffusion and reduced internal resistance. Physicochemical properties, including water activity, rehydration ratio, pH, titratable acidity, color (ΔE), and shrinkage ratio, were not significantly affected by the type of pretreatment or drying conditions. Laser micro-perforation achieved drying efficiency and quality retention comparable to alkaline dipping without chemical additives, demonstrating strong potential as a sustainable pretreatment for industrial-scale fruit drying.

Highlights

- Laser micro-perforation improved drying and quality like alkaline dipping.
- The non-Fickian model fitted large grape drying, showing super-diffusion ($\alpha > 1$).
- Laser pretreatment preserved grape quality better than the chemical method.
- Laser pretreatment is a chemical-free alternative with D_{eff} similar to dipping.

Extended author information available on the last page of the article

Graphical Abstract



Keywords Laser micro-perforation · Alkaline-dipping · Grape drying · Non-Fickian diffusion · Effective moisture diffusivity

Introduction

Grapes are widely cultivated worldwide, with Türkiye playing a key role in production and export (Sabra et al., 2021; Topalović et al., 2020). Favorable climatic conditions of Türkiye support diverse grape varieties (Ataseven, 2024). Among them, Kavacık grapes, cultivated in the Karabağlar district of İzmir, possess geographical indication status and are classified as *Vitis vinifera* L. Alphonse Lavallée (Ministry of Culture and Tourism, 2023). The berries of Kavacık grapes weigh between 4.85 and 6.14 g, while the clusters range from 800 to 1000 g. They are characterized by a thick, glossy, dark purple-black skin (Gülcü, 2016). The variety contains high levels of polyphenols and bioactive compounds (Aubert & Chalot, 2018). This table grape is harvested between September and December, consumed fresh, extensively processed into Turkish delight and molasses (Köroğlu et al., 2025). However, due to its large size, it is less suitable for drying. It is essential to transform this grape, which offers health benefits due to its high bioactive substances, into a form suitable for drying so it can be consumed year-round.

Grapes, with high moisture and sugar content such as Kavacık grapes, are prone to chemical and microbiological spoilage, leading to economic losses if not properly stored or processed (Huang et al., 2019; Srivastava et al., 2021). Drying is a widely used preservation method that reduces moisture and water activity, preventing spoilage

(Bhattacharjee et al., 2024; Huang et al., 2019; Senadeera et al., 2020; Srivastava et al., 2021). Besides extending shelf life, it reduces storage, packaging, and transportation costs by reducing product weight and volume (Ozkan et al., 2022). Drying methods are grouped as natural (sun drying) and artificial (hot air, microwave, spray, infrared, foam, puff drying) (Adeyeye et al., 2022; Günaydın et al., 2022). Hot air drying (HAD), developed as an alternative to sun drying, provides faster drying, shorter processing times, ease of use, relatively low costs, and better quality and safety preservation (Senadeera et al., 2020). HAD has been applied to various products, including melon, mango, tangerine, tomato, strawberry, and grape, to study drying behavior and physical properties (Deng et al., 2019).

Grape drying is energy- and time-intensive due to the waxy skin that restricts moisture diffusion. To accelerate drying, reduce energy use, and improve quality, various pretreatments are applied (Gavahian et al., 2024; Patidar et al., 2021). These are classified as chemical (alkaline or acidic dipping, sulfur or ozone exposure, etc.) and physical (microwaves, pulsed electric fields, freezing, ultrasonic fields, etc.) (Ağçam et al., 2025; Bassey et al., 2021; Deng et al., 2019). The most common chemical method is alkaline dipping, which disrupts the waxy layer, shortens drying time, and preserves color (Carranza-Concha et al., 2012; Doymaz, 2012; Shafiq et al., 2022). Nevertheless, this method is associated with several limitations, including chemical residue concerns, wastewater generation, equipment corrosion, and

increasing environmental and regulatory constraints. As a result, physical pretreatment methods have gained increasing attention as cleaner and more sustainable alternatives. Among physical methods, laser micro-perforation creates microscopic skin channels, enhancing diffusion and reducing drying time. It has been applied in packaging, quality improvement, and process optimization (Qu et al., 2022). Laser micro-perforation also enhances infusion and diffusion (Fujimaru et al., 2012; Silva-Vera et al., 2020), reduces marination times (Figueroa et al., 2020), and increases diffusion coefficients during drying (Araya et al., 2022). Previous drying studies used CO₂ lasers, effective due to strong water absorption but limited by cost and accessibility (Chavan et al., 2023; Chen et al., 2021; Teng et al., 2021). In a previous study, laser micro-perforation applied to apple slices dried using refractive window (RW) drying provided quality attributes comparable to freeze-drying while reducing drying time by 2–25% (Núñez et al., 2023). Also, in another study, raspberries pretreated with laser perforation and dried in a pulse-spouted microwave freeze dryer showed reduced drying time, improved product quality (structure, color, and anthocyanin retention), and no adverse effects on volatile compounds. Moreover, the method was emphasized as suitable for mechanization and automation, enabling industrial-scale application (Qu et al., 2022).

Blue lasers are high-energy systems emitting light in the 445–450 nm range with high-power output, providing precise and localized ablation suitable for controlled material processing (Puértolas et al., 2024). Compared to conventional CO₂ lasers, blue lasers are more affordable, widely available, and easier to implement in experimental settings while maintaining efficient energy delivery for micro-perforation. In food science, they have been used to manipulate powdered foods, enhance interlayer adhesion in 3D printing, and control localized heating in dough, demonstrating their potential to improve mass transfer and processing efficiency (Blutinger et al., 2018; Diaz et al., 2018). Despite these applications, their use as a pretreatment before drying has not yet been reported. Based on their advantages and precision, high-power blue laser micro-perforation was used in this study as an innovative method to enhance mass transfer and preserve quality attributes in fruit matrices.

Mathematical models based on Fick's second law have long been employed to characterize the drying behavior of fruits and vegetables (Onwude et al., 2016). They provide a framework to estimate moisture diffusion and predict drying times (Cavalcanti-Mata et al., 2020). These are based on isotropic diffusion and homogeneous mass transfer assumptions throughout the material (Köroğlu et al., 2025). However, during drying, complex structural changes frequently occur, including shrinkage, anisotropic deformation, and microstructural modifications (Rani & Tripathy, 2020). Such changes lead to heterogeneous water movement and

deviations from classical Fickian behavior (Ramírez et al., 2017; Simpson et al., 2013). These structural transformations can produce sub- or super-diffusion phenomena. In these cases, diffusion rates vary within different regions of the food matrix, resulting in non-uniform moisture transport. Accounting for these complexities, anomalous (non-Fickian) diffusion models have been developed. They are frequently formulated through fractional calculus and provide greater flexibility and improved accuracy in describing the sigmoidal drying kinetics commonly observed in biological materials (Matias et al., 2023; Ramírez et al., 2017; Simpson et al., 2013). These models have been successfully applied to a wide range of widely consumed agricultural products, including apples, bananas, tomatoes, grapes, and wheat. Their broad applicability has been demonstrated in both experimental analysis and the design of efficient drying systems (Aranha et al., 2024; Erenturk et al., 2019; Granella et al., 2022; Köroğlu et al., 2025; Matias et al., 2023; Ramírez et al., 2017).

This study aims to (1) evaluate the effects of blue laser micro-perforation and alkaline dipping pretreatments on the drying behavior of Kavacık grapes (*Vitis vinifera* L. cv. Alphonse Lavallée), (2) analyze the diffusion mechanism during drying using Fick's second law and fractional calculus-based anomalous diffusion models, and (3) examine the impact of pretreatments on selected physicochemical properties, such as water activity, rehydration ratio, pH, total acidity, color, and shrinkage ratio. This study advances the understanding of blue laser micro-perforation as a pretreatment for grape drying by examining the associated drying behavior using a non-Fickian approach. Accordingly, it proposes that blue laser micro-perforation can serve as an effective, non-chemical pretreatment, improving mass transfer while maintaining the quality of large grapes.

Materials and Methods

Materials

Kavacık grapes (*Vitis vinifera* L. cv. Alphonse Lavallée) were obtained from local producers in Kavacık, Karabağlar, İzmir, Türkiye. The grapes were harvested at optimal maturity, as determined by the maturity index (°Brix/TA), which ranged from 20 to 25 (Kaya & Unluturk, 2019). Following harvest, the grapes were immersed in a 5% vinegar solution for 15 min to prevent fungal spoilage, subsequently rinsed, air-dried at room temperature, and stored in 0.5 kg polyethylene packages under refrigerated conditions (4 °C) for up to 15 days. Average berry size was 25.23 ± 3.80 mm in diameter, 25.99 ± 4.92 mm in length, and 8.90 ± 0.48 g in weight, with an initial moisture content of 4.04 ± 0.02 kg water/kg dry solid. Grape skin thickness was measured

following the method developed by Köroğlu et al. (2025) using a phase-contrast microscope (Novex, The Netherlands) equipped with ruler calibration. The captured images were analyzed using a software package Image-Pro Plus 4.5.1 (Media Cybernetics Inc., Silver Spring, MD, USA). The average skin thickness of the grapes was determined to be 0.866 ± 0.012 mm. The groups of samples used in this study are the control group (DG) that is alkaline-dipped grapes in chemical solution and laser micro-perforated grapes (LG16, LG24, and LG32). Before drying, grapes were perforated using a 450-nm blue laser (CNC2418, China) with the same penetration length but different number of holes (16 holes for LG16, 24 holes for LG24, and 32 holes for LG32) with a duration of 7 s. In preliminary studies, the drying time was found to be too long as the control group when the grape is directly dried without any pretreatment. Therefore, alkaline dipping was selected since it is widely used in industry and effectively reduces the drying time without sacrificing the quality attributes (de Souza Matias et al., 2019; Kaveh et al., 2018; Sahin & Sumnu, 2006). For this treatment (DG), grapes were immersed in a solution of 5% (v/v) K_2CO_3 and 1% (v/v) olive oil at 25 °C for 2 min (Doymaz & Altiner, 2012).

Drying Process

Approximately 25 g of grapes from each group—laser micro-perforated (LG16, LG24, LG32) and alkaline-dipped control (DG)—was dried at 60, 70, and 80 °C under convective hot air drying conditions employing a custom-built convective dryer (Kumtel, Türkiye) (Azzouz et al., 2002; Hermassi et al., 2017; Noshad & Ghasemi, 2020). Air velocity of the convective dryer was determined using a thermo-anemometer (Model 451126, EXTECH, Waltham, MA), and average value was found to be 2.5 m/s. Drying temperatures were selected to balance quality retention and microbial safety, with high temperatures potentially causing quality loss and low temperatures prolonging drying and increasing microbial risk (Alp & Bulantekin, 2021). Before drying, the total dry matter of the grapes was determined by oven drying at 105 °C to establish the endpoint criterion for the drying process. Grape weights were measured at regular intervals using a digital scale (ABJ 220-4NM, Kern, Germany) until reaching 0.24 kg water/kg dry solid (19.5% wet basis), and drying curves were plotted for each temperature. According to the Turkish Food Codex (2017), the maximum permissible moisture content for industrially dried grapes is limited to 20%. This value was taken as a reference in this study.

Modeling of Drying Curves

The dimensionless moisture ratio (MR_t) was calculated from the moisture content using Eq. 1. (Sahin & Sumnu, 2006)

$$MR_t = \frac{M_t - M_e}{M_0 - M_e} \quad (1)$$

where MR is the dimensionless moisture ratio, M_t (kg water/kg dry solid) is the dry basis moisture content at any time, and M_0 and M_e are the dry basis initial and equilibrium moisture content (kg water/kg dry solid), respectively. The equilibrium moisture content was estimated from the data when the weight changes stabilized, meaning that the difference between three consecutive weighing values was less than 0.01 g.

Fick's Second Law of Diffusion

The convection drying provides sweeping out the moist air on the surface of the food rapidly. This means that the moisture removal from the out-of-boundary layer occurs through forced convection. The mass transfer inside the grapes is assumed to be one-directional diffusion, disregarding the outer layer of the grapes' anisotropy. The anisotropy in grape tissue, caused by structural variations, leads to non-uniform shrinkage during drying (Mahiuddin et al., 2018). The Fickian diffusion model for spherical geometry assumes a perfect sphere with constant radius and uniform initial moisture content, where moisture transport is diffusion-controlled and governed by Fick's second law. The effective diffusivity is constant, drying is isothermal, external resistance and shrinkage are negligible, surface moisture instantaneously reaches equilibrium, and zero flux is assumed at the center. Equation 2 represents the analytical solution of the one-dimensional diffusion equation for a sphere with initially uniform moisture content under first-type (Dirichlet) boundary conditions (Sahin & Sumnu, 2006).

$$MR_t = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{(2n-1)^2} \exp\left(-\frac{(2n-1)^2 \pi^2 D_{eff}}{R^2} t\right) \quad (2)$$

For longer drying times (Fourier number > 0.6), Eq. 2 can be simplified by retaining only the first term of the series. Therefore, Eq. 2 is reduced to Eq. 3 (Şahin et al., 2016).

$$MR_t = \frac{6}{\pi^2} \exp\left(-\left(\frac{\pi}{R}\right)^2 D_{eff} t\right) \quad (3)$$

where D_{eff} represents the effective moisture diffusion coefficient (m^2/s), t is the drying time (s), and R is the radius of the fresh grape (m). This equation has been used in numerous studies and has been further simplified to Eq. 4 (Kaveh et al., 2018).

$$MR_t = A \exp(-k_F t) \quad (4)$$

where A is the equation coefficient, and k_F is denoted as following:

$$k_F = \left(\frac{\pi}{R}\right)^2 D_{eff} \quad (5) \quad k_{FC} = \left(\frac{\pi}{R}\right)^2 D_{eff} \quad (10)$$

The model in Eq. 3 was employed to estimate D_{eff} based on the Fickian diffusion model.

Anomalous Diffusion Model Based on Fractional Calculus Approach

Classical diffusion theory fails to capture molecular transport in grapes due to their heterogeneous water and sugar distribution. In such systems, the diffusion coefficient varies, and water exhibits anomalous, non-Brownian motion. Fractional calculus-based models, which account for non-Fickian behavior, more accurately describe this complexity and have been widely applied to model anomalous diffusion in food drying (Granella et al., 2022; Köroğlu et al., 2025; Matias et al., 2023; Ramírez et al., 2017). Equation 6 based on the fractional calculus approach is proposed to be used to model non-Fickian behavior (Asemu et al., 2020).

$$\frac{\partial^\alpha M}{\partial t^\alpha} = D_{eff} \left(\frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial M}{\partial r} \right) \right) \quad (6)$$

where M is the dry basis moisture content (kg water/kg dry solid), and D_{eff} is the effective moisture diffusivity (m^2/s). The parameter α defines the order of random motion in anomalous diffusion, ranging between 0 and 1 (Lemus-Mondaca et al., 2015; Matias et al., 2023; Ramírez et al., 2017). $\alpha < 1$ indicates sub-diffusion (slower rate than Fickian model), $\alpha > 1$ denotes super-diffusion (faster rate than Fickian model), and $\alpha = 1$ corresponds to Fickian diffusion (Ramírez et al., 2017).

The analytical solution for Eq. 6 is presented in Eq. 7.

$$MR_t = \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{(2n-1)^2} E_A \left(-\frac{(2n-1)^2 \pi^2 D_{eff}}{R^2} t^\alpha \right) \quad (7)$$

where E_A is the Mittag-Leffler function (de Souza Matias et al., 2019). For long drying times, the first term dominates the entire series, and the equation becomes (Ramírez et al., 2017):

$$MR_t = \frac{6}{\pi^2} E_A \left(-\left(\frac{\pi}{R}\right)^2 D_{eff} t^\alpha \right) \quad (8)$$

As the fractional parameter α approaches 1, the Mittag-Leffler function converges to an exponential function, resulting in the Page equation (Eq. 9), which is widely used for mathematical modeling of food drying processes (Doymaz, 2006).

$$MR_t = A \exp(-k_{FC} t^\alpha) \quad (9)$$

where the k_{FC} is explained as:

Temperature Dependency of Deff

A fractional calculus model was employed to describe the temperature dependence of the effective diffusion coefficient (D_{eff}), offering a more accurate representation of variability in food matrices than the Fickian diffusion model. The model-derived effective diffusivity was used to assess its temperature dependence via the Arrhenius equation (Eq. 11) (Ramírez et al., 2017).

$$D_{eff} = D_0 \exp \left(-\frac{E_a}{RT} \right) \quad (11)$$

where D_0 represents pre-exponential factor (m^2/s), E_a represents activation energy (kJ/mol), R represents gas constant (8.314 J/mol K), and T is temperature (K).

Water Activity

The water activity (a_w) of dried grapes was measured at 25 °C using a digital water activity meter (Rotronic HygroLab 3, UK).

Rehydration Ratio

The rehydration ratio of grapes was calculated by following the method of Köroğlu et al. (2025). Approximately 2–3 g of dried grapes was soaked in distilled water (1:10—dried grape to water ratio, w/v) at 4 °C for 24 h. After removing surface water, the samples were weighed, and the rehydration rate (RR) was calculated using Eq. 12 (Okpala & Ekechi, 2014).

$$RR = \frac{W_F}{W_I} \quad (12)$$

where W_F is final weight (g) after rehydration and W_I is initial weight (g) before rehydration.

pH and Titratable Acidity

The pH of dried grapes was measured with modifications to Unal (2023). Dried grapes were rehydrated, crushed, filtered, and measured using a pH meter (HANNA Instruments, USA). Titratable acidity was determined by titrating 20-mL filtrate with 0.1 N NaOH to pH 8.1 and expressed as tartaric acid equivalent (Mazlum & Nizamlioglu, 2021).

$$TA \left(\frac{\text{gtartaric acid}}{L} \right) = \frac{V \times 0.007505}{m} \times 100 \quad (13)$$

where V is the amount of NaOH consumed (mL), m is the amount of samples titrated (g), and 0.007505 is the conversion factor of molar mass of tartaric acid to gravimetric mass.

Color

Color changes of grapes after drying were evaluated with a colorimeter (3NH, China). Five grapes were randomly selected from each group (dried and fresh grapes). CIE L^* , a^* , b^* values were recorded from 5 points, and total color difference (ΔE) was calculated using Eq. 14 (Bai et al., 2013).

$$\Delta E = \sqrt{(L^* - L_0^*)^2 + (a^* - a_0^*)^2 + (b^* - b_0^*)^2} \quad (14)$$

where L^* , a^* , and b^* are color values of dried grapes and L_0^* , a_0^* , and b_0^* are color values of fresh grapes.

Shrinkage Ratio

Shrinkage ratio (SR) was determined using free bulk density and moisture content of fresh and dried grapes (Baysal et al. 2003; Koroğlu et al., 2025). Approximately 250 g of fresh grapes and 20 g of dried grapes were placed in 500-mL and 50-mL beakers, respectively. Their volumes were recorded, and bulk density was calculated in g/mL. Moisture content was determined as described in the “Materials” section, and the shrinkage ratio (SR) was calculated using Eq. 15.

$$SR = \frac{\rho_{b,UG}}{\rho_{b,DG}} \left(\frac{X_{DG} + 1}{X_{UG} + 1} \right) \quad (15)$$

where ρ_{UG} and ρ_{DG} are bulk density (g/mL) of fresh and dried grapes, respectively. X_{UG} and X_{DG} are wet basis moisture content (g water/g sample) of fresh and dried grapes, respectively.

Model Fitting and Statistical Analysis

Drying model fitting was performed using MATLAB (MathWorks, 2023b), incorporating the Mittag-Leffler equation with $\beta \neq 1$. Model fit was assessed using R^2 , RMSE, and chi-square (χ^2) to identify the best model.

All measurements were performed in triplicate. Statistical differences between treatments were evaluated using ANOVA followed by Tukey's post hoc test ($P \leq 0.05$) in Minitab (ver. 18.1, Minitab Inc., UK). Tables report the mean $\pm$ standard deviation of the triplicate data.

Result and Discussion

Mathematical Modeling

Model Based on Fick's Second Law of Diffusion

Figure 1 illustrates the curve-fitting results obtained using the Fickian diffusion model, with the corresponding goodness-of-fit statistics presented in Table 1. Across all samples, the Fickian diffusion model consistently yielded lower coefficients of determination (R^2) and higher root mean square error (RMSE) and chi-square (χ^2) values compared to the non-Fickian diffusion model, indicating a comparatively inferior fit. In a previous study on apple slices, Fick's second law often overestimated drying time and failed to accurately capture the tail of the diffusion profile, whereas the non-Fickian model provided a better fit. These results indicate that non-Fickian models are more suitable for describing complex mass transfer in biological tissues (Simpson et al., 2015).

The effective diffusion coefficient (D_{eff}) values based on the Fickian diffusion model are shown in Table 2. D_{eff} values of grapes subjected to dipping (DG) and laser micro-perforation (LG) were significantly influenced by drying temperature ($P < 0.05$). Increasing temperature led to higher D_{eff} for both pretreatments, likely due to enhanced molecular mobility and increased water surface tension, promoting moisture diffusivity (Noshad & Ghasemi, 2020). At 80 °C, D_{eff} values of DG and LG samples were similar ($P > 0.05$), while significant differences existed at 60 °C and 70 °C ($P < 0.05$). Forced air movement in convective drying increases the vapor pressure gradient and replaces the saturated air boundary layer, facilitating faster moisture removal. Overall, enhanced heat and mass transfer, reduced external resistance, and increased internal diffusion occur in convective drying. In a previous study, apples were subjected to laser micro-perforation with 1, 3, 10, and 14 holes and subsequently dried using convective hot air drying at 70 °C. The effective moisture diffusivity (D_{eff}) values were reported to range between 6.94×10^{-10} and 8.57×10^{-10} m²/s, and no correlation was observed between the number of laser perforations and D_{eff} (Araya et al., 2022). The D_{eff} values obtained in the present study (Table 2) are comparable to those reported previously, with the differences attributable to the use of a different fruit. Similarly, no correlation was found between the number of laser micro-perforations and D_{eff} in this study. It has been hypothesized that hydrothermal stresses induced during convective drying may have partially offset the initial structural advantages provided by laser micro-perforation. Rapid moisture removal can promote local structural collapse and surface shrinkage, potentially

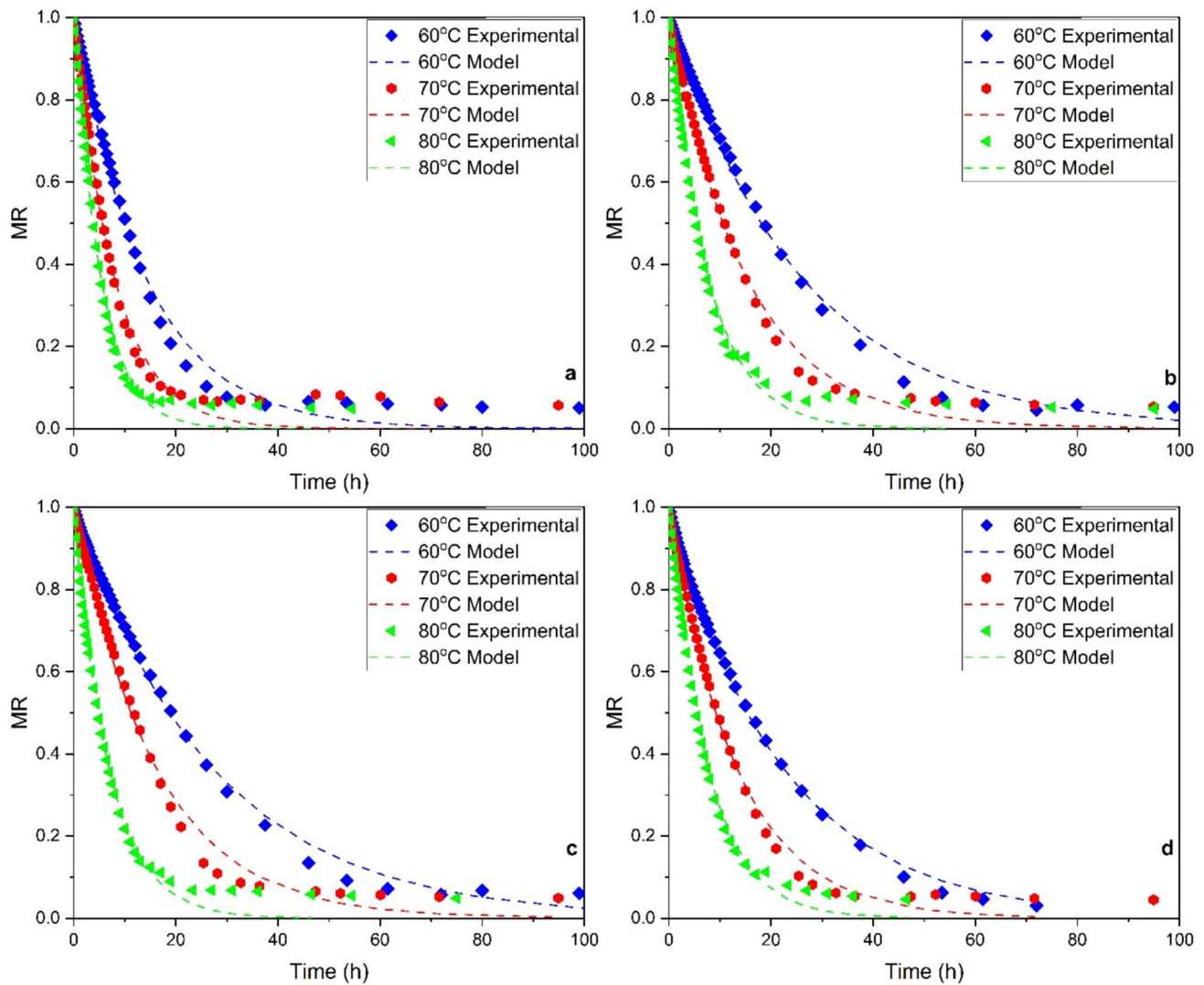


Fig. 1 Experimental and predicted drying curves of grapes according to Fickian diffusion model at 60, 70, and 80 °C for a DG, b LG16, c LG24, and d LG32 (colored markers represent experimental data, while dotted curves indicate model fitting). DG dipped grapes, LG16

laser micro-perforated grapes with 16 holes, LG24 laser micro-perforated grapes with 24 holes, LG32 laser micro-perforated grapes with 32 holes

reducing the connectivity of microchannels created by the laser treatment. Also, in another previous study, the application of various pretreatments to a black grape cultivar produced effective moisture diffusivity values in the range of $8.64\text{--}10.5 \times 10^{-10} \text{ m}^2/\text{s}$. These values fall within a similar order of magnitude to those obtained in the present study, indicating comparable moisture transport behavior under different pretreatment conditions (Doymaz, 2006).

Anomalous Diffusion Model Based on Fractional Calculus Approach

Figure 2 presents curve-fitting results using the fractional calculus-based non-Fickian diffusion model. Drying data

were better described by this model than the Fickian model, with R^2 (0.9979–0.9998), RMSE (0.0041–0.0137), and χ^2 (0.0006–0.0079) values (Table 1). High R^2 and low RMSE and χ^2 indicate reliable representation of grape drying behavior. Graphical analysis and goodness-of-fit statistics show that dipped and laser micro-perforated samples closely conform to the non-Fickian model, suggesting these pretreatments enhance model fit.

The effective diffusion coefficient (D_{eff}) values of laser micro-perforated samples, calculated using the non-Fickian diffusion model, are presented in Table 2. No statistically significant difference in D_{eff} was observed between grapes treated with dipping (DG) and laser micro-perforation (LG) at 80 °C ($P > 0.05$), whereas significant differences were

Table 1 Goodness of fit evaluation of the Fickian and non-Fickian diffusion model parameters

Parameters	DG	LG16	LG24	LG32
Fickian diffusion				
60 °C R^2	0.9931	0.9961	0.9975	0.9989
RMSE	0.0277	0.0188	0.0161	0.0090
χ^2	0.0268	0.0138	0.0114	0.0030
70 °C R^2	0.9951	0.9946	0.9942	0.9965
RMSE	0.0225	0.0242	0.0253	0.0178
χ^2	0.0172	0.0235	0.0269	0.0118
80 °C R^2	0.9939	0.9959	0.9973	0.9963
RMSE	0.0247	0.0200	0.0156	0.0181
χ^2	0.0199	0.0132	0.0078	0.0118
Non-Fickian diffusion				
60 °C R^2	0.9992	0.9992	0.9992	0.9998
RMSE	0.0078	0.0083	0.0085	0.0041
χ^2	0.0021	0.0027	0.0028	0.0006
70 °C R^2	0.9991	0.9979	0.9986	0.9993
RMSE	0.0098	0.0137	0.0127	0.0078
χ^2	0.0033	0.0079	0.0067	0.0022
80 °C R^2	0.9984	0.9990	0.9995	0.9990
RMSE	0.0128	0.0093	0.0063	0.0097
χ^2	0.0054	0.0023	0.0011	0.0028

DG dipped grapes, LG16 laser micro-perforated grapes with 16 holes, LG24 laser micro-perforated grapes with 24 holes, LG32 laser micro-perforated grapes with 32 holes

found at 60 °C and 70 °C ($P < 0.05$). This can be explained by the temperature-dependent dominance of transport mechanisms. At moderate temperatures (60–70 °C), internal diffusion resistance remains relatively high, and structural modifications induced by laser micro-perforation can meaningfully influence moisture transport, resulting in distinguishable diffusivity values. However, at 80 °C, the strong temperature dependence of diffusion enhances molecular mobility and vapor transport to such an extent that the thermal driving force becomes dominant over structural differences. In addition, rapid moisture removal at this temperature may promote tissue shrinkage, cell collapse, and possible surface sugar accumulation, which can homogenize mass transfer resistance across treatments. Consequently, the influence of perforation density becomes less pronounced, leading to statistically similar D_{eff} values at 80 °C. Additionally, the D_{eff} values of DG and LG16 samples were significantly influenced by drying temperature ($P < 0.05$). In a previous study on black grapes dried at 60 °C, the effects of potash dipping and ultrasound pretreatments were investigated. The effective water diffusion coefficient (D_{eff}) was calculated as $0.891 \times 10^{-10} \text{ m}^2/\text{s}$ for grapes pretreated under optimal ultrasound conditions and $1.404 \times 10^{-10} \text{ m}^2/\text{s}$ for those treated with the potash solution. In the present study, higher D_{eff} values were observed. Results from convective drying indicate that, at higher temperatures (80 °C), laser micro-perforation pretreatment has the potential to serve as

Table 2 Effective diffusion coefficient values based on Fickian and non-Fickian diffusion models

$D_{eff} (\text{m}^2/\text{s} \times 10^{-10})$		DG	LG16	LG24	LG32
Model					
Fickian diffusion					
60 °C		3.22 ± 0.30 ^{Ac}	1.73 ± 0.12 ^{Bb}	1.64 ± 0.10 ^{Bb}	2.00 ± 0.40 ^{Bb}
70 °C		5.77 ± 1.18 ^{Ab}	2.93 ± 0.37 ^{Bab}	2.81 ± 0.20 ^{Bb}	3.39 ± 0.38 ^{Bab}
80 °C		8.58 ± 1.03 ^{Aa}	6.50 ± 2.78 ^{Aa}	7.01 ± 2.62 ^{Aa}	6.23 ± 2.23 ^{Aa}
Non-Fickian diffusion					
60 °C		3.41 ± 1.26 ^{Ab}	1.42 ± 0.29 ^{Bb}	1.58 ± 0.35 ^{ABa}	1.55 ± 0.55 ^{ABa}
70 °C		7.18 ± 1.73 ^{Aab}	3.55 ± 0.66 ^{Bab}	2.95 ± 0.40 ^{Ba}	3.28 ± 1.50 ^{Ba}
80 °C		12.80 ± 5.43 ^{Aa}	5.89 ± 1.81 ^{Aa}	6.74 ± 3.93 ^{Aa}	7.79 ± 5.32 ^{Aa}

*Means ± standard deviation within a row followed by different capital letters and within a corresponding section's column followed by different small letters are significantly different ($P < 0.05$). DG dipped grapes, LG16 laser micro-perforated grapes with 16 holes, LG24 laser micro-perforated grapes with 24 holes, LG32 laser micro-perforated grapes with 32 holes

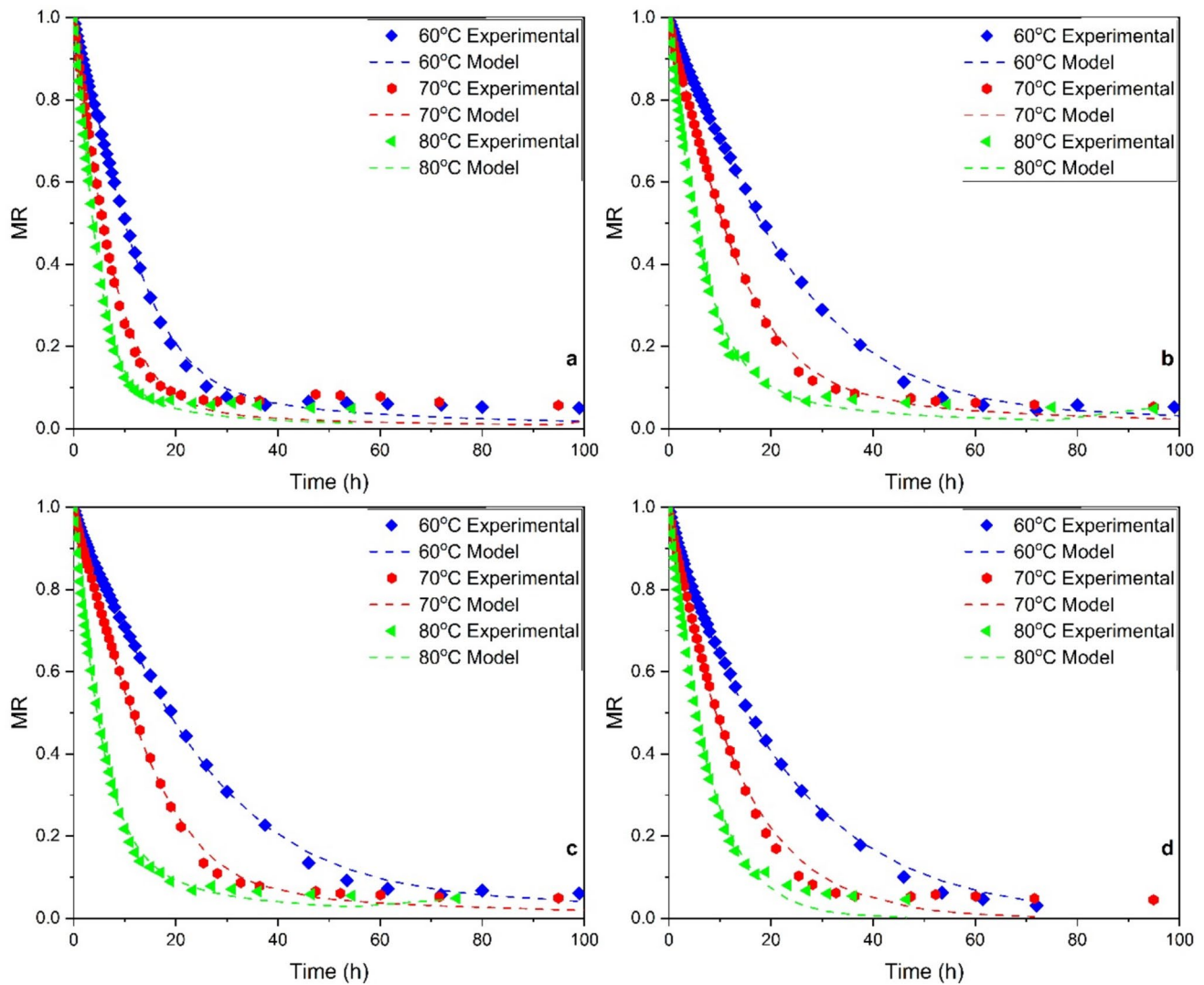


Fig. 2 Experimental and predicted drying curves of grapes according to non-Fickian diffusion model at 60, 70, and 80 °C for a DG, b LG16, c LG24, and d LG32 (colored markers represent experimental data, while dotted curves indicate model fitting). DG dipped grapes,

LG16 laser micro-perforated grapes with 16 holes, LG24 laser micro-perforated grapes with 24 holes, LG32 laser micro-perforated grapes with 32 holes

an alternative to alkaline dipping (Ağçam et al., 2025). The α value represents the fractional (non-integer) order of the model and is used to describe how moisture moves during drying. When $\alpha < 1$, the process is considered sub-diffusion, meaning the movement of moisture is slower and limited, often because the internal structure of the material prevents easy movement. When $\alpha > 1$, the process is described as super-diffusion, meaning moisture moves more quickly, which can happen when the structure becomes more open or pathways for moisture transport become easier (de Souza Matias et al., 2019; Ramírez et al., 2017; Simpson et al., 2013). All pretreated samples showed α values above 1 at all drying temperatures, indicating super-diffusion, except for LG32 at 60 °C (Table 3). This suggests that the drying

Table 3 α values defining the order of random motion in anomalous diffusion for each treatment

α	DG	LG16	LG24	LG32
60 °C	1.27 ± 0.12	1.15 ± 0.13	1.16 ± 0.07	0.96 ± 0.07
70 °C	1.21 ± 0.10	1.24 ± 0.10	1.29 ± 0.03	1.14 ± 0.09
80 °C	1.23 ± 0.07	1.18 ± 0.12	1.09 ± 0.16	1.20 ± 0.07

dipped grapes, LG16 laser micro-perforated grapes with 16 holes, LG24 laser micro-perforated grapes with 24holes, LG32 laser micro-perforated grapes with 32 holes

behavior of alkaline-dipped and laser micro-perforated grapes is better described by the non-Fickian model.

Overall, the results demonstrated that the non-Fickian diffusion model provided a markedly better description of grape drying kinetics than the classical Fickian model, as evidenced by higher R^2 and lower RMSE and χ^2 values. The effective moisture diffusivity (D_{eff}) increased with drying temperature for all samples, confirming the thermally activated nature of moisture transport and reflecting enhanced mass transfer at elevated temperatures. The D_{eff} values obtained in this study were consistent with those reported in the literature for chemically and physically pretreated grapes and other berries. At 60 and 70 °C, laser micro-perforation resulted in significantly higher D_{eff} values compared to the control, in agreement with studies on physical pretreatments such as pulsed electric fields and ultrasound, which disrupt the waxy cuticle and create preferential pathways for moisture migration under conditions where internal diffusion resistance remains limiting. In contrast, at 80 °C, no statistically significant differences in D_{eff} were observed among alkaline-dipped and laser-treated samples, indicating that the strong thermal driving force reduced the relative influence of microstructural modifications and allowed laser micro-perforation to achieve moisture transport performance comparable to alkaline dipping. Similar convergence of diffusivity values at high temperatures has been attributed to tissue shrinkage, hydrothermal stress, and surface sugar accumulation, which tend to homogenize mass transfer resistance regardless of pretreatment intensity. Moreover, the absence of a clear correlation between the number of laser perforations and D_{eff} suggests that beyond a certain perforation density, additional microchannels do not further enhance diffusion. Finally, the observation of $\alpha > 1$ confirmed super-diffusive behavior, supporting the applicability of the fractional, non-Fickian model to describe internal moisture migration throughout the entire drying process, particularly during the early and intermediate stages, and thereby extending current understanding of moisture transport in laser-treated grape tissues.

Temperature Dependency of D_{eff}

In this study, D_{eff} values from the Fickian diffusion model did not fit the Arrhenius equation, so values from the non-Fickian diffusion model were used. The corresponding activation energy (E_a) and pre-exponential factors (D_0) are summarized in Table 4. Activation energy refers to the minimum amount of energy required to initiate and sustain the movement of moisture from the interior of the material to its surface during drying. In other words, it represents the energy barrier that must be overcome for moisture molecules to diffuse through cellular structures and evaporate. A lower activation energy indicates that

Table 4 Arrhenius equation parameters of different treatments

Parameter	DG	LG16	LG24	LG32
E_a (kJ/mol)	64.70	69.69	70.77	78.82
D_0 (m ² /s)	4.90	13	19.2	350
R^2	0.99	0.98	0.99	0.99

DG dipped grapes, LG16 laser micro-perforated grapes with 16 holes, LG24 laser micro-perforated grapes with 24 holes, LG32 laser micro-perforated grapes with 32 holes

Table 5 Physicochemical properties of dried grapes

a_w	DG	LG16	LG24	LG32
60 °C	0.50 ± 0.09 ^{Aa}	0.48 ± 0.01 ^{Aa}	0.50 ± 0.02 ^{Aa}	0.57 ± 0.01 ^{Aa}
70 °C	0.39 ± 0.05 ^{Aa}	0.36 ± 0.02 ^{Ab}	0.40 ± 0.02 ^{Aa}	0.48 ± 0.08 ^{Aa}
80 °C	0.36 ± 0.05 ^{Aa}	0.47 ± 0.08 ^{Aab}	0.46 ± 0.14 ^{Aa}	0.48 ± 0.03 ^{Aa}
RR				
60 °C	1.36 ± 0.14 ^{Aa}	1.16 ± 0.06 ^{Aa}	1.18 ± 0.07 ^{Ab}	1.22 ± 0.04 ^{Aa}
70 °C	1.50 ± 0.29 ^{Aa}	1.30 ± 0.17 ^{Aa}	1.20 ± 0.07 ^{Ab}	1.24 ± 0.10 ^{Aa}
80 °C	1.48 ± 0.35 ^{Aa}	1.53 ± 0.20 ^{Aa}	1.52 ± 0.18 ^{Aa}	1.53 ± 0.21 ^{Aa}
pH				
60 °C	4.58 ± 0.26 ^{Aa}	4.37 ± 0.02 ^{Aa}	4.45 ± 0.08 ^{Aa}	4.35 ± 0.04 ^{Aa}
70 °C	4.43 ± 0.10 ^{Aa}	4.41 ± 0.17 ^{Aa}	4.34 ± 0.09 ^{Aa}	4.43 ± 0.08 ^{Aa}
80 °C	4.41 ± 0.08 ^{Aa}	4.39 ± 0.14 ^{Aa}	4.37 ± 0.07 ^{Aa}	4.38 ± 0.10 ^{Aa}
TA				
60 °C	1.18 ± 0.52 ^{Aa}	0.70 ± 0.16 ^{Aa}	0.82 ± 0.56 ^{Aa}	0.73 ± 0.05 ^{Ab}
70 °C	1.03 ± 0.13 ^{ABa}	1.07 ± 0.27 ^{ABa}	0.89 ± 0.22 ^{Ba}	1.46 ± 0.13 ^{Aa}
80 °C	1.05 ± 0.54 ^{Aa}	0.69 ± 0.22 ^{Aa}	0.81 ± 0.13 ^{Aa}	0.85 ± 0.25 ^{Ab}
ΔE				
60 °C	5.39 ± 0.88 ^{Aab}	4.43 ± 1.72 ^{Aa}	5.37 ± 1.14 ^{Aa}	5.85 ± 3.14 ^{Aa}
70 °C	6.41 ± 0.29 ^{Aa}	6.52 ± 1.00 ^{Aa}	5.53 ± 2.10 ^{Aa}	4.56 ± 1.41 ^{Aa}
80 °C	3.97 ± 1.03 ^{Ab}	4.43 ± 1.54 ^{Aa}	5.00 ± 1.30 ^{Aa}	3.67 ± 2.15 ^{Aa}
SR				
60 °C	1.28 ± 0.14 ^{Aa}	1.09 ± 0.08 ^{Aa}	0.94 ± 0.13 ^{Aa}	1.04 ± 0.17 ^{Aab}
70 °C	1.30 ± 0.19 ^{Aa}	1.19 ± 0.30 ^{Aa}	0.99 ± 0.14 ^{Aa}	0.90 ± 0.05 ^{Ab}
80 °C	1.36 ± 0.20 ^{Aa}	1.06 ± 0.21 ^{Aa}	1.24 ± 0.13 ^{Aa}	1.24 ± 0.08 ^{Aa}

*Means ± standard deviation within a row followed by different capital letters and within a corresponding section's column followed by different small letters are significantly different ($P < 0.05$). DG dipped grapes, LG16 laser micro-perforated grapes with 16 holes, LG24 laser micro-perforated grapes with 24 holes, LG32 laser micro-perforated grapes with 32 holes

the moisture can be more easily mobilized under drying conditions, while a higher value suggests stronger internal binding forces or structural resistance to water removal (Pawar et al., 2023; Sahin & Sumnu, 2006). Under convective drying, the highest E_a was observed for the LG32 sample, likely due to a sugar layer forming from excessive moisture loss and local structural collapses caused by the high number of laser perforations.

Temperature Dependency of D_{eff}

The physical analysis results of samples are presented in Table 5. Water activity is a thermodynamic variable affected by environmental conditions. Moisture sorption isotherms are widely used for estimating shelf life, as they describe the equilibrium relationship between moisture content and water activity in food systems (Choudhury et al., 2011). No significant differences were observed among water activity of samples with equivalent moisture content ($P > 0.05$). Only the LG16 samples exhibited a significant variation with drying temperature ($P < 0.05$). All samples had water activity values below 0.6, consistent with previous findings (Barbosa da Silva et al., 2022).

Dried grapes can be consumed directly or after rehydration, which typically involves soaking in water to allow moisture uptake. The rehydration behavior varies depending on the drying method, and several studies have examined the kinetics of this process (Okpala & Ekechi, 2014; L. Wang et al., 2022a, 2022b). The rehydration ratio (RR) of dried grapes did not differ significantly with respect to pretreatment or drying temperature ($P > 0.05$). The observed RR values were consistent with those reported in previous studies (J. Wang et al., 2022a, 2022b).

pH of the samples was not significantly affected by the drying process ($P > 0.05$). For titratable acidity (TA), no significant differences were observed between pretreatments at 60 °C and 80 °C ($P > 0.05$), while a significant difference occurred at 70 °C ($P < 0.05$). TA values of LG32 samples were significantly influenced by temperature ($P < 0.05$). These findings align with previous research (Unal, 2023), although Kavacık grapes showed higher TA values (0.73–1.46), likely due to varietal differences.

Color is a key quality parameter influencing consumer sensory perception. During grape drying, both enzymatic and non-enzymatic browning reactions may occur as a result of oxygen exposure and applied heat (Adiletta et al., 2016). In the assessment of ΔE values, pretreatments generally did not result in significant differences in color change ($P > 0.05$), although the DG sample was significantly affected by drying temperature ($P < 0.05$).

During drying, structural deformation, known as shrinkage, occurs as a result of skin anisotropy and internal pressure reduction. Shrinkage affects drying kinetics by shortening the diffusion path and decreasing the effective mass transfer area (Mahiuddin et al., 2018). Regarding shrinkage ratio, no significant differences were observed between pretreatments ($P > 0.05$); however, the LG32 samples were significantly influenced by drying temperature ($P < 0.05$). These variations are likely associated with differences in drying duration and sample characteristics arising from varying harvest times.

Conclusion

The findings of this study show that laser micro-perforation and alkaline dipping pretreatments have similar effects on the drying behavior and quality characteristics of Kavacık grapes. The non-Fickian diffusion model described the drying process accurately and reflected the changes in effective moisture diffusivity and activation energy at different temperatures. Higher drying temperatures increased D_{eff} , indicating faster internal moisture movement due to more facilitated mass transfer. Both pretreatments helped to speed up moisture removal, and importantly, laser micro-perforation maintained key quality properties, including pH, titratable acidity, color (ΔE), and shrinkage ratio at levels similar to those obtained with alkaline dipping. These results demonstrate that laser pretreatment can provide the same drying efficiency as chemical pretreatment while avoiding the use of alkaline solutions. Since laser micro-perforation does not introduce chemicals and can be applied in a controlled manner, it presents a more sustainable and environmentally responsible option. Overall, this study shows that laser micro-perforation is a promising pretreatment method for grape drying and may also be applied to other fruits where maintaining quality and improving drying efficiency are important.

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Data Availability The data that support the findings of this study are available from the corresponding author upon reasonable request.

Declarations

Competing interests The authors declare no competing interests.

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Authors and Affiliations

Berkay Berk¹  · Ebru Köroğlu¹  · Sevcan Ünlütürk¹ 

✉ Sevcan Ünlütürk
sevcanunluturk@iyte.edu.tr

¹ Department of Food Engineering, Faculty of Engineering, Izmir Institute of Technology, Gülbahçe Köyü, Urla, Izmir 35430, Turkey