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Kinetic Modeling of Hydrodistillation Extraction of Essential Oils from Roots and Leafy Stems of *Elionurus Hensii* Schum from Congo

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Abstract

Elionurus hensii is a plant belonging to the Poaceae family, widely used in food and traditional medicine as a tea drink. It strengthens the immune system, fights coughs, and alleviates joint pain. Due to the therapeutic and nutritional interest of its Essential Oil, the question of its production in terms of extraction yield arose. This work aimed to model the kinetic extraction of essential oil from the plant *Elionurus hensii* K. SCHUM from Congo in order to optimize its yield and develop the most suitable technologies for producing its essential oil at higher contents. In this study, the extractions were carried out by hydrodistillation on the roots (continuous extraction) and stem-leaves (continuous and discontinuous extraction) of the plant at different times corresponding to 15, 30, 45, 60, 90, 120, 150, 180, 210 and 240 minutes. The extraction results gave maximum contents of 1.81% on the roots and 2.7% on the stem-leaves for an extraction time of 180 min. The modified Monod models and the pseudo-diffusional Mafarat and Bélliard model showed a good fit with the experimental data. The values of R^2 are such that $R^2 = 0.995$ for roots and $R^2 = 0.9922$-0.9928; 0.9726-0.978 for stem-leaflets respectively for the modified Monod models and the pseudo-diffusional Mafarat and Bélliard model. These two models were therefore found to be the best suited for the extraction kinetics of essential oil from the *Elionurus hensii* plant.

Keywords

Modeling; Extraction kinetics; Essential oil; *Elionurus hensii*; Hydrodistillation

1. Introduction

Plants now represent a significant market share in many fields, including cosmetics, therapeutics, and agri-food [1]. Today, players in these different fields, after having favored the use of synthetic molecules because they are less expensive, tend to turn to natural molecules, therefore the constituents of plants. Aromatic plants represent an inexhaustible source of food and especially effective traditional remedies thanks to the active ingredients they contain: flavonoids, polyphenols, essential oils, etc. [2]. They also have a considerable asset thanks to the progressive discovery of the applications of their essential oils, as well as their use in other areas of economic interest [3]. On the market, their many uses mean that they are experiencing an increasingly strong demand. Indeed, if the demand for bioactive molecules is constantly growing, that of essential oil is even greater, thus allowing its production to increase. In this regard, the optimization of oil extraction will have to meet this expectation.

The economic importance of these essences due to their biological and therapeutic properties and their use in the treatment of certain diseases for which synthetic chemicals are less and less active or in the preservation of food against oxidation as alternatives to synthetic chemicals are attracting more interest from researchers.

Much attention has been paid in recent years to natural compounds. Recently, there has been a global interest in the extraction of beneficial compounds from plant by-products. Many studies have launched into the search for bioactive molecules and are now exploring the possibility of their transformation either into an ingredient that can be incorporated into different food products to make functional foods or foods, or into cosmetics or pharmaceuticals [4]. These functional foods can provide benefits for human health in response to the overconsumption of certain foods that can have health pathologies. The consumption of functional foods has subsequently spread throughout the world [5]. Encouraged by the growing interest in food by consumers [6], the essential oil sector has developed into a flourishing sector.

In Congo, there are craft industries and know-how already exists since many small workshops extract and market these essences. Today, it is a question of developing it to make this sector an additional source of income and a tool for sustainable development that targets the agri-food market and many others. Conventional processes for extracting these natural substances from plants require real know-how in order to have them in large quantities. Mastering the extraction processes that allow the optimization of the extraction of bioactive substances has become very useful because it meets scientific and technological challenges.

Plants, sources of medicine, have the advantage of being accessible to all and particularly to developing countries [7]. Some aromatic plants mentioned in the literature have various uses: therapeutic and food (medicine and food), such as *Occimun basilicum, Occimum gratissimum, Cymbopogon citratus or Lippia multiflora*... [8, 9]. In Congo, a country with remarkable forest wealth [10], there is a plant called *Elionurus hensii* belonging to the Poaceae family, widely used in food and traditional medicine as a tea drink. It strengthens the immune system, fights coughs, and relieves joint pain [11]. Its essential oil contains active ingredients that are greatly appreciated for their biological properties: antimicrobial, antioxidant [12-14], etc. As these properties were discovered, and the therapeutic and nutritional interest of its EO, the question of its production from the point of view of extraction yield arose. Because few studies have been done on the kinetics of extraction of the essential oil of this plant. While extraction is the first operation applied to obtain these essential oils.

Essential oils are increasingly appreciated for their therapeutic and industrial applications. Among these, *Elionurus hensii* SCHUM, an aromatic food and medicinal plant from Congo, has demonstrated significant antimicrobial and antioxidant properties. This study focuses on optimizing the extraction process to maximize oil yield and explores suitable kinetic models to support industrial scalability.

2. Methods

2.1 Plant material

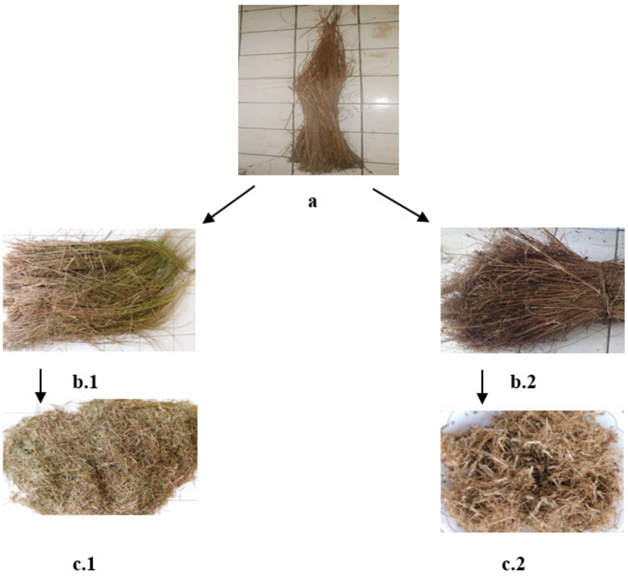

Figure 1. a) Whole plant, b.1) aerial parts, b.2) underground parts, c.1) cut aerial parts, and c.2) cut underground parts.

The plants were harvested during the months of February and April from Loukoko located in the Plateaux de cataractes in the Department of Pool. After harvesting, the aerial and underground parts of the plants are separated, cut into small pieces of about 2 cm, and dried at room temperature in a ventilated place in the shade to better preserve the molecules sensitive to heat and light. Figures 14 b.1 and 14 b.2 respectively represent the cut aerial and underground parts of the plant *Elionurus hensii* Schum K. (Figure 1a).

2.2 Extraction of essential oils

The extraction of the essential oil was carried out using a hydrodistillation device. Hydrodistillation was performed under atmospheric pressure at a boiling temperature of around 100°C. The main feature of this process is that there is direct contact between the boiling water and the plant material. Thus, a quantity of 100 g of plant material is introduced into a 2000 mL flask filled with distilled water up to two (2) thirds of its capacity so that the plant material is completely immersed in the water. The whole is then brought to a boil for a set period of time. After monitoring the extraction kinetics, the flask thus heated produces steam loaded with volatile products or essential oils. This steam condenses in contact with a refrigerant. The condensate is collected in a test tube and then poured into a separating funnel where the separation of the two immiscible phases is carried out: the aqueous phase and the organic phase. The organic phase composed of essential oil was treated with anhydrous sodium sulfate to remove all traces of water. The oil obtained is stored in a refrigerator in glass bottles wrapped with aluminum foil. The yield defined as the ratio between the mass of the essential oil obtained after extraction and the mass of the plant material used is calculated according to the [15] the following formula (1):

$$R_{EO}(\%) = \frac{M_{EO}}{M_{PM}} \times 100 \tag{1}$$

$R_{EO}(\%)$: Essential oil yield in percentage; M_{EO}: Mass of essential oil after extraction at time t; M_{PM}: Mass of plant matter

2.3 Monitoring of extraction kinetics

The purpose of extraction kinetics is to set the time required to extract the maximum amount of oil contained in a plant material and to avoid losses in time and energy [16]. This kinetics consists of determining the yield as a function of the duration of the extraction. In this study, two types of extraction were carried out: continuous and batch. For continuous extraction, the yield was determined at durations of 15, 30, 45, 60, 90, 120, 150, 180, 210 and 240 minutes. For batch extraction, the plant material was replaced or changed at each time. The operation was repeated several times under the same conditions.

2.4 Modeling of extraction kinetics

Mathematical modeling is used to predict the extraction process of essential oils from *Elionurus hensii* and to determine the optimal extraction parameters and process performance.

To model the kinetics of hydrodistillation extraction of essential oils from the stems-leaves and roots of *Elionurus hensii*, four models selected from the literature were used. These are the Mafarat and Béliard II models, the Mafarat and Béliard pseudo-diffusional model, the Monod model, and the power law model. In the literature, the authors have described several empirical models. Models whose application requires taking into account certain parameters such as the form of the plant material, the type of extraction used, the temperature and pressure of the operation, etc. were not taken into account in this study.

2.5 Statistical processing

The parameters of the statistical models for the modeling of the extraction of the essential oil of *Elionurus hensii* were processed using the Originpro2018 software and Excel 2013.

Two statistical parameters were taken as selection criteria for the best-fit models. These are the coefficient of determination R^2 and Chi-square (χ^2) and adj. R^2.

3. Results and discussion

3.1 Extraction yield of essential oil from *Elionurus hensii*

We recall that the extraction of the essential oil (EO) of *Elionurus hensii* was extracted by hydrodistillation in continuous and batch mode. Table 1 shows the yields obtained in essential oils from the parts of the plant studied. This extraction reveals maximum yields in EO of the order of 2.7% and 0.6% respectively in continuous and batch mode for the aerial part and 1.81% for the underground part in continuous mode. These results are higher than the yields obtained by [13], 1 to 2% on the stem-leaves and 0.5 - 0.9% for the roots. The same is true for those mentioned by [17] considering the species harvested during the same period of the year, namely: March (0.81%, 0.43%), April (0.89%; 0.51%), June (0.80%; 0.48%) and July (0.98%; 0.53%) for stem-leaflets and roots respectively. On the other hand, it is similar to the yield obtained by [18] which reaches maximum yields of 2.7-2.9% at the level of stem-leaflets but, the yield of 1.5% reported at the level of roots by the same authors remains slightly lower than the yield reported by the present study. Other results reported by [19], on the essential oil obtained by hydrodistillation of the same species show yields ranging from 0.4 to 0.9% on the stem-leaves which is also lower than our results. On the other hand, the discontinuous yields on the aerial part are relatively close to those given by [19]. This difference is quite normal since it can be attributed to several factors: the harvest period, environmental conditions (climate, geographical area, degree of drying), degree of fragmentation, and the extraction method [20].

Table 1. Extraction yield of essential oil from the stems-leaves and roots of *Elionurus hensii* in continuous and discontinuous mode

t(min)	15	30	45	60	90	120	150	180	210	240
Continuous extraction										
Stems-leaves	-	0,021	0,056	0,097	0,2	0,334	0,478	0,596	0,608	0,609
Roots	-	0,006	0,031	0,049	0,179	0,265	1,615	1,79	1,80	1,81
Discontinuous extraction										
Stems-leaves	0,07	0,419	0,868	1,568	4,268	5,038	5,998	6,009	6,011	6,017

3.2 Extraction kinetics

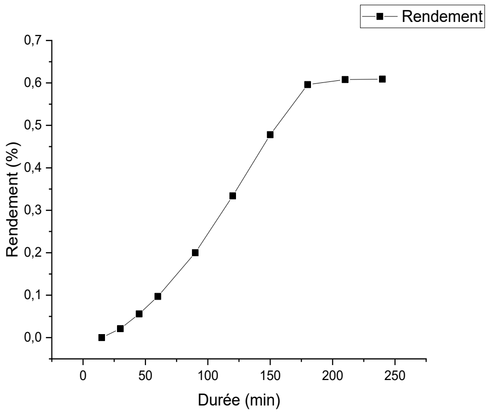

Figure 2a. Curve of the kinetics of essential oil extraction from discontinuous stem-leaves.

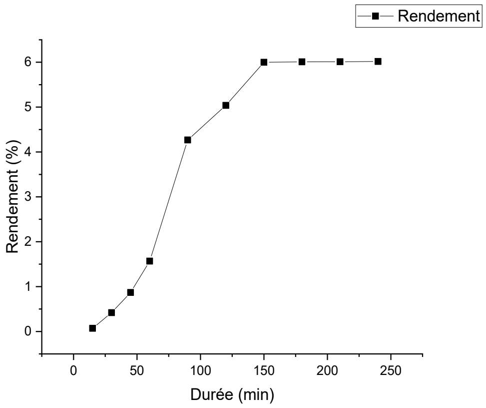

Figure 2b. Curve of the kinetics of essential oil extraction from continuous stem-leaves.

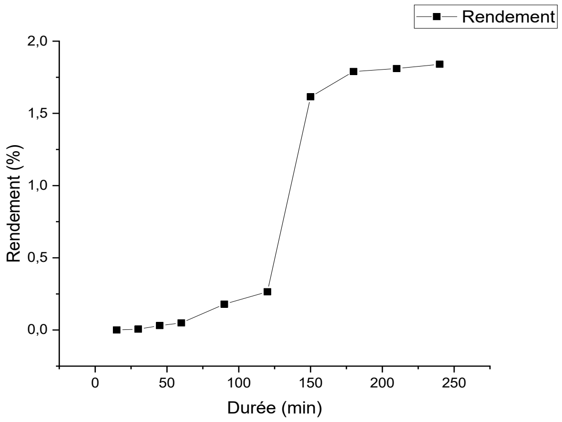

Figure 2c. The kinetic curve of continuous extraction of essential oil from roots.

The following Figures 2a, 2b, and 2c show the variation of the mass yield of essential oil of *Elionurus hensii* as a function of the extraction duration.

The monitoring of the extraction kinetics shows that it goes through three phases for the stem-leaflets and roots according to the appearance of Figures 2a, 2b, and 2c. These curves clearly show the general appearance expected of a classic extraction kinetics curve, which shows the variation of the oil mass yield as a function of the extraction duration. The first phase corresponds to heating where the extraction is almost zero or low. It is represented by a straight line. The second is an ascending straight line showing the ease of extraction from 15 to 60 minutes and from 30 to 60 minutes, respectively for Figures 2a and 2b. This rapid extraction process was observed just at the beginning of the extraction and then slowed down to a time t = 150 minutes and t = 180 minutes. This correlates with the conclusion of [21]. In their study on the extraction of *Terminalia catappa* L. seed oil by n-hexane, it was observed that the initial rapid extraction process was due to the free oil disposed on the surface of the plant. The oil exposed on the surface is soluble in the solvent and hence leads to rapid extraction. According to authors [22-25], the rapid extraction rate at the beginning and slow at the end of the extraction process could be explained by rapid initial washing action. The higher yield was obtained for a duration of 150 minutes and beyond. Whereas a lower yield was recorded at t = 15 minutes. Due to shorter diffusion paths.

In Figure 2c, the yields fluctuate during the first 45 minutes. Afterwards, a sharp increase in the yield is observed from 120 to 180 minutes and slowed down beyond 180 minutes, probably due to the solubility of the extract in the solvent when heating the plant material at the first level, followed by internal diffusion. The rapid extraction process was due to the presence of free oil on the surface of the solid. Since the plant was cut, the oil was exposed and this made the oil soluble in the solvent. Note that some extractions are limited only by solubility. Other extractions are limited only by the internal diffusion of the extract in the solvent. Indeed, the access of the solvent into the material to be extracted is particularly difficult. The extraction is carried out solely according to the diffusional parameters.

The increase in yield from 45 to 180 minutes is due to the washing of oil located on the surface of the plant material followed by a final slowdown phase where the extraction is limited by internal diffusion. It is observed beyond 150 minutes for Figure 2c and is characterized by a second level where the oil extraction yield reaches a constant rate or no longer changes significantly over time. The different figures show that the maximum oil is extracted in 180 minutes, which leads us to believe that the optimal hydrodistillation time would be 180 min although the roots require more heating time. This could be explained by the presence of heavy molecules.

3.3 Kinetic models of essential oil extraction from *Elionurus hensii*

The nonlinear kinetic curves from the models studied are illustrated in Figures 3a, 3b, and 3c below.

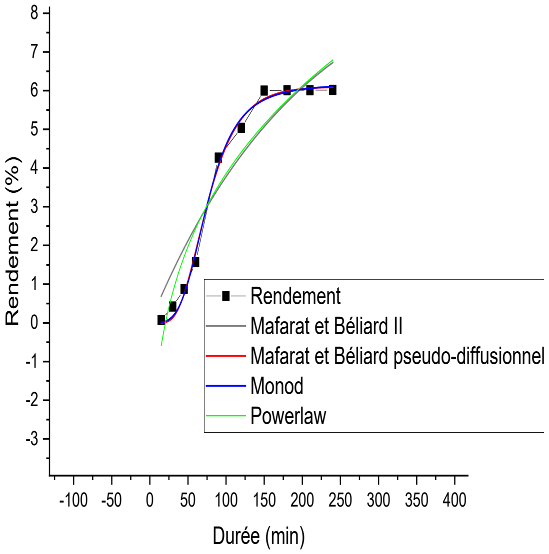

Figure 3a. Continuous stem-leaves modeling curve.

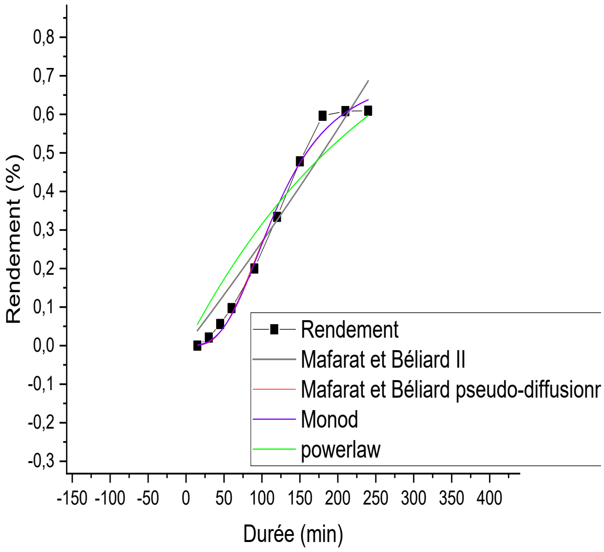

Figure 3b. Discontinuous stem-leaves modeling curve.

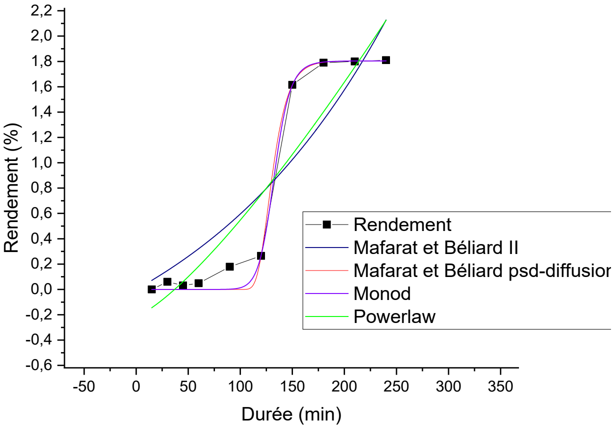

Figure 3c. Modeling curve of continuous extraction of HE from roots.

Examination of the figures of the modeling of the kinetics at the level of the stem-leaflets allows us to note that the extraction yields in oils obtained from the nonlinear models are generally close to those obtained from the experimental data with regard to the Mafarat and Béliard pseudo-diffusional models and the Monod model. On the other hand, the two other models are very far from the experimental data.

The curves of the Mafarat and Béliard pseudo-diffusional and Monod models are merged in both cases (discontinuous and continuous extractions). They are very close to that of the experiment in the entire domain of a discontinuous extraction, whereas in the continuous extraction, the two curves differed little with the experimental data from 150 to 210 minutes just at the end of the extraction process.

Table 2. Parameters of the models used

Model settings	**Extraction mode**		
	Continuous	Continuous	Discontinuous
	Part of the plant used		
	Roots	Leafy stems	
	Monod model		
$\mathbf{Y_{max}}$	$5{,}868.10^{-14} \pm 3{,}97.10^{-21}$	0,71743±0,05192	16,865 ±9,272
$\mathbf{K_m}$	$7{,}58.10^{-16} \pm 5{,}13.10^{-23}$	120,0289±8,1805	354,677 ±286,422
	Modified Monod model		
$\mathbf{Y_{max}}$	1,80415±0,04416	0,71743±0,05192	0,71743±0,05192
$\mathbf{K_m}$	132,689±2,0975	120,0289±8,1805	120,0289±8,1805
n	17,327±2,45650	2,998±0,39700	2,998±0,397000
	Model of Mafarat and Béliard II		
$\boldsymbol{C_l(\infty)}$	-0,8866±1,1313	-3,0168±7,870	7,9140±12,010
$\boldsymbol{\lambda}$	-0,0051±0,004	$-8{,}542.10^{-4}$	0,00295±0,005
	Mafarat and Béliard Pseudo-diffusional model		
$\boldsymbol{C_l(\infty)}$	1,8042±0,04243	0,68904±0,0428	2,89094±0,1566
$\boldsymbol{\lambda}$	0,09498±0,01574	0,01724 ±0,00306	0,08474±0,00365
c	170827,40052±329	4,8281±1,3598	212,0488±475,132

In both models, the deviations of the points are observed from 60 and 90 minutes respectively in the discontinuous and continuous curves (Figures 3a and 3b). On the other hand, these deviations remain smaller at the very beginning of the extraction operation. This shows that initially, the transfer of matter is governed mainly by diffusion. This differs from Figure 3c on the roots whose extraction is slow with values that fluctuate from 0 to 90 minutes (first level). This shows that the beginning of the extraction is limited by the solubility of the extract in the solvent. The second phase is represented by a straight line that shows the speed of the extraction.

Table 2 shows the parameters of the different models used.

The values of the kinetic parameters of the Monod model are shown in Table 2. It was observed that the Ymax and Km parameters for the Monod model varied depending on the extraction mode. The increase in Ymax and Km was associated with the increase in the oil yield observed with respect to time. Batch extraction of essential oils from roots and stem-leaflets gave higher values compared to those reported by [26]. However, continuous extraction of essential oils from stem-leaflets gave lower Ymax values than the value reported by the same authors (km = 5.88 min^{-1}). Some insignificant differences were recorded between the yield predicted by the model and the experimental values at the root level. Furthermore, the introduction of a power n allowed not only to reduce the value of Ymax and km of the continuous extraction of root essential oil and discontinuous extraction of stem-leaf essential oil but, improves the monod model (Table 2). Both continuous extraction modes showed a good correlation with the experimental data.

The parameters $C_l(\infty)$ and of the Mafarat and Béliard II models were found to be low. Similar results on (Table 2) were observed by [27] for an extraction of essential oil of *Salvia Coccinea* by hydrodistillation. After the introduction of the power c in this Mafarat and Béliard II model leading to the pseudo-diffusional Mafarat and Béliard model, the values of the parameters $C_l(\infty)$, and were found to be higher than the previous values of $C_l(\infty)$, and c of the roots and stem-leaves illustrated in Table 2.

These results are comparable to the results obtained by [27]. However, the values of recorded were all much higher (0.08474) (Table 2) than the values of reported by him (0.00306), while the values of the power c, obtained were very high than the values reported by [27] which were around 0.431 min^{-1}.

On the other hand, the values of c are much higher than those obtained by [21] on the solvent extraction of Jatropha oil. The higher (or lower) values of $C_l(\infty)$, and C as reported by these authors imply respectively a rapid and higher (or lower) initial oil extraction rate and a slower final diffusion rate respectively for oil extraction from different parts of the plant studied.

The following Table 3 shows the statistical parameters of the different models applied to the extraction of EOs from different parts of the plant studied.

Table 3. Statistical parameters (roots and stem-leaves)

Part of the plant used (Extraction mode)	Models	Statistical parameters		
		R^2	Adj. R^2	χ^2
Roots (continuous extraction)	Mafarat and Béliard II	0,82745	0,80589	0,146
	Pseudo-diffusional Mafarat and Béliard	0,99491	0,99345	0,0049
	Monod	0,788	0,7619	0,1805
	Modified Monod	0,99501	0,9936	0,0048
Stem-leaves (continuous extraction)	Mafarat and Béliard II	0,94281	0,9356	0,0042
	Pseudo-diffusional Mafarat and Béliard	0,9922	0,990	$6{,}5.\mathbf{10^{-4}}$
	Monod	0,95021	0,93076	$6{,}054.\mathbf{10^{-4}}$
	Modified Monod	0,9928	0,9907	$6{,}054.\mathbf{10^{-4}}$
Stem-leaves (discontinuous extraction)	Mafarat and Béliard II	0,8388	0,812	0,3227
	Pseudo-diffusional Mafarat and Béliard	0,9726	0,9617	0,0656
	Monod	0,788	0,7619	0,1805
	Modified Monod	0,978	0,9694	0,0525

The results of R^2, Adj. R^2 and Chi-square ($\boldsymbol{\chi^2}$) were shown in Table 3.

Generally, higher values of R^2 and Adj. R^2 i.e. close to 1 and smaller or low values of would indicate a better quality of fit of the model to the experimental data.

According to this table, R^2 and Adj. R^2 increase with time while the value of decreases. It was also observed that these values were all less than or equal to 1. The Mafarat and Béliard II, Monod and power law models presented values of R^2 less than 1 and values of greater than 0. Only two models presented values of R^2 and Adj. R^2 close to 1 and the standard error, low. The values of R^2 obtained in this study were slightly higher than the values reported by [24] who reported values of 0.9584. [25] reports a value of 0.9883 for an oil extraction from *Irvingia gabonensis*. This indicates that these two models could better describe the kinetics of essential oil extraction from *Elionurus hensii* than the extraction of the different oils studied by these authors.

The Mafarat and Béliard pseudo-diffusional and modified Monod models had higher R^2 Adj. R^2 values and the standard error and lower and were chosen as the best kinetic models for the kinetic extraction of this essential oil whether continuous or batchwise.

3.4 Validation of models

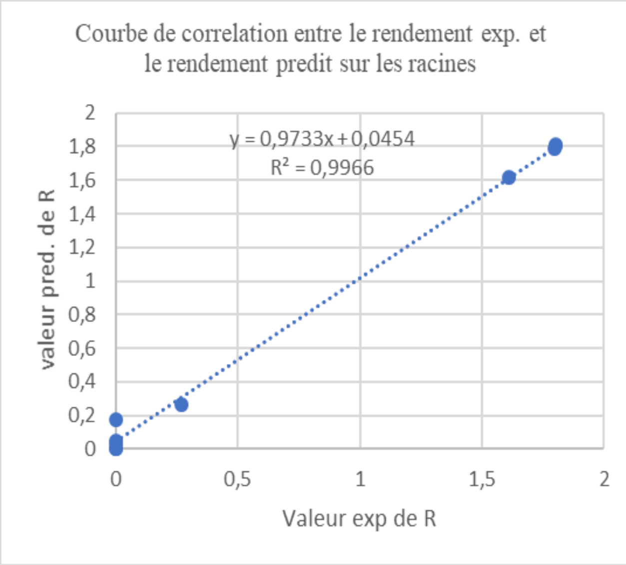

Figure 4a. Correlation curve between experimental yields and yields predicted by the Modified Monod model for the continuous extraction of essential oils from the roots of *Elionurus hensii*.

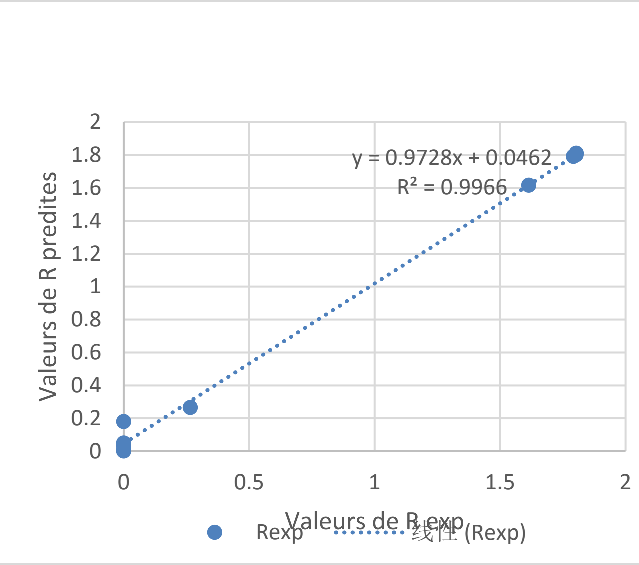

Figure 4b. Correlation curve between experimental yields and yields predicted by the Pseudo-diffusional Mafarat and Béliard model for the continuous extraction of essential oils from the roots of *Elionurus hensii*.

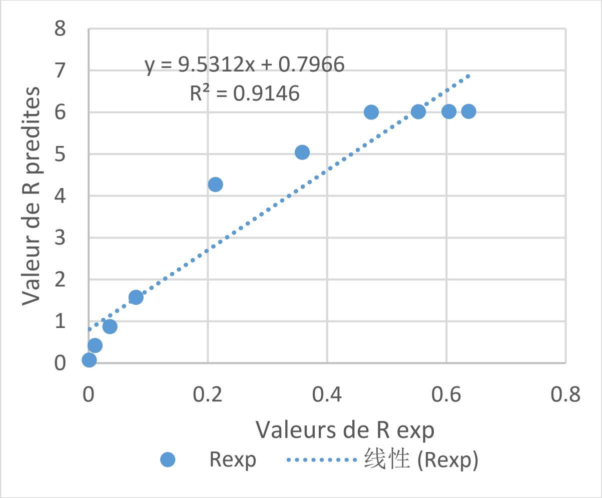

Figure 4c. Correlation curve between experimental yields and yields predicted by the Modified Monod model for the continuous extraction of essential oils from the stems-leaves of *Elionurus hensii*.

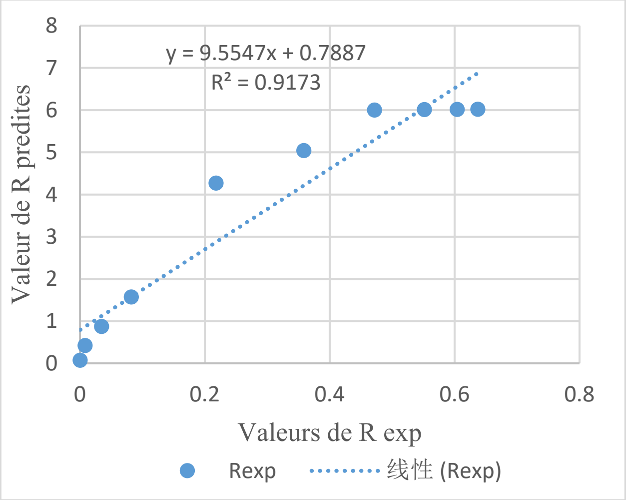

Figure 4d. Correlation curve between experimental yields and yields predicted by the Pseudo-diffusional Mafarat and Béliard model for the continuous extraction of essential oils from the stem-leaves of *Elionurus hensii*.

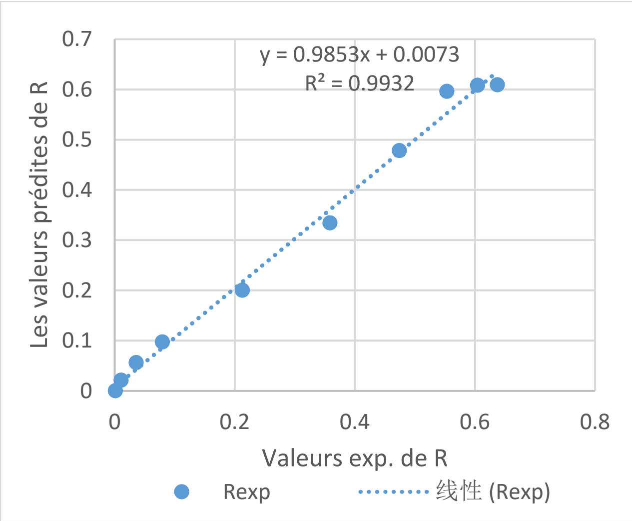

Figure 4e. Correlation curve between experimental yields and yields predicted by the Modified Monod model for the discontinuous extraction of essential oils from the stems-leaves of *Elionurus hensii*.

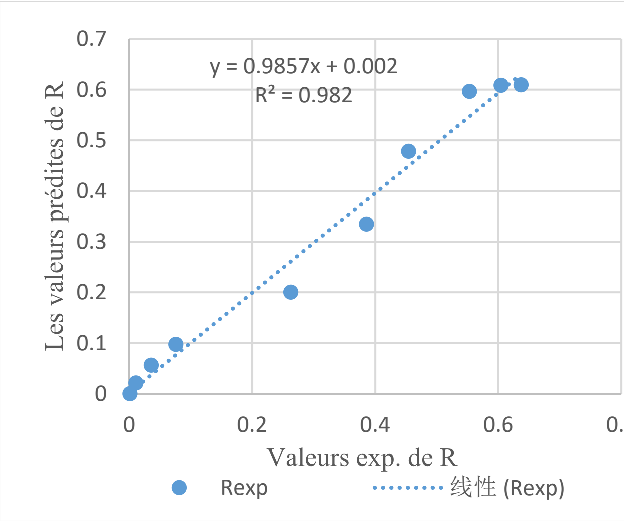

Figure 4f. Correlation curve between experimental yields and yields predicted by the Pseudo-diffusional Mafarat and Béliard model for the discontinuous extraction of essential oils from the stem-leaves of *Elionurus hensii*.

Figures 4 (a to f) show the fits of the modified Monod and pseudo-diffusional Mafarat Beliard models with experimental data for extractions (continuous and discontinuous) of essential oils from the roots and leafy stems of *Elionurus hensii*.

Looking at Figures 4a and 4b, we see that the experimental data appear to be closely scattered around the line representing the theoretical data from the modified Monod and pseudo-diffusional Mafarat and Beliard models. This indicates the relevance of these two models for describing the continuous extraction process of essential oils from the roots of *Elionurus hensii*. This is confirmed by the values of the regression coefficient close to 1 observed in these figures (R^2=0.9966). However, the R^2 values obtained for the continuous extractions of essential oils from the leafy stems of *Elionurus hensii* (0.9146; 0.9173) indicate a less perfect correlation of the experimental data with the data predicted by the two models.

From these results, we can conclude that the modified Monod and pseudo-diffusional Mafarat Beliard models better represent the process of continuous extraction of essential oils from the roots and discontinuous extraction of essential oils from the leafy stems of *Elionurus hensii*.

4. Conclusion

The objective of this study was to carry out an essential oil extraction and to establish representation models to describe the extraction kinetics (in batch and continuous mode) in the case of *Elionurus hensii*. The results of the extractions on the different parts of the plants gave yields of 2.7% and 0.6% for the roots in batch and continuous mode respectively and 1.81% for the roots. The essential oil obtained from the roots was solid. It also appears that a duration of 180 min gives a better yield for a complete distillation (total exhaustion of essential oil in the plant material). In other words, a duration (Optimal THD) of 180 min is sufficient to extract the maximum of essential oil contained in the plant material. The bibliographic models Mafarat and Béliard II, Monod, and the power law applied in this study have shown that they are not able to correctly represent the extraction kinetics from the plant *Elionurus hensii* by hydrodistillation over a wide time domain. However, with some modifications of the model Mafarat and Béliard II and Monod allow to have respectively the models Mafarat and Béliard pseudo-diffusional and Monod modified. These gave a good approximation of the data of the experiment. The high yields of oil obtained prove that this oil can be the subject of industrial production. Thus, the optimized extraction process and validated kinetic models provide a foundation for scaling up the production of E. hensii essential oils, contributing to sustainable agricultural and medicinal practices in Congo and beyond.

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