Drying kinetics and mathematical modeling of Moroccan Peppermint tea (*Mentha Piperita*)

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Peppermint leaves (*Mentha x piperita* L.) washed with water or citric acid solution (2.5 $g.l^{-1}$) were dried under open sun. The drying kinetics on a thin layer of peppermint tea was studied at temperature of 80 °C and at drying air velocities of 200 $m^3.h^{-1}$. An indirect forced convective solar dryer in continuous was used in the experiments. The drying time is almost identical for the pilot and treated peppermint tea. It is 80 min. Ten mathematical models have been used for describing the drying curves. The Wang and Singh models showed the best fitting of experimental data of treated peppermint tea with citric acid solution and diffusion approach model is the best to describe the drying curves of peppermint tea washed with distilled water.

Keywords: *Mentha Piperita;* Solar drying; Drying kinetics; Modelling.

Nomenclature

CDC	Characteristic drying curve	X_0	Initial moisture content, (% dry matter)
$\left(-\frac{dX}{dt}\right)_0$	Initial drying rate, (kg water/(kg dry matter min))	X_f	Final moisture content, (% dry matter)
$\left(-\frac{dX}{dt}\right)_t$	Drying rate at any time of drying, (kg water/(kg dry matter min))	X_{eq}	Equilibrium moisture content, (% dry matter)
D_v	Drying air flow rate, ($m^3 h^{-1}$)	X^*	Moisture ratio
DM	Dry matter	N	Number of observations (data points)
Exp.	Experiment	n	Number of constants of models
f	Dimensionless drying rate	r	Correlation coefficient
Rh	Equilibrium relative humidity, (%)	ESM	Average systematic error
X	Moisture content at any time of drying (% dry matter)	χ^2	Reduced chi-square
t	drying time (min)		

1. Introduction

Medicinal and aromatic plants (MAPs) and their preparations are widely used by human beings all over the world (Reid et al. 2018). They are important factors in sustainable development, environmental protection and public health. About 80% of the world population still relies on MAPs for their primary health needs (Dubick 1996; Zhang 2002). Aromatic and medicinal plants are also an important source of raw materials used in the pharmaceutical, fragrance, cosmetic, flavour and perfumery industries (Bogers et al. 2006; Bruhn 1989; Miguel et al. 2004).

Peppermint (*Mentha piperita* L.) is an important medicinal plant belongs to the family Lamiaceae. It is a hybrid of *Mentha spicata* L. (spearmint) and *Mentha aquatic* (Lawrence 2006). Peppermint is a perennial herb native to Europe, naturalized in the northern USA and Canada, and cultivated in temperate areas of the world, particularly in Europe, North America and North Africa but nowadays cultivated throughout all regions of the world (Leff et al. 2004). Peppermint leaves (fresh and dried), the essential oil extracted from the aerial parts of the flowering plant, the fresh flowering plant and the whole plant are used as the medicinal parts (Briggs 1993). Best known for its flavoring and fragrance properties, peppermint have been used as an antispasmodic, aromatic, antiseptic and also in the treatment of cancers, colds, cramps, indigestion, nausea, sore throat and toothaches (Marwa et al. 2017; Neeraj et al. 2013).

Other methods of decontamination and their impact on quality parameters of peppermint, such as color and ash content, as well as fingerprint components such as phenols and essential oils have been evaluated (Machhour et al. 2011).

Drying is the most important step in the preparation of peppermint leaves for marketing. This method increases the shelf life of the final product by slowing microbial growth and preventing certain biochemical reactions that may alter the organoleptic characteristics (Díaz-Maroto et al. 2003). Several methods are used for drying mint as vacuum drying (Uribe et al. 2016), thermal (Arslan et al. 2010; Park et al. 2002) and infrared treatments (Ashtiani et al. 2017).

Peppermint plant tea has been generally sun dried under ambient conditions in Morocco. This type of drying process allows a safe final product free of any chemical preservatives or any harmful electromagnetic radiation (Kouhila et al. 2002).

To integrate experimental knowledge into industrial applications, drying kinetics should be modeled mathematically (Jayas et al. 1991). Depending on experimental data, several models are used (i) theorical models generally based on diffusion equations or simultaneous heat and mass transfer equations, (ii) semi-theoretical models (approximation theoretical equations) and (iii) empirical models (Mghazli et al. 2017).

The main objective of this work is to study the solar drying kinetics of Moroccan peppermint tea and to model the obtained data for a better understanding of the mechanisms involved.

2. Materials and Methods

2.1. Samples

Sun-dried specimens of peppermint (*Mentha piperita*) were obtained from the *Impériale des Thés et Infusions* society (Marrakesh, Morocco) and used without any sorting or cleaning treatment. Fresh samples were kept in plastic containers at 5 °C until analysis and samples for vacuum drying were processed immediately. In order to avoid leaf wilting they were packed in polyethylene bags and were stored at 4 °C until quality analyses.

2.2. Drying process

The drying experiments were conducted during the period of October 2014 in Marrakech (Morocco). A mass of 25 g of peppermint tea were sprayed with a volume of 25 ml of citric acid (2.5 $g.l^{-1}$). The prepared samples were uniformly spread in thin layer on the surface of a weighing apparatus (Fig.1). The drying experiments were conducted at 80 °C ± 0.1 °C and air flow of 200 $m^3.h^{-1}$ ± 7 $m^3.h^{-1}$ (Table 1). The mass loss of the product during the drying experiments was measured by a digital weighing apparatus (± 0.001 g). During each drying experiment (80 min), the weight of the product on the tray was measured by removing it from the drying cabinet for approximately 15-20 s. These measurements were performed every 5 min. The initial and dried moisture contents of peppermint tea were measured after drying samples for 24h in an oven at 105 ± 1 °C.

Table 1: Models of drying applied to the description of the curves of drying

Model name	Model expression	Reference
Newton	$X^* = \exp(-kt)$	Lewis (1921)
Page	$X^* = \exp(-kt^n)$	Page (1949)
Henderson et Pabis	$X^* = a \exp(-kt)$	Yaldiz et al. (2001)
Logarithmic	$X^* = a \exp(-kt) + c$	Koukouch et al. (2015)
Two-term	$X^* = a \exp(-k_0t) + b \exp(-k_1t)$	Henderson (1974)
Two-term exponential	$X^* = a \exp(-kt) + (1-a) \exp(-kat)$	Sharaf-Eldeen et al. (1980)
Wang et Singh	$X^* = 1 + at + bt^2$	Wang and Singh (1978)
Diffusion approach	$X^* = a \exp(-kt) + (1-a) \exp(-kbt)$	Yaldiz et al. (2001)
Modified Henderson and Pabis	$X^* = a \exp(-kt) + b \exp(-gt) + c \exp(-ht)$	Madamba et al. (1996)
Verma *et al.*	$X^* = a \exp(-kt) + (1-a) \exp(-gt)$	Verma et al. (1985)

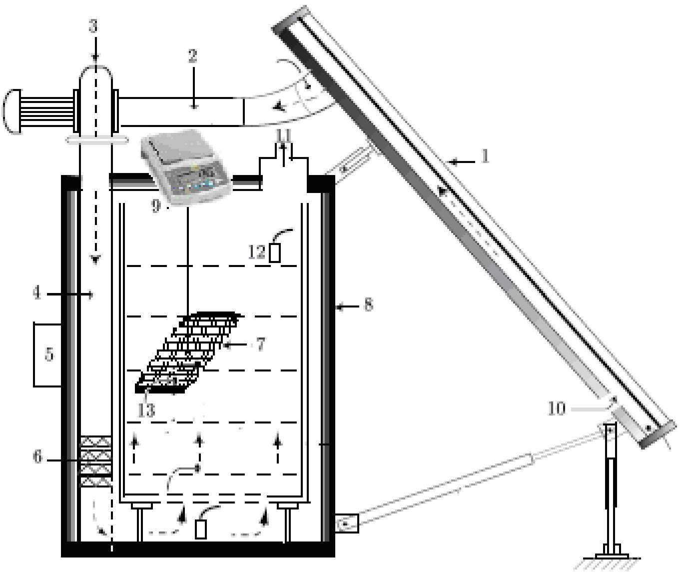

Figure 1 : Schematic representation of the solar dryer ; (1) solar collector; (2) circulation fan; (3) fan; (4) air flow direction, (5) control box; (6) auxiliary heating system; (7) tray containing the product; (8) drying cabinet; (9) balances precision; (10) control foot; (11) exit of air; (12) humidity probes; (13) thermocouples.

2.3. Mathematical Treatment

Drying experiments were performed by using a convective pilot air dryer. The Van Meel transformation (Van Meel 1958) was applied for describing kinetics and determining the characteristic drying curves. Lopez et al. (2000) and Kouhila et al. (2003) used only the initial moisture (X_0) and equilibrium (X_{eq}) moisture contents to normalize the moisture content obtained at any drying time (X). The moisture content obtained at any time of solar drying experiment is calculated as following (Moussaoui et al. 2019):

$$X(t) = \frac{X - X_{eq}}{X_0 - X_{eq}} \quad (1)$$

The dimensionless drying rate (f) was also determined for each experiment as following:

$$f = \frac{\left(-\frac{dX}{dt}\right)_t}{\left(-\frac{dX}{dt}\right)_0} \quad (2)$$

The experimental variations of moisture ratios versus drying time (X(t)=f(t)) of each treated and not treated peppermint samples were described using ten models (Table 2). The corresponding experimental characteristic drying curves given by plotting the drying rates (f) as a function of moisture ratios were also described by the mathematical correlation giving the best fit of experimental data. The equilibrium moisture content X_{eq} was determined from the desorption isotherms. The Marquardt-Levenberg non-linear optimization method (Marquardt 1963), using the computer program Curve Expert (version 3.1) was used for fitting the characteristic drying curve.

Table 2: Experimental conditions for the kinetics of drying of peppermint tea

Test	θ ± 0.1 (°C)	Rh ± 2 (%)	X_0 ± 0,001	X_f ± 0,001	T(min)
Treated peppermint tea	80.0	44	0.986	0.121	60
Pilot peppermint tea	80.0	39	0.781	0.081	50

The parameters of each model were determined by minimizing the difference between calculated and experimental data. The adequacies of the models were evaluated by using two statistical parameters: the standard error (ESM) and the correlation coefficient r. These parameters are defined as following.

$$ESM = \frac{100}{N}\sum_{i=1}^{N}\left|\frac{Xeq_{i,exp} - Xeq_{i,pre}}{Xeq_{i,exp}}\right| \quad (3)$$

$$r=\sqrt{\frac{\sum_{i=1}^{N}\left(Xeq_{i,pre}-\overline{Xeq}_{i,exp}\right)^2}{\sum_{i=1}^{N}\left(Xeq_{i,exp}-\overline{Xeq}_{i,exp}\right)^2}} \quad (4)$$

Where “pre” is the value of the moisture ratio or of the drying rate (f) calculated by using the tested model, “exp” is the experimental value of the moisture ratio or drying rate.

3. Results and discussion

3.1. Drying characteristics of peppermint

The initial moisture content of the treated peppermint leaves by water and by citric acid were respectively 0.8 % and 1 %. The evolution of the wet mass of the product during the drying process is represented in figure 2. The obtained experimental curve showed very high moisture content during the initial phase of the drying. No constant rate-drying period were observed, and the curve was typically a falling-rate period. At the end of the drying process, the higher final moisture content (X_f) was attributed to citric acid treatment better than distilled water. It should be noted that X_f represents an optimal and important characteristic value for which the product is not damaged with maintaining its nutritional and organoleptic qualities (shape, texture, aroma and essential oils.

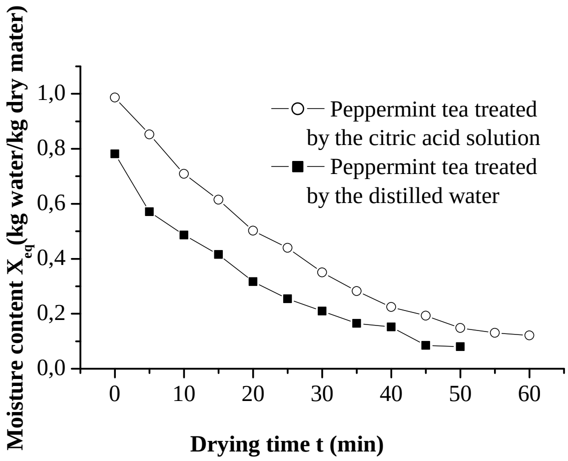

Figure 2: Peppermint tea treated by both citric acid solution and distilled water: variation of moisture content according to the time of drying.

Plots of the drying rate versus time and water content curves are shown respectively in Fig. 4 and 5. The obtained curves showed that the drying rate decreases with time. However, we note the absence of phase 0 (phase of increasing speed) and phase I (constant speed), and the unique presence of phase II (decreasing speed) in the solar convective drying curves of thin layers of the peppermint tea. In this drying phase, the speed is no longer limited by the thermal conditions, but rather by the characteristics of the product to dry as the internal migration of water, the structure of the product, etc. The same results were obtained for different plant products (Belghit et al. 2000; Koukouch et al. 2015; Lamharrar et al. 2007). The drying during the decreasing speed phase is governed by water diffusion in the solid. It is a complex mechanism involving two water liquid and vapour states, which is often characterized by the effective dissemination. This property depends on temperature, pressure and moisture content of the product.

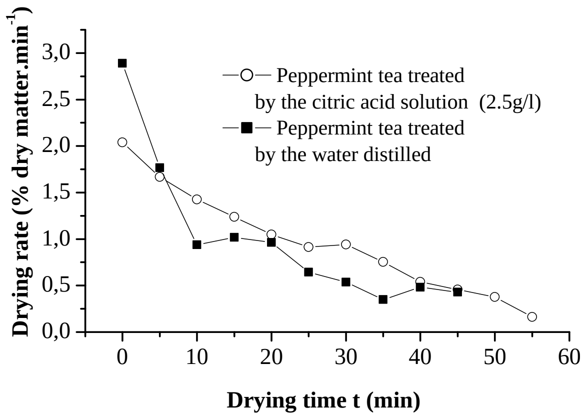

Figure 3: Evolution of drying rate of the peppermint tea treated by both citric acid solution and distilled water according to the time of drying

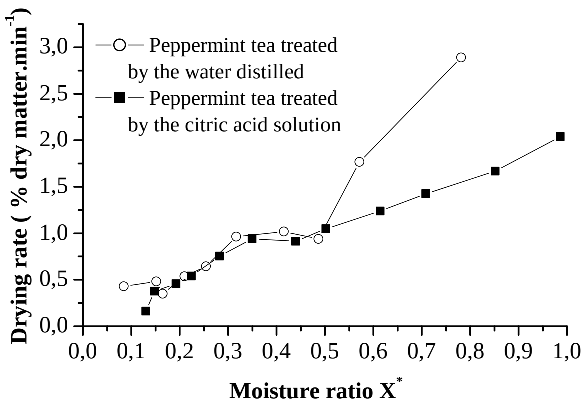

Figure 4: Evolution of drying rate of the tea of peppermint treated with both citric acid solution and distilled water according to the moisture ratio

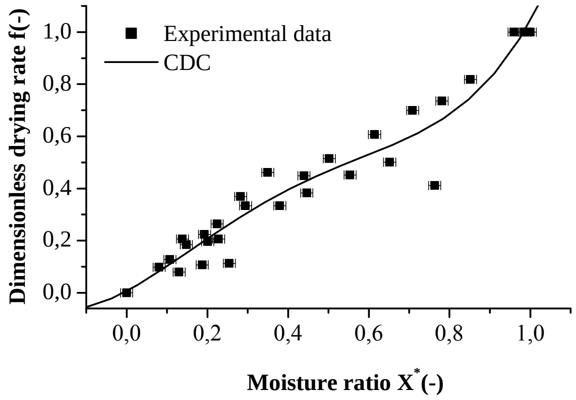

Figure 5: Characteristic drying curve (CDC) of the peppermint tea

3.2. Evaluation of the models

The experimental drying curves of the treated peppermint tea with citric acid to 0.25 % and distilled water are illustrated in Fig. 6. A good correlation between drying curves have been shown in spite of the effect of the two following parameters: changes in air conditions and the natural treatment. The method of nonlinear optimization Levenberg-Marquard has been conducted using appropriate software and treating all data points. The best fitting is obtained by choosing as a criterion for evaluating a correlation coefficient r and a higher average bias ESM minimum. The fitting of the CDC peppermint tea can determine the equation of the drying rate in the form of a polynomial of degree 4:

$$f = 0,9021X^{*} + 1.3797X^{*2} - 4.0243X^{*3} + 2.7841X^{*4} \quad (5)$$

r = 0.9410 and ESM = 0.0796

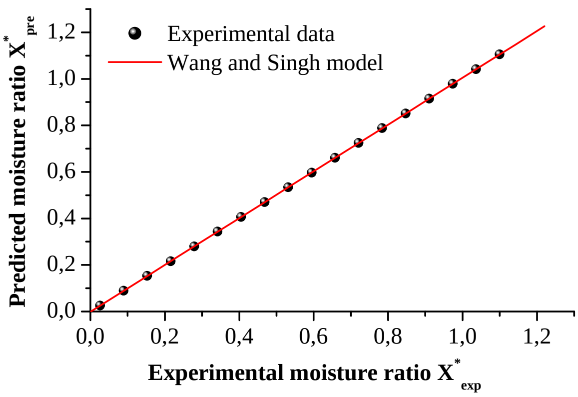

Figure 6: Experimental and predicted Wang & Singh model moisture ratio values for treated peppermint tea

The coefficients of each model of drying are determined using the method of nonlinear optimization based on the Marquard-Levenberg algorithm with appropriate software (Tables 3 and 4). The method involves the determination of the parameters of each model and the comparison of their statistical parameters. The model that provides the greatest value of *r* and the lowest values of χ^2 and ESM is considered as the best model to describe the drying kinetics of peppermint tea.

The reduced chi-square (χ^2) used for selecting the best equation to describe the thin-layer drying curves of *peppermint tea*. This parameter can be calculated as follows:

$$\chi^2 = \frac{\sum_{i=1}^{N}\left(Xeq_{i,exp} - Xeq_{i,pre}\right)^2}{N-n} \quad (6)$$

The model of Wang and Singh showed good correlation with the experimental curves with *r* of 0.9995 and χ^2 of 1.01 10^{-4} for the tea treated with citric acid. However, the model of the diffusion approximation seems more appropriate to describe the kinetics of drying the tea of control treated with water under the same conditions with r = 0.9955 and χ^2 = 1.17 10^{-3}. Indeed, Fig. 7 and 8 show a perfect correlation between experimental relative water contents and calculated relative water contents by the both models of Wang and Singh as well as the approximation diffusion model. Concerning the experimental and predicted relative water contents for the treated and control tea *versus* drying time illustrated in Fig. 9, the results of the latter (predicted water contents) match well with those of experimental values.

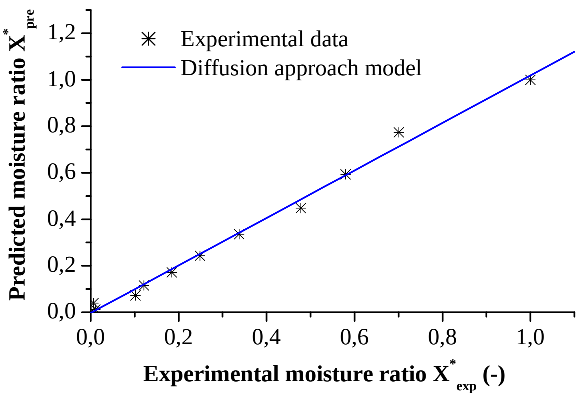

Figure 7: Experimental and predicted Diffusion approach model moisture ratio values for pilot peppermint tea

Tea treated with citric acid

(Wang and Singh model)

$$X^{*}_{pre} = 1.004 X^{*}_{\exp}$$

R = 0.9995

$$\chi^2 = 1.01 \times 10^{-4}$$

Pilot peppermint tea

(Diffusion approach model)

$$X^{*}_{pre} = 1.017 X^{*}_{\exp}$$

R = 0.9995

$$\chi^2 = 1.17 \times 10^{-3}$$

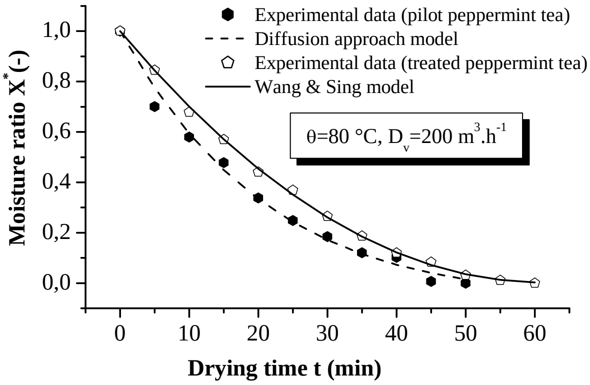

Figure 8: Water content reduced experimental and predicted according to the drying time for the treated and pilot peppermint tea

Table 3: Modelling of the water content reduced according to the time of drying of treated herb tea with citric acid to 0.25%

Model	Coefficients	r	ESM	χ2
Newton	k=0,0453	0.9882	0.0507	$2.50\ 10^{-3}$
Page	k=0.0171 ; n=1.2981	0.9971	0.0260	$6.60\ 10^{-3}$
Henderson and Pabis	a=1.054 ; k=0.047	0.9902	0.0485	$2.06\ 10^{-3}$
Logarithmic	a=1.224 ; k=0.0311 ; c=-0.2134	0.9991	0.0149	$1.49\ 10^{-4}$
Two-term	a=0.8351 ; k0=0.0475 ;b=0.2193 ;k1=0.0479	0.9902	0.0536	$2.54\ 10^{-3}$
Two-term exponential	a=1.8193 ; k=0.0637	0.9964	0.0291	$2.53\ 10^{-3}$
Wang et Singh	a=-0.0326 ; b=0.0002	0.9995	0.010	$1.01\ 10^{-4}$
Diffusion Approach	a=10.13 ; k=0.0194 ; b=0.9047	0.9992	0.0138	$1.93\ 10^{-4}$
Modified Henderson and Pabis	a=0.2466 ; k=0.048 ; b=0.4422 ; k0=0.047 ; c=0.3658 ; k1=0.0474	0.9902	0.0607	$3.26\ 10^{-3}$
Verma *et al.*	a=-2.0337 ; k=0.0147 ; k0=0.0218	0.9992	0.0139	$1.53\ 10^{-4}$

Table 4 : Modelling of the water content reduced according to the time of drying of pilot herb tea treated by water

Model	Coefficients	r	ESM	$\chi2$
Newton	k=0.0573	0.9927	0.0381	$1.44\ 10^{-3}$
Page	k=0.048 ; n=1.054	0.9931	0.039	$1.53\ 10^{-3}$
Henderson and Pabis	a=0.9956 ; k=0.0570	0.9927	0.0401	$1.60\ 10^{-3}$
Logarithmic	a=1.0883 ; k=0.0431 ; c=-0.1206	0.9966	0.0291	$9.91\ 10^{-4}$
Two-term	a=0.8077 ; k_0=0.057 ; b=0.1879 ; k_1=0.057	0.9927	0.0455	$2.06\ 10^{-3}$
Two-term exponential	a=1.4859 ; k=0.0673	0.9935	0.0379	$1.87\ 10^{-3}$
Wang and Singh	a=-0.041 ; b=0.0004	0.9902	0.0466	$2.29\ 10^{-3}$
Diffusion Approach	a=7.6638 ; k=0.033 ; b=0.9217	0.9995	0.0333	$1.17\ 10^{-3}$
Modified Henderson and Pabis	a=0.3318 ; k=0.0570 ; b=0.3318, k_0=0.0570 ; c=0.3318 ; k_1=0.0570	0.9927	0.0538	$2.87\ 10^{-3}$
Verma *et al.*	a=-1.2502 ; k=0.0258 ; k_0=0.0366	0.9955	0.0333	$1.17\ 10^{-3}$

4. Conclusion

The study of drying kinetics of Moroccan peppermint tea showed a single stage in which the drying rate decreases (phase II), with required conditions of temperature at 80° C and citric acid treatment, essential for microbiological decontamination from *Escherichia coli*. The solar convective drying enabled lowering the cost; and so, the duration of drying peppermint tea does not exceed 80 min.

The description of the experimental curves of drying was carried out using ten models of thin film drying. Wang and Singh model describes all the experimental points of the drying of the treated tea, while the model of the diffusion approximation is more appropriate to describe the kinetics of drying of the tea of control. The two products exhibit the same behavior with respect to drying kinetics.In addition, the combined biochemical treatment based on the use of citric acid at 0.25 % has no effect on the kinetics of drying of the peppermint tea.

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