# Modeling the extraction process of pomelo (citrus maxima (burm.) Merr.) Peel essential oil by steam distillation on a production scale (500kg/batch)

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**Abstract.** The development of larger-scale extraction processes depends on process kinetics. A production scale of around 500 kg/batch was used in this study to experimentally extract pomelo essential oil by steam distillation. In this study, three models including Model of simultaneous washing and diffusion, Model of instantaneous washing followed by diffusion, and Model of diffusion without washing are considered for evaluation. The most appropriate kinetic model kinetics were chosen using the coefficient of determination ($R^2$) and percentage of variance (%q). The findings demonstrate that the analyzed model of concurrent washing and diffusion suited the experimental data. The results show that in this model, the value of $R^2$ = 0.99795 and q = 0.89285 ± 0.0061, which has the highest value of the three models. The extraction process through this production system was successfully described using a kinetic model, which gave information on the extraction process.

## 1. Introduction

The enormous number of functional, dietary, and biological benefits of pomelo (*Citrus grandis* or *Citrus maxima*) are well known in many nations [1-3]. Since they have been scientifically shown to have medicinal potential and are safe for humans, many components of this plant, including the leaves, pulp, and peel, have been utilized in traditional medicine for decades. Compounds from citrus maxima have medicinal benefits [2, 4]. The plant has been shown to have a wide range of pharmacological actions, including cytotoxic, hepatoprotective, nephroprotective, antibacterial, anticancer, antioxidant, anti-inflammatory, antiepileptic, stomach tonic, and heart stimulating properties [5, 6]. Flavonoids, polyphenols, coumarins, limonoids, acridone alkaloids, essential oils, and vitamins are frequently added to fruits to enhance their pharmacological effects on the body. Vitamin-enriched fruit is highly nutritious. It contains minerals that support bone health and correct bone formation and the body's electrolyte balance, such as calcium, phosphorus, salt, and potassium [2, 7]. In addition, new research on the fruit's various components, such as its pulp, peel, leaves, seeds, and essential oils, may uncover additional pharmacological actions that could be helpful to people [8–9].

Pomelos have a low percentage of edible sections, one of its issues (being fruit pulp). *Citrus t*rees frequently have this characteristic because of their thick bark, which often contributes up to 50% of the byproducts. It is challenging to cope with a significant amount of by-products from this agricultural product's peel processing due to the industrial processing method, which solely consumes fruit pulp. The suggested technique for generating essential oils from fruit peels demonstrates the viability of processing leftovers. These essential oils are created as valuable commodities and are frequently utilized in food, medicine, and cosmetics [10–12]. Most countries with advanced science, including Europe, the US, Japan, and Korea, have conducted research on essential oils; the findings have been applied to manufacturing industries that contribute significantly to the economy and society.

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There are numerous ways to extract essential oils from plants, including mechanical methods, steam terminal distillation methods, extraction methods using volatile solvents, and high-tech extraction processes including $SCO_2$, microwaves, and ultrasonics [12–15]. The benefits and drawbacks of each approach vary. For instance, cold pressing is a great mechanical way to gather citrus essential oils with a variety of components. However, this method has limited yields and necessitates washing the fruit to remove its color after pressing. Although solvent extraction solves performance concerns, it has the drawback that solvent removal is required after extraction, which is regarded as a middle stage. Currently, high-tech processes like $SCO_2$, microwaves, and ultrasonics that use reusable solvents, good-quality essential oils, and high efficiency completely comply with green chemistry guidelines. The methods' energy costs and practical applicability are constrained, though. The steam distillation process is taken into consideration to solve the aforementioned issue. A common technique for extracting essential oils from plant tissues is steam distillation. Due to its ease of installation and operation, as well as its viability on an industrial scale. It is the simplest and most popular technique for obtaining essential oils from aromatic and therapeutic plants. The plant matter and water don't come into touch throughout this process. The essential oil of the material is drawn to the boiling steam, where it is condensed. With this technique, the essential oil is partially protected from high temperatures because when the material is soaked, it comes into contact with steam, which protects it from them. Additionally, the evaluation of the laboratory extraction procedure connected to the production is necessary for the current applied study.

In terms of the extraction process, the kinetics of essential oil extraction shows the importance of the basic understanding and the possibility of optimization, control, and design of industrial distillation processes career [16-17]. On the other hand, the scientific publications on this issue are not wide. The lack of material to perfect extraction technology with high recovery causes the problem of excess energy supplying the process and the waste of domestic raw materials. The kinetic study of pomelo peels essential oil extraction by steam distillation on a production scale was carried out. The research not only meets a simple essential oil extraction process but also solves the problem of phase control and deepens the mechanism of the extraction technology. More specifically, it provides an in-depth overview of the production-scale extraction process that current applied studies want to focus on.

## 2. Materials and Methods

2.1. *Materials*

*Citrus maxima* (Burm.Merr.) is a fruit tree that is grown quite popularly in the Mekong Delta (Mekong Delta) in which it is concentrated in one area some provinces such as Ben Tre, Tien Giang, and Vinh Long. Pomelo fruit with green skin is spherical, weighing on average between 1.2 and 2.5 kg per fruit, with a peel that ripens to a green to yellowish green color. They also have an aroma, are seedless to slightly grainy, and have a meat to fruit ratio of more than 55%. After being selected, fruits are preliminarily treated and peeled off. The weight of peels accounted for approximately 15–20% weight of the fruit. Before distillation, the albedo was removed from pomelo peel.

2.2. *Extraction of pomelo oil by steam distillation*

The process of steam distilling pomelo peels essential oil was shown in Figure 1. After harvesting, raw materials are pre-treated, removing dirt, damaged parts, and carefully separating the green skin of pomelo. Next, 500 kg of pomelo peel was processed for size and put into the extraction system. Heat the system until steam appears, then start the timer. After the extraction process, the vapor mixture passes through the condenser to obtain liquids consisting of essential oils and water. After the extraction process, crude essential oil was obtained from this mixture. Essential oil with a bit of water should be anhydrous with $Na_2SO_4$ salt to get pure essential oil.

**Fig. 1.** Pomelo essential oil extraction system

2.3. *The kinetics modelling on extraction process*

Building suitable kinetic models applied on the distillation of pomelo essential oil based on two mechanisms [18, 19]:

(1) Washing: Using raw materials' surface to extract essential oils

(2) Diffusion: when an essential oil moves from the inside of a substance to its surface without being blocked by membranes or other barriers present in the material

The kinetic model of the procedure for removing the essential oil from the skin of a green pomelo, which includes three models as follows, is based on these two mechanisms:

Model of simultaneous washing and diffusion:

$$\frac{q}{q_\infty} = 1 - fe^{-k_1 t} - (1-f)e^{-k_2 t} \tag{1}$$

Model of instantaneous washing followed by diffusion:

$$\frac{q}{q_\infty} = 1 - (1-f)e^{-k_2 t} \tag{2}$$

Model of simple diffusion without washing:

$$\frac{q}{q_\infty} = 1 - e^{-k_2 t} \tag{3}$$

Calculation of parameters: The parameters of the model: k, $q_\infty$ and $R^2$ are determined by linear regression method based on experimental values based on the correlation graph q and t. Data processing using OriginPro 9 software.

The statistical values are determined as follows:

Coefficient of determination:

$$R^2 = 1 - \frac{\sum_{i=1}^{N}(q_{\infty,exp} - q_{\infty,cal})^2}{\sum_{i=1}^{N}(q_{\infty,exp} - q_{\infty,mean})^2} = \frac{\sum_{i=1}^{N}(q_{\infty,cal} - q_{\infty,mean})^2}{\sum_{i=1}^{N}(q_{\infty,cal} - q_{\infty,mean})^2 + \sum_{i=1}^{N}(q_{\infty,cal} - q_{\infty,exp})^2} \tag{4}$$

Percentage difference between the essential oil quantity measured at saturation and the essential oil quantity estimated from the model:

$$q\% = \frac{q_{\infty,\mathrm{exp}} - q_{\infty,cal}}{q_{\infty,exp}} \times 100 \tag{5}$$

In there,

$q_\infty$, exp (%): The quantity of essential oil collected by the experiment

$q_\infty$, cal (%): The quantity of essential oil obtained from the model using Origin 9 software

$q_\infty$, mean (%): The average value of $q_t$;

N: is the observation coefficient.

The kinetic model was chosen to describe the experimental data of the process based on the calculation findings, comparison of three kinetic models, and coefficient of determination $R^2$, %q, and rate constant k. Citrus peel extraction mechanisms, quick or slow extraction rates, and process rate constants are all factors in the process of distilling citrus peel essential oil via distillation.

2.4. *Data processing methods*

Each experiment was conducted three times, with the results shown as standard deviations. Using Microsoft Excel, the results' standard deviation is determined. Utilizing Origin 9.0 software, process kinetics-related experimental data were examined.

## 3. Results and Discussion

Three experimental kinetic models were used to fit the recovery efficiency of pomelo essential oil at various distillation periods (from 30 minutes to 3 hours). Figures 2, 3, and 4 show how the essential oil content rose over time. The hypothesized essential oil extraction mechanism and the proposed kinetic models were validated using the nonlinear form of equations (1) - (3) over time (Figure 2 – 4).

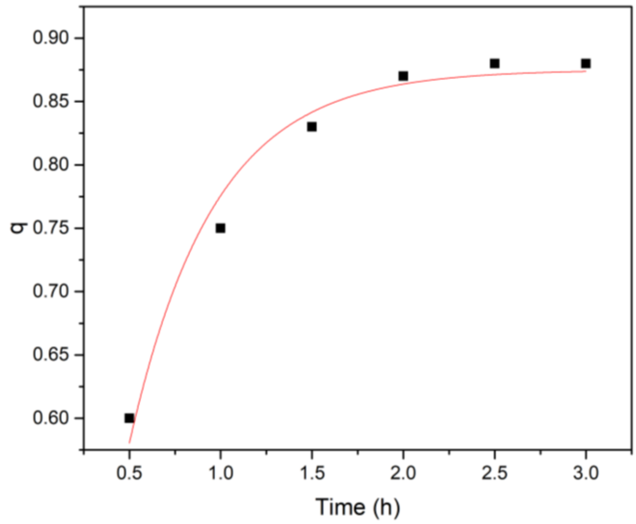


**Fig. 2.** Simultaneous washing and diffusion model of pomelo essential oil extraction at different time

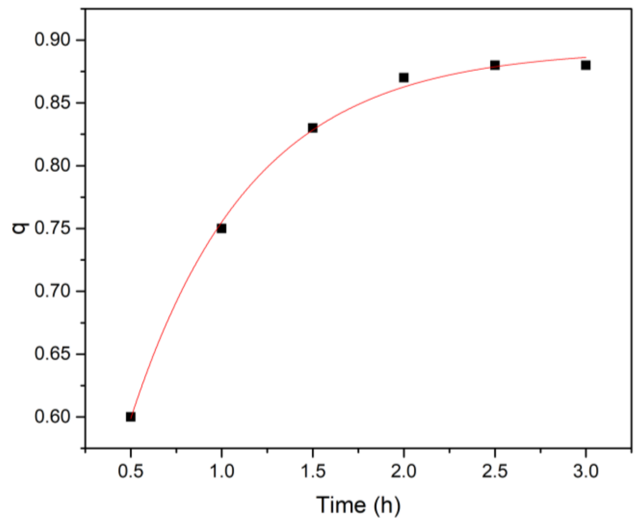


**Fig. 3.** Instantaneous washing followed by diffusion model of pomelo essential oil extraction at different time

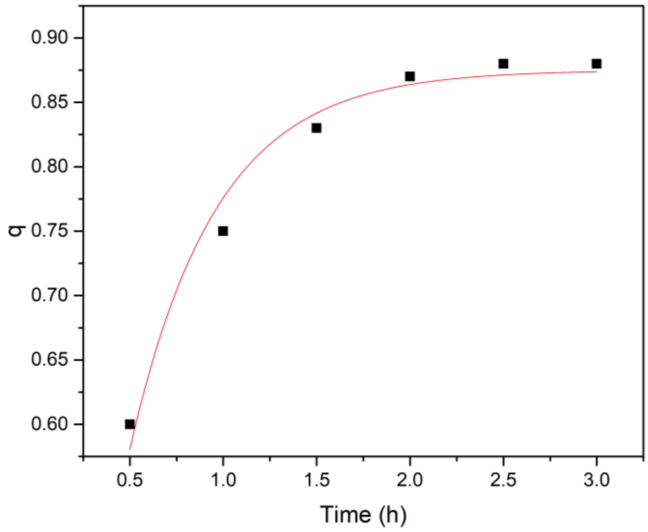


**Fig. 4.** Diffusion without washing model of pomelo essential oil extraction at different time

Figure 2 – 4 illustrates how the experimental data points appear to be dispersed throughout the curve, suggesting that the kinetic model equations are appropriate for describing the experimental data. Further examination of the experimental kinetic curves for extraction revealed that there are typically two stages to the extraction process: the rapid extraction (washing stage), during which the essential oils are located on the outer surface of the washed material, and the slow extraction (diffusion stage), during which the essential oils are transferred from the interior of the material to the outside environment. The output of the recovered essential oil slowly and gradually increases during the second stage. The sigmoid or S-shaped curves on this scale that depict the variation in pomelo peel essential oil output are upward curving. This is consistent with earlier experimental results (Figures 2-4), which demonstrate that the production of essential oils grows quickly in the early stage (i.e., rapid distillation, immediate washing phase), slows down until virtually constant oil yield, and then increases again (i.e. slow, diffuse distillation phase) [20, 21].

Three kinetic stages can be seen for the majority of the time points, as shown in Figure 2-4. First, the extraction yield increases quickly between 30 and 60 min, then slowly as the extraction time increases until a constant time is reached after 2 h of extraction. The second stage was where more than 90% of the total recovered oil was removed (after 2 h).

The essential oil fraction was extracted during the washing step with unhindered and hindered diffusion. To assess the link between experimental data and the model, the coefficient $R^2$ is a frequently utilized parameter (table 1). Nevertheless, practically all models under various extraction conditions attained very high $R^2$ ($R^2 > 0.97$) and these models can be thought of as being appropriate to characterize the process kinetic. The next evaluation coefficient, value%q, is utilized to determine which model is most suited. This metric measures the percentage difference between the experimental and experimental values for the essential oil content at saturation. figured using the model. The fast wash, followed by diffusion, 2.3 model has better experimental data than the (2) and (3) models, according to the $R^2$ and%q values. The greater $R^2$ coefficient of determination and the discrepancy between the calculated and real amounts of essential oil falling within the permitted range (less than 5%) are the causes of this outcome. These findings demonstrate that %q is a useful support factor when evaluating the model's fit.

Additionally, the model of simultaneous washing and diffusion without interference in the nonlinear form, which has an $R^2$ coefficient of 0.99795 and %q of 0.89285 and 0.0061%, is chosen as the appropriate model and form. The extraction kinetics are well captured by the non-linear model. Given the aforementioned information, it is reasonable and trustworthy to interpret the experimental data of the extraction process using the nonlinear equation of the instantaneous wash model followed by diffusion.

**Table 1.** Kinetic parameters of pomelo essential oil extraction process

| Model | q | $k_1$ | $k_2$ | f | $Chi^2$ | $R^2$ | $Adj.R^2$ |
|---|---|---|---|---|---|---|---|
| Model of simultaneous washing and diffusion | 0.87506 ± 0.02338 | 2.17651 ± 1.21832E7 | 2.1765 ± 4.65547 x$10^6$ | -0.61596 ± 1.3806E9 | 6.7032 x$10^{-4}$ | 0.97805 | 0.94513 |
| Model of instantaneous washing followed by diffusion | 0.89285 ± 0.0061 | - | 1.51978 ± 0.11273 | 0.29529 ± 0.04088 | 4.16586 x$10^{-5}$ | 0.99795 | 0.99659 |
| Model of diffusion without washing | 0.87506 ± 0.01052 | | 2.17649 ± 0.12823 | | 3.3516 x$10^{-4}$ | 0.97805 | 0.97257 |

Based on the assumptions of model (1-3) with the combination of experimental data, conclusions can be made. The kinetics of removing pomelo peel was first described using a concurrent unconstrained washing and diffusion model. Second, the two processes that make up the primary process in the kinetic model of direct water extraction are washing and diffusion. When we refer to washing, we mean the process of cleaning the raw material's surface both internally and externally of the essential oil. At the start of the extraction process, the amount of essential oil increases quickly during this stage. The diffusion phase, which is indicative of the gradual extraction of essential oils, is the next stage that is crucial to the extraction mechanism. The process involves diffusing the essential oil from the pomelo peel's interior to its exterior, where it is then dispersed by the steam. Diffusion is the unrestricted mass transport of essential oils from ruptured organs. During distillation, the essential oil output slowly increases during the diffusion phase [22]. Indeed, the current extraction processes of pomelo essential oil are described by instantaneous washing followed by diffusion. The extraction processes were demonstrated from experimental data at experimental scales of 100g [23] to 1000g scales [21], pilot scales of 5000g [18], and, more specifically, in this study at a production scale of 500 kg. The aforementioned arguments demonstrate that Equation (2) accurately captures the experimental data of pomelo essential oil extraction across all production scales.
The kinetic model for instantaneous simultaneous washing and diffusion at the optimal location for steam distillation of pomelo essential oil was as follows:

$$\frac{q}{q_\infty} = 1 - (1 - 0.29529).e^{-1.51978t}$$

Where q was the amount of essential oil that was obtained in the material up to time t, and $q_\infty$ was the amount of essential oil that was obtained up to saturation.

## 4. Conclusions

The extraction process of pomelo peel essential oil by steam distillation on an experimental production scale (500 kg/batch) was examined in this study. The estimation and simulation of the extraction process were done using these models. These models were used to estimate and simulate the extraction process. The results show that the instantaneous washing followed by diffusion model was the most suitable for the production-scale pomelo essential oil extraction process, with an $R^2$ coefficient of 0.99659 and a process rate constant of 1.52 $min^{-1}$.

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