

# Mass Transfer Coefficients of the Extraction Process of Essential Oil from *Myrtus Communis L.* Plants Using Different Solvents

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**Abstract:** Essential oils are highly concentrated substances having flavor and therapeutic properties. They are used in food, cosmetic and pharmaceutical products. The main objectives of this research is to extract essential oil from different parts of *Myrtus Communis L.* Plants ( leaves, flowers, and stems) by solvent extraction method using water as a green solvent compared with other solvent ( n- hexane and ethanol). The description and simulation of this process with mathematical model based on mass transfer fundamentals was investigated in this research. The process parameters affected on extraction oil yield and mass transfer coefficients, when different extraction solvents were used. Results demonstrated that different parts of *Myrtus Communis L.* plants influence on the extraction mechanism, as well as the process controlled by mass transfer diffusion.

**Key words:** mass transfer coefficient, solvent extraction, essential oil, *Myrtus Communis L.*

## I. INTRODUCTION

Essential oils have been used by many cultures around the world for centuries. Many plants yield essential oils that may be used as perfumes, food flavorings, medicines, and pharmacy [1]. Essential oils also called volatile are mixture of hydrocarbons, alcohols, aldehydes, ketons, phenols and esters [2]. In medicine, the volatile oil obtained from different parts of *Myrtus communis L.* is used for decreasing glucose level in the blood. But, the therapeutic using of this plant has a little alertness payment [3].

Myrtle plants emit a pleasant odor mainly due to volatile oil that is stocked in the secretor cells, which are placed in the plants flowers, leaves and berries [4]. Myrtle is majorly used as a pectoral when the vapors produced after leaf decoction and may be used for treatment different disease like stomach ulcers, dysentery and urinary tract infections [5].

The extraction and/or fractionation of natural complex mixtures for allowing a spontaneous and complete separation of required compounds or concentrated of benefits from the extract are the main objective of research [6].

For this purpose, different extraction processes have been studied particularly to increase the essential oil yield, reduce the processing time, and moderate the consumption of energy [7].

These methods include hydro-distillation, steam distillation, microwave extraction, supercritical fluid extraction, as well as solvent extraction. The appropriateness of extraction technique differs from plant to plant [8].

Lately, using water as an extraction solvent has awful interests as a green solvent. Specially, water is cheap, environmentally genial, nontoxic, non-flammable, providing clean processing way and prevents pollution [9]. Besides, to the environmental feature to used water in state of organic solvents, separation of products is mostly simplified. Water is a polar solvent, its density, viscosity, as well as activity can weakest its solubility and the contacting with the plant matrix [10]. Varying extraction parameters can solve this problem. Volatile oil is not soluble at room temperature with water, which start-up separation by cooling, and simplified the process steps. Thus, increasing the extraction temperature can be increase the interaction between solvent and the plant matrix, also the mass transfer and diffusion [11].

The principle of solvent extraction is that when solvent contacts with solid material, soluble compounds in the solid materials transfer to the solvent phase. However, solvent extraction in plant results a mass transfer of soluble volatile oil to the solvent phase from solid plant phase, by a concentration differences. When the concentration of active sites in the solvent increase the rate of mass transfer decrease, until equilibrium is reached. Then no longer mass transfer will be between the plant material and the solvent [12].

In general, a good solvent should be; dissolved the demand essential oil, having a suitable boiling point, can separated easily when extraction process finished, solvents like; ethanol, benzene, hexane, acetone, ethyl-alcohols etc.,[13]

Mathematical modeling for any extraction process can be represented the principle step along operation of an active industrial process. They are

using for simulating different processes without needing many number of experimental runs to optimize the preferable operating conditions to each process [14]. Cassel et al.,(2009) was used the experimental data for extraction Rosemary essential oil to investigate a mathematical model in order to perform steam distillation extraction process[15]. Xavier et al.(2011) developed a model for a fluidized bed extractor, where two extraction periods describe the concept of broken and intact cells, the extraction curves depends on mass transfer fundamental for the solute [16]. Franco-Vega et al.(2016) demonstrate a model depends on utilizing mass transfer for predicting the extraction physical kinetics, for orange peel extracted with assisted microwave extraction process, where solute diffused from the inter cells. Parameters of the model approach two observed phases, and calculated the mass transfer coefficients of the fluid-phase and solid-phase [17]. The aim of this study is to evaluate the effect of using water as a green solvent compare with other solvent ( n- hexane and ethanol) in a new packed bed extractor on the extraction yield. Since, it should be expected that essential oil extraction process can be modeled by a mass transfer mechanism, performing the behavior of three parts of *Myrtus communis* L plant, and evaluate the mass transfer coefficient.

## II. MATERIALS AND METHODS

### 2.1 Reagents and chemicals

N-hexane and ethanol that used in this research, with analytical grade product was manufactured by BDH Company.

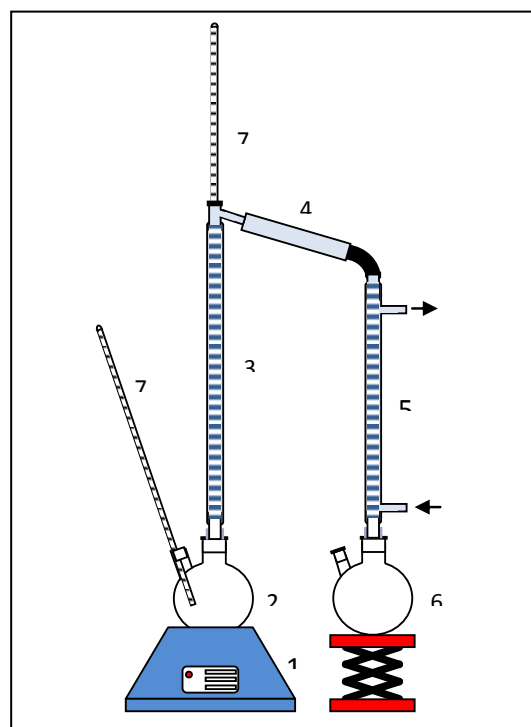
### 2.2 Plant material

The essential oil was extracted from three aerial parts of *Myrtus Communis* L. Plants (leaves, flowers, and stems); they were exposure to gradual and partial drying, after that a manual cutting were used.

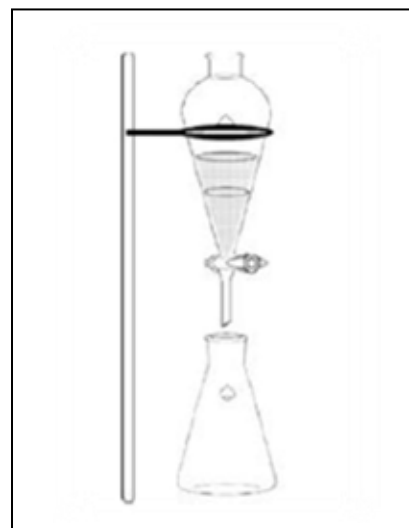
### 2.3 Packed bed extraction process

*Myrtus Communis* L. essential oils was extracted using a new extraction unit consist of a flask of boiling solvent, packed column of the required extracted part of the plant, condenser and collection flask. Figure 1, display a schematic description of the process unit. While Figure 2, show the schematic diagram of the separation of essential oil apparatus.

Fifty gram of fresh leaves of *Myrtus Communis* L. was packed in the extraction column. The required extraction solvent ( water, n- hexane and ethanol) was placed in the boiling flask. During the process, the solvent is boiled then flow the packed column and extract the required essential oil. The essential oil and the extraction solvent were collected in a receiving flask and then separated by separation funnel.



**Fig. 1: The Schematic diagram of the laboratory extraction unit:(1) mantel heater; (2) solvent flask; (3) packed column (4)connection; (5) condenser; (6) collection flask; (7) thermometer.**



**Fig. 2: The Schematic diagram of the laboratory separation of essential oil**

Concentration of essential oil in the solvent was determined by using UV-VIS.

The same procedure was used for other parts of the plant ( flowers and stems). The extracted essential oil was drying by anhydrous sodium sulfate and kept at 4°C.

The yield percent was calculated by using the following relationship:

$$\% \text{yield} = \frac{\text{amount of essential oil extracted}}{\text{total amount of grated leaves}} \times 100$$

## 2.4 Physical properties

The apparent density ( $\rho_p$ ) of the essential oil extracted from *Myrtus Communis L.* was measured using pycnometer, while the bulk density ( $\rho_s$ ) is the ratio of the mass of the particle bed for the sample in a container to the volume of the container (which include the bed volume as well as the voids among particles). The bed void fraction ( $\varepsilon$ ) can be calculated using the ( $\rho_p$ ) and ( $\rho_s$ ) values. The superficial area ( $a_o$ ) of the bed is calculated from the surface area of the whole particles in the bed. These physical properties were used in the mathematical modeling.

## III. MATHEMATICAL MODELING

In solid-liquid extraction process, the mass transfer of the solute from solid to liquid take place two steps of process. The first step is the diffusion from the inside of the solid material to its surface, and the second, is the mass transfer from the surface of the solid material to liquid [18]. The first part of the extraction curve is correlated to the extraction of the free solute from demolished cells. In this extraction step, the fluid phase is in balance with solid phase along the extractor. Second stage is guided by diffusion the solute from internal cells of the vegetable substructure and consonant to the diffusion from intact cells. In Xavier et al. model, mass transfer of extracted oil was considering constant along the process [16].

Since the solid material is small in size, so we can assumed that the solute concentration is always homogeneous and there is no concentration gradient along the solid material during extraction process. That is mean; the effective diffusivity inside the solid material can be neglected. Therefore the overall mass transfer is controlled by the mass transfer between the two phases [17].

The general model is based on the mass balances per unite volume of extraction bed for the transfer of the solute from the solid phase to the fluid phase.

$$\rho_f \varepsilon \frac{\partial y}{\partial t} + \rho_f u \frac{\partial y}{\partial t} = N(x, y) \quad 1$$

$$-\rho_s(1 - \varepsilon) \frac{\partial x}{\partial y} = N(x, y) \quad 2$$

Associated mass fluxes are:

$$N(x, y) = k_f a_o \rho_f (y^* - y) \quad \text{for } x > x_s, \text{ first period} \quad 3$$

$$N(x, y) = k_s a_o \rho_s x \quad \text{for } x \leq x_s, \text{ second period} \quad 4$$

The solution of the model can be expressed in maximum value of extracted essential oil terms ( $E_\infty$ ), described as:

$$\frac{E_t}{E_\infty} = \frac{\int_0^t m y dt}{E_\infty} \begin{cases} \frac{k_e t}{E_\infty} & \text{for } x > x_s \\ (1 - e^{k_d t}) & \text{for } x \leq x_s \end{cases} \quad 5$$

Considering the two extraction periods, then  $k_e$  is the mass transfer coefficient for the first period, and  $k_d$  is the mass transfer coefficient for the second period, as can seen in the equations (6) & (7).

$$k_e = m y \quad 6$$

$$k_d = \frac{k a_o}{(1 - \varepsilon)} \quad 7$$

As it can be seen, the first interval is a linear function with time, but the second interval has an exponential expression. The normalized curve was get if you plot the fraction of mass of recovered volatile oil ( $E_t / E_\infty$ ) against the extraction time.  $k_e$  and  $k_d$  were calculated by means of nonlinear regression, the mass transfer coefficient  $k$  was calculated using  $k_e$  and  $k_d$  values.

## IV. RESULTS AND DISCUSSION

### 4.1 Effect of type of solvent on extraction yield

Extraction yield of the essential oil obtained from *Myrtus Communis L.* (leaves, flowers, and stems) using different solvents are shown in Fig.3 to Fig.5.

As it can be seen in these figures, the extraction time is an important parameter for extraction process. It helps in deciding the maximum extraction yield when equilibrium is reached. Furthermore, any increases in extraction time have no effect in increasing the oil yield. In general the maximum extraction yield for n-hexane (60.97%) is greater than other solvent (53.1%, 46.8%), because this solvent has higher polarity than others used. This results is agree with Saxena et al.[13]

Extraction by water is a very clean method; this solvent has additional advantage versus other solvents in separation of essential oil from the collected product because water has a higher density compare with other solvents. Then maximum yield of extraction by water 46.8% is good, with the many advantages of this solvent inexpensive, green solvent, nontoxic, etc.[9]

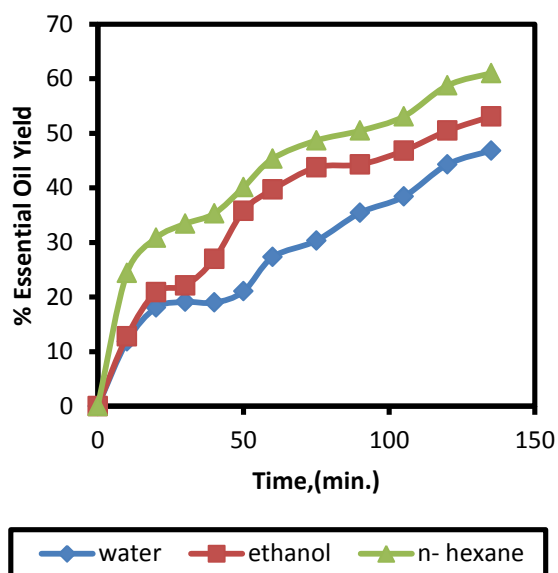


Fig. 3: The effect of different solvent on oil extraction yield with time for *Myrtus Communis* L. leaves.

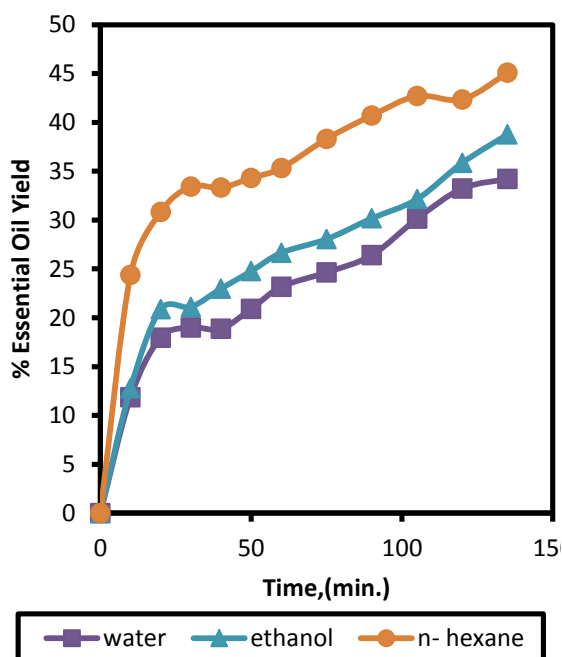


Fig. 4: The effect of different solvent on oil extraction yield with time for *Myrtus Communis* L. flowers.

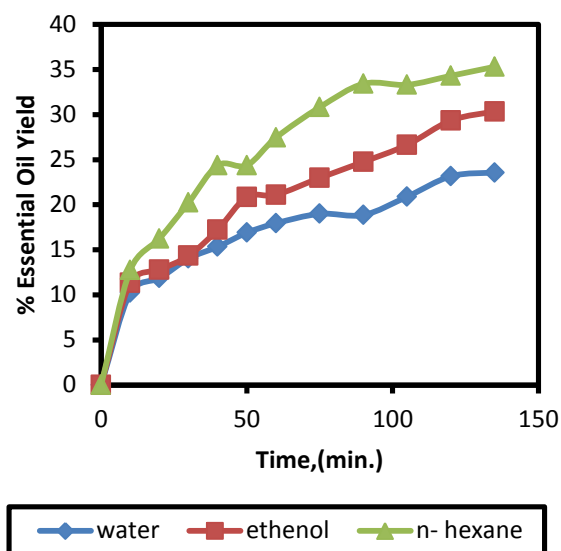


Fig. 5: The effect of different solvent on oil extraction yield with time for *Myrtus Communis* L. stems.

#### 4.2 Effect of the *Myrtus Communis* L. parts on extraction yield

Extraction yield of the essential oil obtained from *Myrtus Communis* L. ( leaves, flowers, and stems) using n-hexane is shown in Fig.6.

As it can be seen in this figure, there is varying in quality and yields from one organ to another as characterized for the three parts of *Myrtus Communis* L. plant. Since leaves can give the best extraction oil yields ( 60.97%) in compare with flowers (45.3%) and stems (35.6%). Leaves contained high volatile cells than other parts of plants that can be broken and liberated the required essential oil to extract this is agree with Pereira et al.[8].

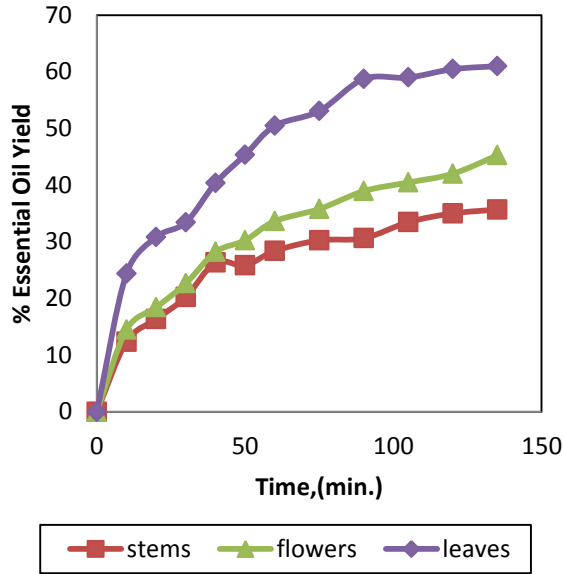


Fig. 6: The effect of different parts of *Myrtus Communis L.* plant on oil extraction yield with time for n- hexane as solvent.

#### 4.3 Extraction kinetics

The analysis of the resulted curves in Fig. 7 to Fig. 9 indicates the extraction kinetics for different solvent on oil extraction yield with time for *Myrtus Communis L.* leaves.

It can be noticed that the rate of oil extraction yield is increases rapidly first while followed by a gradual increase. This is due that plant matrix is exposed to the fresh solvent, the free oil on the surface of particles extracted quickly leading to rapid extraction rate. Since, in the starting of extraction process the oil concentration in the solvent is low, the oil diffuses very fast from the solid phase to the liquid phase due to the external mass transfer mechanism, this represent the first period of extraction. After that, the concentration of oil will increase in the solvent with increasing time so; a decreasing in the rate of diffusion will take place. The extraction yield of *Myrtus Communis L.* essential oil will reach a constant value even by increasing the time of extraction, this value is named the maximum amount of extractable oil. Equilibrium will reached, which represent the second period of extraction. This results are in agreement with the results which obtained by Franco et al.[17]

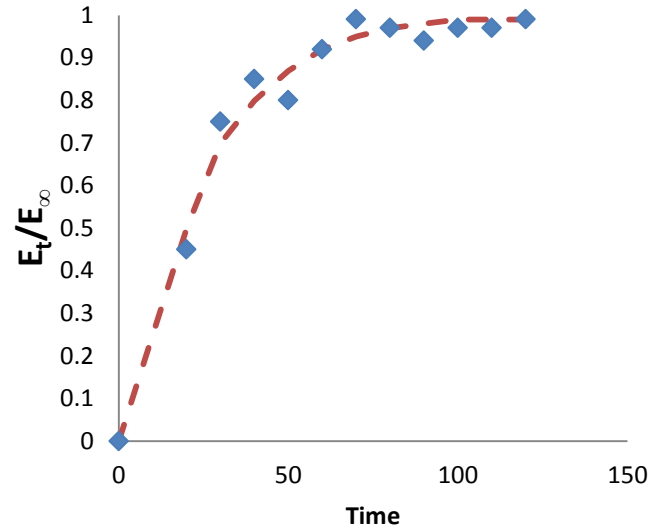


Fig. 7: Extraction kinetics of *Myrtus Communis L.* essential oil using n-hexane as a solvent. ( -- theoretical, ◆ experimental)

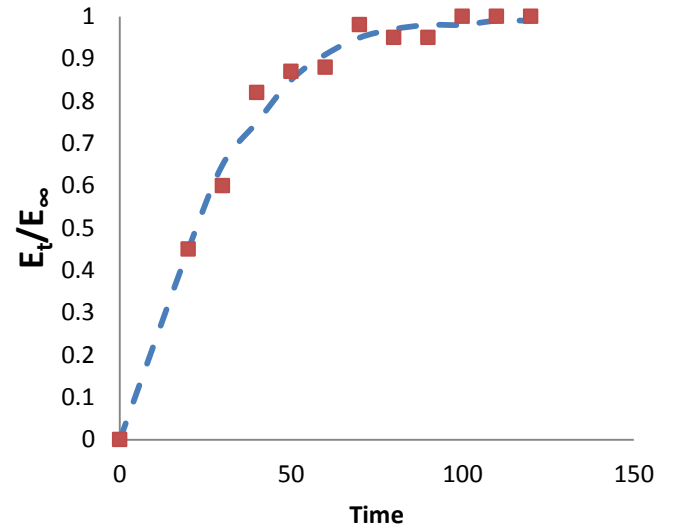
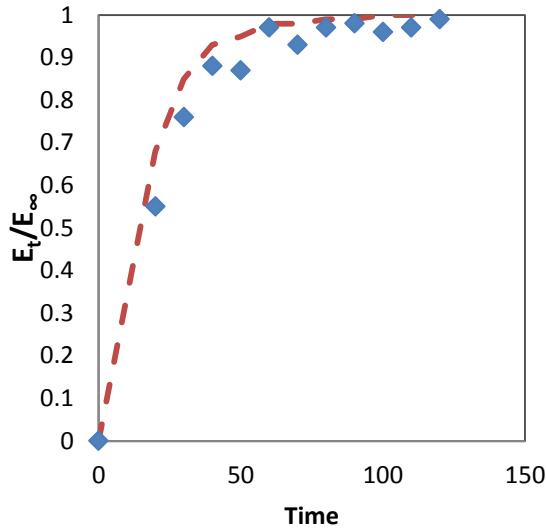


Fig. 8: Extraction kinetics of *Myrtus Communis L.* essential oil using ethanol as a solvent. ( -- theoretical, ■ experimental)



**Fig. 9: Extraction kinetics of *Myrtus Communis* L. essential oil using water as a green solvent. ( -- theoretical, ◆ experimental)**

The maximum yield of extracted essential oil and mathematical model parameters  $k_e$ ,  $k_d$  and  $k$  are shown in Table 1. From this table mass transfer coefficients for extraction using water as a green solvent has a higher value than the other solvents, this due to the stable physical properties of water in camper to the other solvents with changing of the temperature during extraction process.

**Table 1: Maximum extraction yield and mass transfer coefficient values for essential oil extracted from *Myrtus Communis* L. plant.**

Solvent	Plant part	% maximum yield	$k_e$	$k_d$	$k$
			g/min	min <sup>-1</sup>	g/min.cm <sup>2</sup>
n-hexane	leaves	0.610	0.0612	0.0275	0.0393
	flowers	0.451	0.0553	0.0256	0.0370
	stems	0.353	0.0436	0.0163	0.0327
ethanol	leaves	0.531	0.0575	0.0241	0.0358
	flowers	0.387	0.0491	0.0195	0.0343
	stems	0.303	0.0382	0.0142	0.0268
water	leaves	0.468	0.0753	0.1225	0.0448
	flowers	0.342	0.0606	0.1170	0.0417
	stems	0.236	0.0452	0.1151	0.0388

## V. CONCLUSIONS

The present study demonstrated the extraction of essential oil from three parts of *Myrtus Communis* L. plant ( leaves, flowers, and stems) using packed bed column with different solvents ( n-hexane, ethanol and water). N-hexane has the greatest oil extraction yield (60.97%), while water gives a moderated extraction yield (46.8%) in addition water is a green solvent and the produced essential oil may be used in many food and pharmaceutical industries. The main advantage of the mathematical model is it can be simulated the extraction curves with the mass transfer coefficients ( $k_e$ ,  $k_d$  and  $k$ ). The value of these coefficients is differenced due to the type of the solvent and the part of the plant used.



### Nomenclature

$a_o$	[m <sup>2</sup> /kg]	Superficial area
$E_t$	[kg]	Amount of extracted oil at time t
$E_{\infty}$	[kg]	Maximum amount of extractable oil
$k$	[kg/min.m <sup>2</sup> ]	Mass transfer coefficient
$k_d$	[min <sup>-1</sup> ]	Constant for to the diffusion controlled interval
$k_e$	[kg/min]	Constant for the equilibrium interval
$k_f$	[m/min]	Mass transfer coefficient in fluid phase
$k_s$	[m/ min]	Mass transfer coefficient in solid phase
	[kg/ min]	Solvent flow rate
$N$	[kg/m <sup>3</sup> . min]	Flux of solute
$t$	[min]	Extraction time
$u$	[m/ min]	Superficial fluid velocity
$x$	[kg/kg]	Mass ratio in solid phase
$x_s$	[kg/kg]	Mass ratio of solute in solid phase
$y^*$	[kg/kg]	Equilibrium mass ratio in fluid phase
$y$	[kg/kg]	Mass ratio in fluid phase
		Porosity (bed void fraction)
$\rho_f$	[kg/m <sup>3</sup> ]	Solvent density
$\rho_p$	[kg/m <sup>3</sup> ]	Particle density
$\rho_s$	[kg/m <sup>3</sup> ]	Solid density, bulk density

### References

- [1] W. A. Wannes, B. Mhamdi, J. Sriti, M.B. Jemia, " Antioxidant activities of the essential oils and methanol extracts from myrtle (*Myrtus communis* L.) leaf, stem and flower", Food and Chemical Toxicology, 48, (2010), pp.1362–1370
- [2] G. K. Oloyede, I. A. Oladosu and A. F. Shodia, "Chemical composition and cytotoxicity of the essential oils of *Crinum ornatum*(Ait.) Bury", African Journal of Pure and Applied Chemistry, (2010), Vol. 4(3), pp. 035-037.
- [3] A. Sepici, I. Gürbüz, C. Çevik , E. Yesilada, " Hypoglycaemic effects of myrtle oil in normal and alloxan-diabetic rabbits", Journal of Ethnopharmacology, 93, (2004), pp. 311–318
- [4] B. Berka-Zougalia, A. Hassanib, C. Besombesa, "Extraction of essential oils from Algerian myrtle leaves using instant controlled pressure drop technology", Journal of Chromatography A, 1217, (2010), pp. 6134–6142
- [5] N. Hayder, I. Skandrani, S. Kilani, I. Bouhlel, A. Abdelwahed, "Antimutagenic activity of *Myrtus communis* L. using the Salmonella microsome assay", South African Journal of Botany, 74, (2008), pp 121–125.
- [6] A. Zermanea, O. Larkecheb, A.-H. Meniaib, C. Cramponc, E. Badensc, " Optimization of essential oil supercritical extraction from Algerian *Myrtus communis* L. leaves using response surface methodology", Journal of Supercritical Fluids, 85, (2014), pp. 89– 94

- [7] K. Assamia, D. Pingretc, S. Chemata, B.Y. Meklatia, " Ultrasound induced intensification and selective extraction of essential oil from *Carum carvi* L. seeds", Chemical Engineering and Processing, 62, (2012), pp. 99– 105
- [8] P. Pereira, G. Bernardo-Gil, M. J. Cebola, " Supercritical fluid extracts with antioxidant and antimicrobialactivities from myrtle (*Myrtus communis* L.) leaves. Response surface optimization", J. of Supercritical Fluids, 83, (2013), pp. 57– 64
- [9] A. Filly, A. S. Fabiano-Tixier, C. Louis X. Fernandez, "Water as a green solvent combined with different techniques for extraction of essential oil from lavender flowers", Comptes Rendus Chimie, 19, (2016), pp. 707-717
- [10] J. Kabuba and R. Huberts, " Steam Extraction of Essential Oils: Investigation of Process Parameters", The Canadian Journal of chemical Engineering, Vol. 87, (2009), PP. 915-920.
- [11] G.R. Boucard, R.W. Serth, "Practical design of a distillation plant for the separation of essential oils from aromatic row materials", Texarome Inc., vol.(5), (2005).
- [12] M. Oligae, R. Sean, J. Androoz, "Hexane solvent extraction: Definition, Glossary details", Journal of Chromatography A, vol.(11), pp. 30-45.
- [13] D.K. Saxena, S.K. Sharma and S.S. Sambi, "Comparative Extraction Of Cottonseed Oil By N-Hexane And Ethanol", ARPN Journal of Engineering and Applied Sciences, Vol. 6(1), (2011), PP. 84-89.
- [14] E. Ghasemi , F. Raofie, N. M. Najafi, "Application of response surface methodology and central composite design for the optimization of supercritical fluid extraction of essential oils from *Myrtus communis* L. leaves", Food Chemistry, 126, (2011), pp. 1449–1453
- [15] E. Cassel, R.M.F. Vargas, N. Martinez, D. Lorenzo, "Steam distillation modeling for essential oil extraction process", Industrial Crops and Products, 29, (2009), pp.171-176.
- [16] V.B. Xavier, R.M.F. Vargas, E. Cassel, A.M. Lucas, M.A. Santos, " Mathematical modeling for extraction of essential oil from *Baccharis* spp. by steam distillation" , Industrial Crops and Products, 33, (2011), pp. 599-604.
- [17] A. Franco-Vega, N. Ramirez-Corona, E. Palou, A. Lopez-Malo, "Estimation of mass transfer coefficients of the extraction process of essential oil from orange peel using microwave assisted extraction", Journal of Food Engineering, 170, (2016), pp. 136-143
- [18] H. Sovov, S.A. Aleksovski, "Mathematical model for hydro-distillation of essential oils", Flavor Fragr. J. 21, (2006), pp. 881-889.

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