

**APPLICATION OF HERVAS AND PELEG KINETIC MODELS TO STUDY
MICROWAVE-ASSISTED EXTRACTION OF ANTIOXIDANT SECONDARY
METABOLITES FROM *TRICHILIA ROKA* ROOT BARK**

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ABSTRACT

Application of the Hervas and Peleg kinetic model was investigated for predicting kinetics of extraction of polyphenols, flavonoids and limonoids from *T.roka* root bark through microwave assisted process under four independent variables (variable irradiation time X1, and fixed irradiation power X2, liquid-to-solid ratio X3 and methanol concentration X4). The maximum extraction efficiency was achieved with microwave irradiation power: 400 W, liquid-to-solid ratio: 1/20 mL/g and methanol concentration 95%. From Hervas model, the concentration of total limonoid (TL) and total polyphenol (TPP) release at equilibrium C_0 were respectively 425 and 542010 $\mu\text{gE/gDW}$ with a diffusion coefficient k of 0.02 and 0.06 s^{-1} . According to Peleg's model, the maximum extraction quantities (a) of TF, TL and TPP from *T. roka* root bark (C_0) are 500.00, 50.00, and 833333.33 $\mu\text{gE/gDW}$ respectively and with the respective extraction velocity coefficients (k) of 0.014, 0.03; and 0.04 s^{-1} . These parameters were obtained at the maximum irradiation time of 60 and 80 s respectively for limonoids and phenolic compounds with the extraction kinetic constants of 40 and 45 s^{-1} . The Hervas model best adjust the extraction of TL and TPP, Peleg model best adjusts the extraction of the three secondary metabolites. The correlation analysis of the mathematical regression model indicated that a first and second-order polynomial model could be employed to obtain the highest recovery of secondary metabolites. The closeness of experimental values, with the predicted ones indicating the good similarity of the models used.

KEYWORDS: *Trichilia roka*, Microwave assisted extraction, antioxidant activity, DPPH radical scavenging activity, β -carotene bleaching test.

INTRODUCTION

Liquid-solid extraction is a process of transferring one or more solutes from a solid to an adjacent fluid that corresponds to the extraction solvent. Kinetic data for the extraction of secondary metabolites from plants in general and from *Trichilia roka* in particular are very scarce because the implementation of the extraction process requires the resolution of delicate problems such as modelling and the determination of certain parameters that are often non-existent in the data banks (Herzi et al., 2013). The implementation of a mathematical model of the kinetics of extraction of secondary metabolites could on one hand allow the understanding of the phenology of the extraction process and on the other hand minimize the number of experiments to be carried out, while ensuring the quality of the extract in terms of bioactive compounds (Cassel et al., 2009).

There are several methods that can be used for such a purpose namely maceration, reflux extraction, Soxhlet extraction and supercritical fluid extraction called conventional method. However, these methods are either time, energy and solvent-consuming or too expensive for small-scale implementation (Chumnanpaisont et al., 2014). Therefore, between alternative methods of extraction, microwave-assisted extraction (MAE) has been proposed to solve these problems. This method is extensively used in the extraction of bioactive secondary metabolites from plant materials due to its high efficiency in terms of its extraction conditions such as good extraction yields, less solvent, energy consumptions, extraction time and easy implementation (Chumnanpaisont et al., 2014). In addition, during MAE heat diffuses in the matrix from the inside to the outside, consequently it improves the extraction and solubilisation of the molecules. Indeed, MAE has shown its efficiency in improving the yield of extraction of limonoids from *Trichilia roka* (Nana et al., 2021). While the maximum yield of extraction is the aim during extraction of molecules, the kinetic of extraction is a prerequisite to achieve that goal. Indeed, irradiation time is the most valuable parameter as the speed of extraction is the main advantage in microwave extraction (Prasad et al., 2011; Liu et al., 2010; Farhat, 2010). In order to improve the use of *Trichilia roka* extract, it appears important to study the kinetic extraction of the bioactive compounds. Improved extraction yield of metabolites is a function of extraction method in the first hand and how fast the secondary metabolites is dissolved and the equilibrium in the liquid is reached in the second hand. When solute diffusivity in vegetal matrix is considered, the rate limiting step of extraction is the diffusion of dissolved solute from the solid to the solvent (Gertenbach, 2002). The extraction rate therefore increases with a greater concentration gradient. Recovery of compounds from plant materials is typically accomplished through MAE techniques taking into account their chemistry and uneven distribution in the plant matrix. The result of extraction is a function of how fast the secondary metabolites are dissolved and the equilibrium in the liquid is reached (Herzi et al., 2013).

Among bioactive compounds are limonoids and polyphenols which exhibit a wide variety of biological properties including antioxidant and antiplasmodial activities. In addition, they exhibit many biological activities such as insecticidal, antifungal, antibacterial, antiviral, and anticancer effects (Braga et al., 2020). *Trichilia roka* (Chiov) (Meliaceae) has been used in traditional medicine long time ago in many tropical and sub-tropical countries, thanks to their content in limonoids, and phenolics. Our recent findings reported several isolated compounds from several genera of Meliaceae with antimalarial, antimicrobial, anti-inflammatory, antischistosomal, anticonvulsant, anticancer, antitrypanosomal and antimutagenic activities (Nana et al., 2013). To the limit of our knowledge, no study has yet been reported on the kinetics of microwave-assisted extraction of these secondary metabolites. The importance of studying extraction kinetic is not only for modelling the process, but also for optimisation purposes. The optimisation will go a long way improving the extraction yields and the biological activities.

Many kinetic models have been proposed by many authors to follow the extraction of metabolite. The typical kinetic models of liquid-solid extractions include unsteady state diffusion empirical equation of Hervas, Peleg model (Peleg, 1988) and Fick's law of diffusion (Cacace and Mazza, 2003). Mathematical models are useful engineering tools which facilitate the simulation, optimization, design and control of extraction processes. In fact, mathematical modelling can simplify the process design and control to obtain the optimization condition and provide correct information for large-scale extraction and preparation (Yonggang *et al.*, 2021). Besides the optimum condition, the optimization will provide mathematical models able to properly predict the behaviour of the system considering the factors that influence the MAE process.

In this study, research on kinetic modelling of microwave extraction of limonoids and polyphenols using the Hervas and Peleg kinetics models was undertaken to provide a comprehensive follow up of the leaching of the metabolites.

2. MATERIALS AND METHODS

2.1. Plant materials and chemicals

The root bark of *Trichilia roka* was harvested in December 2020, from a forest in a suburb around Ngaoundere (Adamawa Region of Cameroon). The plant sample was taxonomically authenticated by Pr Mapontmesem, a botanist in the Department of Biological Sciences, Faculty of Science of the University of Ngaoundere. The plant material was freed of extraneous material, shade-dried at room temperature for two weeks and milled to a fine powder, using a waring blender. Two kilograms of powder was packaged in an air tight container, labelled and stored until used.

Chemicals used included Folin–Ciocalteu reagent, 2,2-diphenyl-1-picrylhydrazyl (DPPH), gallic acid, sodium carbonate, potassium acetate, potassium hexacyanoferrate, iron (III) chloride, β -carotene, linoleic acid, chloroform, and tween 40, obtained from the Sigma Chemical Co. (St. Louis, MO, USA). Metertech Spectrophotometer UV/vis sp 8001, rotary evaporator system (Büchi, Switzerland) and a modified microwave oven (DAE WOO, KOG- 360 and Combi Grill) were also used.

2.2. Microwave-Assisted Extraction (MAE)

Microwave-assisted extraction is one of the "green extraction" methods that are highly effective in the recovery of polyphenols (Panja, 2018). We have adapted a domestic microwave for the extraction of limonoids, polyphenols and flavonoids as shown in Fig 1. For the extraction operation, 1 g of dried powder of vegetal material was suspended in 20 mL of aqueous methanolic solution (20:80) (v/v) in a 150 mL Teflon extraction vessel. The vessel was placed at the centre of the microwave apparatus and heated at different time (0, 20, 30, 40, 50, 60, and 80 s) at 400 W. after the extraction, the vessel was allowed to cool down (1 min at 25°C), and the mixture filtered on Whatman filter paper N° 1 and funnel. Then the filtrate was kept at 4°C for total polyphenol, total limonoids, total flavonoids content, antiradical and antioxidant investigations.

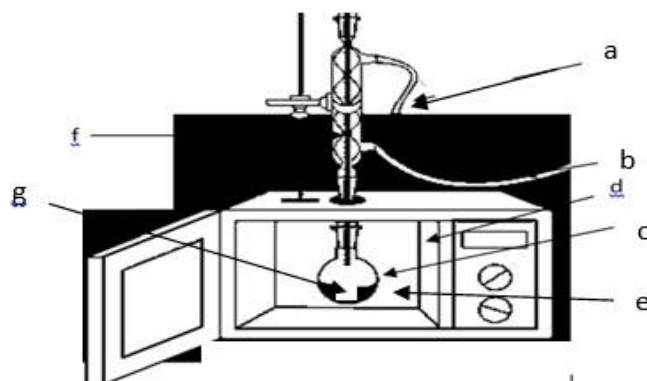


Fig. 1: Domestic microwave oven (Daewoo KOG-360 combi grill) adapted with a condenser.

a: outlet water, b: cold water inlet, c: boiling flask, d: microwave cavity, e: microwave zone, f: condenser tube, g: sample.

Analysis of chemical and antioxidant activities

The total phenolic content of extract was spectrometrically analyzed in adherence to the Folin-Ciocalteu method (Cheng *et al.*, 2009) and the results expressed as μg Gallic Acid Equivalents (EAG) per g dry matter. Total flavonoids were brought up by using the method previously implemented by Zhishen *et al.* (1999) and expressed as μg Quercetin Equivalents/g dry matter. Total limonoids content, expressed as μg Rubenscin Equivalents (ERUB)/g dry matter, was carried out by colorimetric method as previously described (Abbasis *et al.*, 2005). The radical scavenging property using 2, 2-diphenyl-1-picrylhydrazyl (DPPH) radical was determined according to Mansouri (2005). The antioxidant property of the extract on β -carotene-linoleic acid system was determined by measuring the inhibition of β -carotene degradation by the oxidation products of linoleic acid according to the method described by Kartal *et al.* (2007).

2.3. Mathematical modelling of kinetic extraction of phenolics and limonoids

Two empirical models were employed because of their relative ease of use (Tuhran *et al.*, 2002a, b): the Hervas and Peleg empirical models.

2.3.1. Hervas model

Hervas model is based on the assumptions that the limiting step in the extraction is the diffusion process and not that of solubilization, the solute partition coefficient between the two phases is equal to unity, the mass loss of the solid phase subsequent to the solute diffusion is compensated by an equivalent mass gain of solvent migrating into the solid phase.

The kinetic mechanism proposed by Hervas *et al.* (2006) followed the equation:

$$C(t) = C_0 (1 - e^{-kt}) \quad (1)$$

Where C and C_0 ($\mu\text{g E/gDW}$) are the concentration of metabolite in the medium (solvent) at respectively extraction time t (s) and equilibrium, and k (s^{-1}) is the first order kinetic constant of extraction.

Empirical model of Peleg

The empirical model of Peleg is generally used to describe sorption kinetics. He proposed a well-known hyperbolic model describing moisture sorption curves (Peleg, 1988). Since there is a similarity between the shape of the extraction and the sorption curves, Peleg's model has been adapted and used to describe the solid-liquid extraction of various metabolites from plant materials such as oregano, coffee silverskin, jamun (*Syzygium cumini*) seeds, forestry lignocellulosic by-products, brown seaweed (*Ascophyllum nodosum*) (Milićević *et al.*, 2021). The Peleg model estimates

the theoretical (C_e) extraction yields as a function of time according to the equation as well as the extraction rate in the first minutes (C_0). The modified Peleg's equation is expressed as follows,

$$C(t) = C_0 + \frac{t}{K_1 + K_2 t} \quad (2)$$

where $C(t)$ is the yield of solutes (g / 100 g) at time t , C_0 is the yield of solutes (g / 100 g) at time 0, K_1 is related to the constant rate of extraction of solutes expressed in min.g/100 g, K_2 provides information on the equilibrium extraction yield of solutes ($C_\infty = 1/K_2$, g/100 g). In the present work, $C_0 = 0$ at time $t = 0$ then:

$$C(t) = \frac{t}{K_1 + K_2 t} \quad (3)$$

Considering the equilibrium condition, it emerges from these equation three kinetic parameters namely:

The initial extraction rate B_0 ($\mu\text{g E/ (s.gDW)}$), the phenolic compounds extraction capacity of the Peleg model in equilibrium (C_0) ($\mu\text{g E/gDW}$), and the coefficient of extraction speed k (s^{-1}); defined in equation:

$$C_0 = \frac{1}{K_2}; k = \frac{K_2}{K_1}; B_0 = \frac{1}{K_1}$$

2.4. Statistical analysis

The results reported in this work are the average values of three measurements. Analysis of variance (ANOVA) was used to determine the influence of each factor and the degree of significance of each of these effects. It then examines the statistical significance of each effect by comparing the squared average against an evaluation of the experimental error. The significance of each factor is determined by the Fisher test, which is defined as the ratio of the mean square of the regression (MSR) to the experimental error (EE) ($F = \text{MSR}/\text{EE}$), representation of the meaning of each variable controlled on the model examined. The regression equations were also subjected to the Fisher test to determine the regression coefficient R^2 . The adequacy of the model with respect to the experimental measurements can be expressed using a determination coefficient R^2 (the more closely R^2 value is close to 1, the more suitable the model). The agreement between the experimental data and the models were assessed by using Absolute Mean Deviation Analysis (AMDA) and the adjusted correlation coefficient (R^2 adj). A low AMDA and a high R^2 adjusted value denote good fit

$$AMDA = \frac{\sum_{i=1}^p \left(\frac{|Y_{iexp} - Y_{ical}|}{Y_{iexp}} \right)}{p} \quad (7)$$

With: Y_{iexp} the experimental response and Y_{ical} the calculated response from the model for an experiment i ; p being the total number of experiments.

3. RESULTS AND DISCUSSION

3.1. Extraction kinetics

The detailed experimentation on four independent variables as mentioned in the abstract have been studied before applying the kinetic models (Nana *et al* 2021). Figure 1 shows the variation in metabolites (flavonoids, limonoids and polyphenols) extraction as a function of time. For these metabolites, we observe increasing curve up to 60 s for limonoids and 80 s for phenolic compounds. After this growth, follows a decrease which seems to stabilize for limonoids and rise for phenolic compounds. It happens that, the rapid temperature rise and the internal pressure increase

generated by the microwave radiation resulted in the release of target compounds. For the kinetics of extraction of total flavonoids (TF) and total polyphenols (TPP), the maximum values are (402 $\mu\text{g QE/g DW}$ for TF and 533421 $\mu\text{g GAE/gDW}$ for TPP) obtained at 80 s, after which a phenomenon of degradation of the molecules is observed thus lowering the various contents. On the other hand, at 60 s the total limonoids (TL) reached the maximum of extraction (258.387 $\mu\text{g RubE/gDW}$) and immediately the degradation took place. Limonoids therefore being heat-sensitive molecules compared to phenolic compounds. The shape of the curves is explained by the fact that an observable equilibrium does not have time to establish itself as in conventional extraction method. With microwave heating, at time 60s for the limonoids and 80s for the phenolic compounds, these needing metabolites are already extracted and, because of the higher temperature a phenomenon similar to a precipitation, or complexation of metabolites is observed, such as flocculation phenomenon. After these times increasing the extraction time up to 120 s does not significantly ($p > 0.05$) improve the extraction of phenolic and limonoids compounds. This observation could be explained by Fick's second law on diffusion stipulating that the equilibrium would be reached between the concentration of the solute (active substances) of the solid matrix (plant matrix) and the solution (solvent) after a certain time.

$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2}$ where D is the diffusion coefficient, a constant. Therefore, excessive extraction time will not be necessary to extract secondary metabolites such as phenolic and limonoids compounds (Chan *et al.*, 2009).

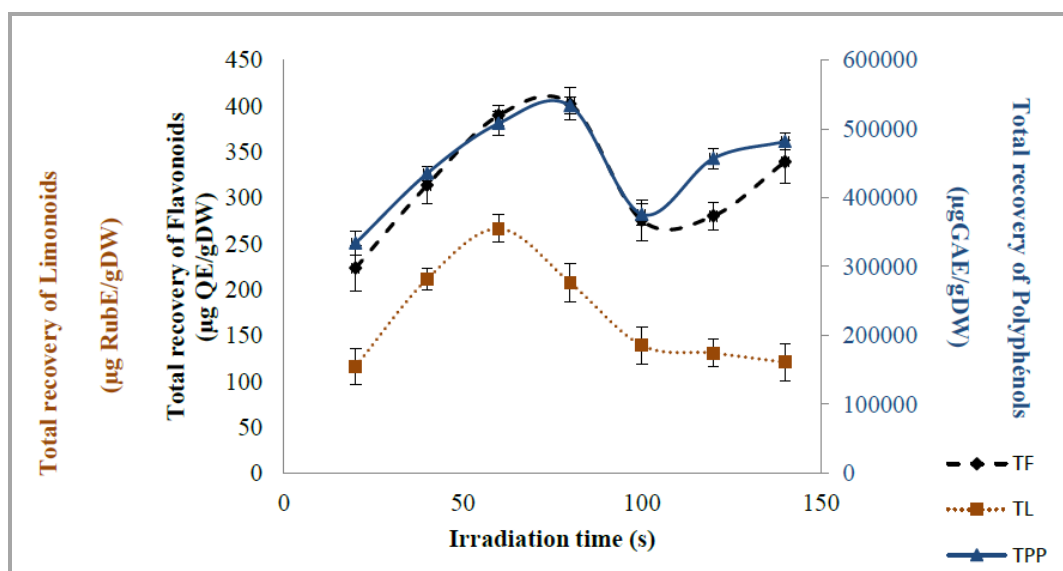


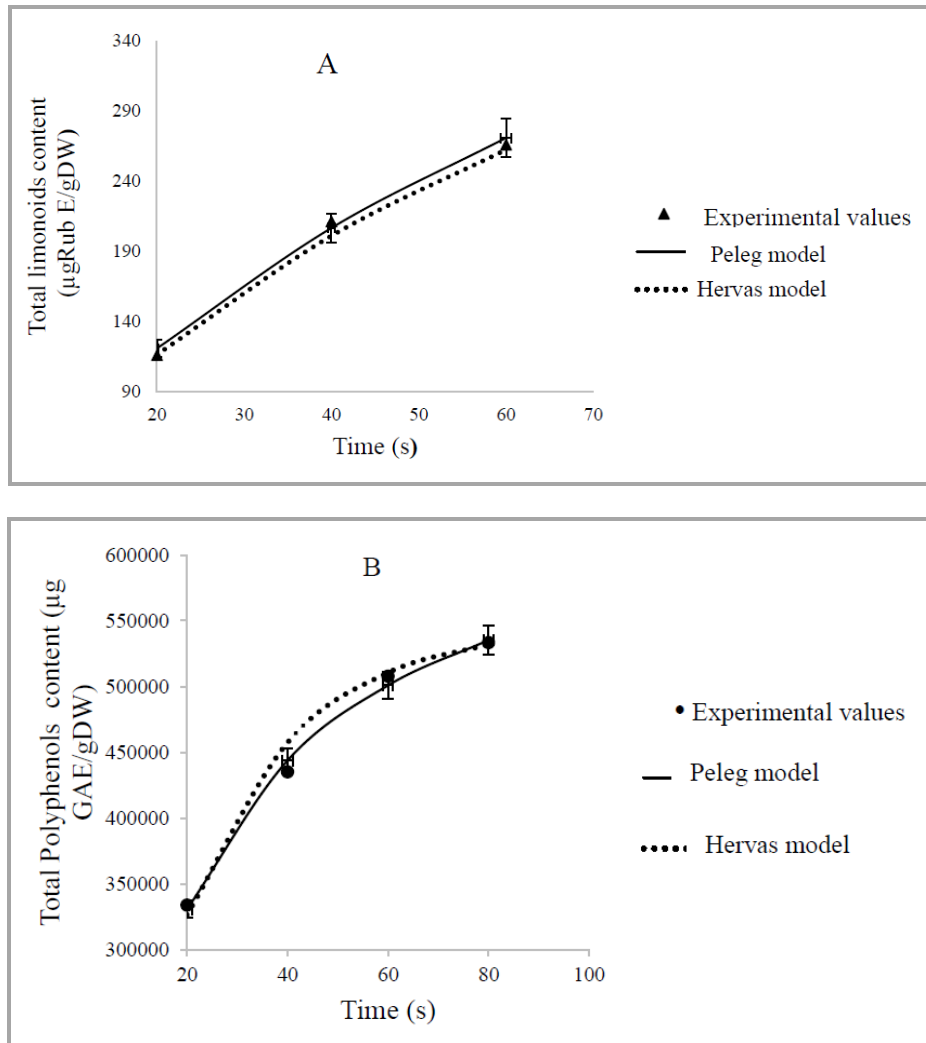
Fig. 1: Effect of irradiation time on the content of total flavonoids, total limonoids, and total polyphenol in *Trichilia roka* microwave-assisted extraction of root bark.

3.2. Secondary metabolites extraction kinetic models

In order to determine the effective diffusion coefficient, assuming no change in the effective diffusivity (proportionality constant between the molar flux due to molecular diffusion and the concentration gradient) with solute concentration, the rate of diffusion of the limiting step can be described by Fick's second law knowing that at the second extraction stage, the internal diffusion, which is slower than the external elution, is the limiting process. In addition, our study will be limited to the diffusion phase, because the fast leaching phase is characterized by a much lower solubilization constant than the extraction yield at equilibrium during the diffusion phase (Franco *et al.*, 2007).

3.2.1 Hervas and Peleg kinetics model

The two models described above were employed to fit the experimental data. Indeed for the model fit curves, the figures (2, A, B, and C) below indicate that there is a progressive increase in total flavonoids, total limonoids and total polyphenols contents over time, corresponding to the diffusion of those secondary metabolites from the plant material to the solvent following leaching and diffusion phenomenon. But, Hervas kinetic model can't be used for MAE of total flavonoids (fig. 2 C) despite the R^2 value of 0.96. Otherwise, microwave assisted extraction efficiency of those metabolites increases until an optimum level is reached. Except for the extraction of total flavonoids, all curves agreed well with the two models; the different R^2 values were ≥ 0.96 . The appropriateness of the two kinetic equation in fitting the MAE of limonoids and polyphenols is attributed to the capacity of the equation to explain why the initial state of extraction is rapid and be very slow after the equilibrium state. According to this statement, it can be clearly seen that, The R^2 values (0.97–0.99) for Peleg kinetic model and (0.96–0.99) for Hervas kinetic model are similar, but a slight difference is observed for total flavonoids. Peleg kinetic model best fits the MAE of flavonoids with an R^2 value of 0.97 than that of Hervas with an R^2 value of 0.96, more the AMDA value of Peleg model is significantly smaller (0.04) than that of Hervas value (0.06). Hence the following adjusted curves.



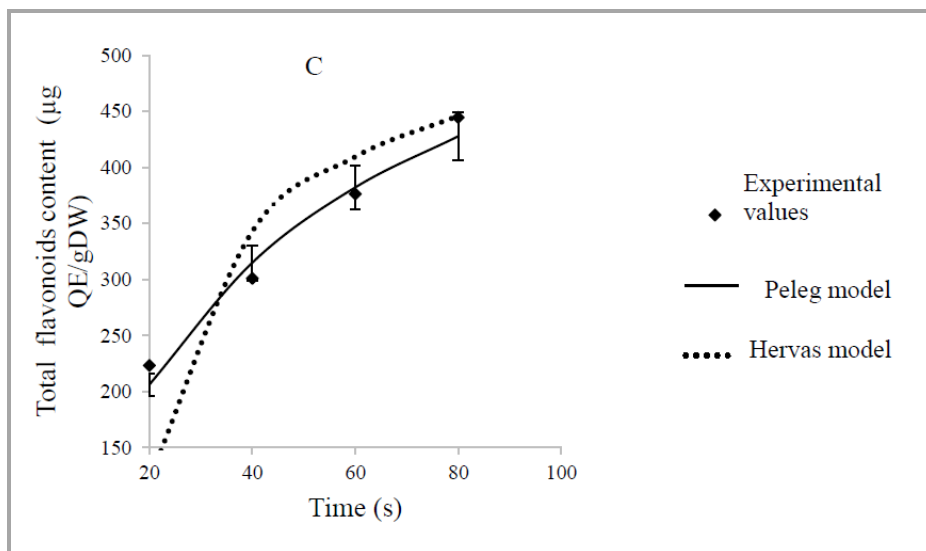


Fig. 2: Effect of extraction time on the MAE for diffusion step; A: for total limonoids; B: for total polyphenols; and C: for total flavonoids.

3.2.2. Hervas and Peleg kinetic models parameters

The two kinetic models equations were generated by fitting the experimental data of microwaveassisted extraction of the three secondary metabolites from *Trichilia roka* root bark. According to the two models, the concentrations of TF, TL and TPP released at equilibrium (Co) are 490, 425 and 542010 µgE/gDW respectively and the respective diffusion constants (k) are 0.03 and 0.02; and 0.06 s⁻¹ for Hervas kinetic model. The appropriateness of the Hervas kinetic equation in fitting the MAE of secondary metabolites is attributed to the capacity of the equation to explain why the initial state of extraction is rapid and very slow after the equilibrium state. In the same vein, the concentrations of TF, TL and TPP released at equilibrium (Co) are 500.00, 50.00 and 833333.33 µgE/gDW respectively and the respective extraction velocity coefficients (k) are 0.03; 0.014 and 0.04 s⁻¹

Table 1: Related parameters of the Hervas model for MAE of different responses.

Responses	Co (µg E/gDW)	k(s ⁻¹)	R ²	AMDA
TF	490	0.03	0.96	0.06
TL	425	0.02	0.99	0.02
TPP	542010	0.06	0.98	0.02

Co (µgE/gMS): the concentration of metabolites at the equilibrium K(s⁻¹): the kinetic constant of extraction

AMDA: Absolute Means Deviation Analysis

The solid–liquid extraction process can be considered as the reverse of an adsorption operation, therefore the bases of the adsorption kinetic equations can be applied to solid–liquid extraction and the Peleg kinetic model can be applied to experimentally evaluate the extraction rate constant such as Peleg rate constant (K1), Peleg capacity constant (K2) and others. The fitting curve and four kinetic parameters (K1, K2, Bo, Co) were obtained, during the diffusion phase and for the three secondary metabolites. This diffusivity value describes how fast these metabolites diffuse through microwave extraction solvent. From Peleg's kinetic model, the concentrations of TF, TL and TPP released at equilibrium (Co) are obtained and the extraction velocity coefficients (Table 2). In addition, the initial extraction rates Bo of TF, TL and TPP obtained are 14.12, 0.88 and 32467.53 µg E/ (s.gDW). The higher value of these secondary metabolites obtained is related to the nature of the plant matrix and also to the nature of the extraction solvent. K1 is a

constant related to mass transfer rate, e.g., the lower the K_1 , the higher the initial solid-liquid extraction rate and K_2 is a constant related to maximum solid-liquid extraction capacity, i.e., the lower the K_2 , the higher the solid-liquid extraction capacity. K_1 ($3.6 \cdot 10^{-4}$ gMs/ μ gE) and K_2 ($4.7 \cdot 10^{-4}$ gMs/ μ gE) values of this work are in close agreement with those of Milićević (2021) in their work on application of Peleg model to study Kinetic modelling of ultrasound-assisted extraction of phenolics from cereal brans were K_1 and K_2 were respectively K_1 ($2 \cdot 10^{-4}$ gMs/ μ gE) and K_2 ($8 \cdot 10^{-4}$ gMs/ μ gE) for control reagent, and Tuhiran and *al* (2002) on their work on application of Peleg model to study water absorption in chickpea during soaking were K_1 and K_2 were respectively $17.1 \cdot 10^{-3}$ s gMs/ μ gE) and ($4.26 \cdot 10^{-3}$ gMs/ μ gE). But we can noticed that when the Peleg model was applied to the experimental data of MAE extraction (Figures 2 A, B and C), the overall fitting of the experimental data was good, but the applicability of the whole extraction process was poor, especially for the second stage of extraction. It cannot well describe the behaviour of MAE process during the entire extraction period. It is necessary to explore more extraction models (Yonggang *et al.*, 2021).

Table 2: Related parameters of the Peleg model of different responses.

Responses	K_1 (s.gDW/ μ gE)	K_2 (gDW/ μ gE)	Bo (μ gE/s.gDW)	Co (μ gE/gDW)	k (s^{-1})	R^2	AADM
TF	0.067	0.002	14.92	500	0.030	0.97	0.04
TL	0.138	0.02	0.88	50	0.014	0.99	0.03
TPP	$3.1 \cdot 10^{-5}$	$1.2 \cdot 10^{-6}$	32467.53	833333.33	0.038	0.99	0.01

K_1 (s.gMS/ μ gE): the Peleg rate constant of solute; K_2 (gMS/ μ gE): the Peleg capacity constant of solute; Bo (μ gE/s.gMS): the initial extraction rate of solute; Co (μ gE/gMS): The equilibrium extraction yield; K (s^{-1}): the kinetic constant of extraction; AMDA: Absolute Means Deviation Analysis

3.3. Antioxidant activities

It was successfully shown that samples with high level of phenolic content also contain flavonoids in great amount. The rich-flavonoid plants could be a good antioxidant source that would help increase the overall antioxidant capacity of an organism and guard it against lipid peroxidation (Shariifar *et al.*, 2009). Concerning limonoids, limonin, the aglycone and glycosides of *Citrus* limonoids have shown to display numerous pharmacological activities including anticancer, antimicrobial, antioxidant, antidiabetic and insecticidal among others. Compared to the two other secondary metabolites limonoid structures are less indicated to provide direct radical-scavenging activity as they lack aromatic and phenolic structures. Therefore, Limonin (Lim) and Limonin-17- β -D-glucopyranoside (LG) were investigated using the two antioxidant method used in this work, it is found that Lim and LG inhibited less than 7% using β -carotene-linoleate model system and showed 0.5% and 0.25% respectively free radical scavenging activity using the DPPH method (Jun yu *et al.*, 2005). Nevertheless, Hamdan *et al.* (2011) evaluated the antioxidant activity of different extracts from the fresh peel of *Citrus jambhiri* by measuring their DPPH radical-scavenging effects and found that the chloroform fraction exerted the strongest DPPH free radical scavenging activity with IC50 value of 119.4 ± 0.8 M (Guldani *et al.*, 2016).

According to antioxidant activity, the R^2 value of the Hervas and Peleg kinetic model for the DPPH scavenging assay was determined to be 0.934 and 0.982 for β -carotene assay only 6.6 % and 1.8% of that total variations were not explained by the model (Nana *et al.*, 2021).

The β -carotene assay showed a high degree of precision of the reliability of the experimental values compare to the DPPH scavenging activity. In addition, the results for antioxidant assay respectively the DPPH scavenging assay and β -

carotene assay were promising (90.12 and 90.49%) and demonstrated in a comparative manner that, more than the practical aspect of MAE to substitute the conventional extraction method is recommended. An appropriate kinetic model provides a best antioxidant efficiency for polyphenolic and limonoids compounds from *T. roka* root bark MAE extract.

4. CONCLUSION

The results showed that Hervas and Peleg's model fits adequately the MAE of limonoids and phenolic compounds extract from *T. roka*. The experimental results, different from those used in the two kinetic models, were compared with the predicted values in order to confirm the accuracy of the kinetic model. For the whole MAE process effective diffusivity values were determined for each response. Food supplements and drugs based on limonoids and phenolic compounds extract from *T. roka*, under these conditions and according to these models will be endowed with optimal biological activities such as anticancer, antiplasmodial, antioxidant, antiinflammatory and much more. Any model will be valid if manipulated under exactly the same conditions, the square of the correlation coefficient R^2 tends towards 1 and to confirm, the values of the Absolute Mean Deviation Analysis (AMDA) of the different models are between 0 and 0.2. (Karam, 2004). Hence Hervas and Peleg kinetic models best adjust these secondary metabolites microwave assisted extraction. According to the diffusion step, Hervas models best adjust the extraction of limonoids and polyphenol and Peleg models best adjust the extraction of the three studied secondary metabolites. In addition to the advantages linked to the microwave-assisted extraction mentioned above, the knowledge of a good kinetic model allowing the transfer of the matter from the vegetal or animal matrix is necessary in order to optimize the extraction technique and thus maximize the biological activities through the active principle.

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Author's contribution

All of us made substantial contributions for this manuscript writing. ON has performed all the manipulations of this work, interpreted the results, and wrote the entire manuscript JM structured, supervised the drafting of this manuscript, and reviewed it entirely. NYN made analysis of some data and reviewed it entirely, TRC drew some figure of this work and MBN provided the necessary laboratory equipment for this work.

REFERENCES

1. N. Herzi, S. Camy, J. Bouajila, P. Destrac, M. Romdhane, J.S. Condoret, Supercritical CO₂ extraction of *Tetraclinis articulata*: chemical composition, antioxidant activity and mathematical modeling. *Journal of Superfluid*, 2013; 82: 72-82. <http://dx.doi.org/10.1016/j.supflu.2013.06.007>
2. E. Cassel, R.M.F. Vargas, N. Martinez, D. Lorenzo, E. Dellacassa, Steam distillation modeling for essential oil extraction process. *Industrial Crops and Products*, 2009; 29: 171-176. DOI:10.1016/j.indcrop.2008.04.017
3. N. Chumnanpaisont, C. Niamnuy, S. Devahastin, Mathematical model for continuous and intermittent microwave-

- assisted extraction of bioactive compound from plant material: Extraction of β -carotene from carrot peels. *Chemical. Engineering Science*, 2014; 116: 442–451. DOI:10.1016/j.ces.2014.05.010
4. O. Nana, J. Momeni, F.F. Boyom, M.B. Ngassoum, Microwave assisted extraction of antiplasmodial and antioxidant limonoids from *Trichilia roka* (chiov). *Journal of Phytopharmacology*, 2021; 10(3): 185-191. [https://doi.org/10.31254/phyto.\(2021\)10307](https://doi.org/10.31254/phyto.(2021)10307)
 5. T.M. Braga, L. Rocha, T.Y. Chung, R.F. Oliveira, C. Pinho, A.I. Oliveira, J. Morgado. A. Cruz, Biological Activities of Gedunin-A Limonoid from the Meliaceae Family. *Molecules*, 2020; 25(493): 1-24. <https://dx.doi.org/10.3390/molecules25030493>.
 6. K.N. Prasad, .A.H. Fouad, Y. Bao, W.K. Kin, N.R. Ramakrishnan, A. Azrina. A. Ismail, Response surface optimization for the extraction of phenolic compounds and antioxidant capacities of underutilised *Mangifera pajang* Kosterm. Peels. *Food Chemistry*, 2011; 128: 1121–1127. DOI 10.1016/j.foodchem.2011.03.105
 7. W. Liu, Y. Yanying, Y. Ruzhen, W. Chunpeng, X. Binbin, and C. Shuwen, Optimization of Total Flavonoid Compound Extraction from *Gynura medica* Leaf Using Response Surface Methodology and Chemical Composition Analysis. *International Journal of Molecular Sciences*, 2010; 11: 4750-4763. <https://doi.org/10.3390/ijms11114750>
 8. A. Fahrat, Vapo-Diffusion assistée par Micro-ondes: Conception. Optimisation et application. Thèse de Doctorat, Université d'Avignon et des Pays de Vaucluse & L'Ecole Nationale d'Ingénieurs de Gabès. 2010; 3-12.
 9. O. Nana, J. Momeni, F.F. Boyom, N.Y. Njitang and M.B. Ngassoum. Microwave assisted extraction as an advanced technique for optimisation of limonoids yields and antioxidant potential from *Trichilia roka* (Meliaceae). *Current Research in Green and Sustainable Chemistry*, 2021; 4: 100147. <https://doi.org/10.1016/j.crgsc.2021.100147>
 10. D.D. Gertenbach, Solid–liquid Extraction Technologies for Manufacturing Nutraceuticals. In: J.Shi, G. Mazza, and M.L. Maguer. (Eds.). *Functional Foods: Biochemical and Processing Aspects*. CRC Press Inc. Boca Raton. FL. 2002; 332–365.
 11. W.Yonggang, W. Chenliang, X. Hongyan, J. Yongming, Y. Mingjun Y, L. Feifan, Comparative analysis of three kinds of extraction kinetic models of crude polysaccharides from *Codonopsis pilosula* and evaluate the characteristics of crude polysaccharides. *Biomass conversion and Biorefinery*, (2021) <https://doi.org/10.1007/s13399-022-02518-w>.
 12. M. Peleg, An empirical model for the description of moisture sorption curves. *Journal of Food Science*, 1988; 53: 1216–1219. <https://doi.org/10.1111/j.1365-2621.1988.tb13565.x>
 13. J.E. Cacace, G. Mazza, Mass transfer process during extraction of phenolic compounds from milled berries. *Journal of Food Engineering*, 2003; 59: 379–389. [https://doi.org/10.1016/S0260-8774\(02\)00497-1](https://doi.org/10.1016/S0260-8774(02)00497-1)
 14. S. Karam, Application de la méthodologie des plans d'expériences et de l'analyse de données à l'optimisation des processus de dépôt, Thèse de Doctorat, Université de Limoges, 2004; 8-12: 110-120.
 15. P. Panja, Green extraction methods of food polyphenols from vegetable materials. *Current Opinion in Food Science*, 2013; 23: 173. <https://doi.org/10.1016/j.cofs.2017.11.012>.
 16. S.M. Cheng, M. Luther, W. Kenworthy, & L. Yu, Antioxidant properties and phenolic isoflavone, tocopherol and carotenoid of maryland-grown soybean lines with altered fatty acid profiles. *Food Chemistry*, 2009; 114: 20–27. <https://doi.org/10.1016/j.foodchem.2008.09.007>
 17. J. Zhishen, T. Mengcheng, and W. Jianming, The determination of flavonoid contents in mulberry and their

- scavenging effects on superoxide radicals. *Food Chemistry*, 1999; 64 (4):555–559. [https://doi.org/10.1016/S0308-8146\(98\)00102-2](https://doi.org/10.1016/S0308-8146(98)00102-2).
18. Z. Abbasis, P. Zandi, E. Mirbagheri, Quantitation of limonin in Iranian orange juiceconcentrates using high-performance liquid chromatography and spectrophotometric methods. *European Food Research Technology*, 2005; 221: 202-207. DOI:10.1007/s00217-005-1136-1.
 19. A. Mansouri, G. Embarek, E. Kokkalou, P. Kefalas, Phenolic profile and antioxidant activity of the Algerian ripe date palm fruit (*Phoenix dactylifera*). *Food Chemistry*, 2005; 89: 411-420. DOI: 10.1016/j.foodchem.2004.02.051.
 20. C. Sanchez-Moreno, Methods used to evaluate the free radical scavenging activity in foods and biological systems. *International Journal of Food Science and Technology*, 2002; 8: 121-137. DOI: 10.1177/1082013202008003770.
 21. N. Kartal, M. Sokmen, B. Tepe, D. Dafetera, M. Poulissou, A. Sokmen, Investigation of the antioxidant properties of *Ferula orientalis* L using a suitable extraction procedure. *Food Chemistry*, 2007; 100: 584–9 <https://dx.doi.org/10.1016/j.foodchem.2005.09.084>.
 22. M. Turhan, S. Sayar, S. Gunasekaran, Application of Peleg model to study water absorption in chickpea during soaking. *Journal of Food Engineering*, 2002; 53(2): 153–159 Doi: 10.1016/S0260-8774(01)00152-2.
 23. M. Turhan, & S. Gunasekaran, Kinetics of in situ and in vitro gelatinization of hard andsoft wheat starches during cooking in water. *Journal of Food Engineering*, 2002; 52: 1-7 [https://doi.org/10.1016/S0260-8774\(01\)00058-9](https://doi.org/10.1016/S0260-8774(01)00058-9).
 24. F. Hervas, S.P. Celma, T.C. Berlanga, M.F. Pérez, Q. Gratacós, & C. Ribas, Study of the extraction kinetic of glycosaminoglycans from raw sheepskin trimmings. Proceedings of International United of Environment Commission of IULTCS. Chengdu.China. 2006; 14–16 October.
 25. N. Milićević , P. Kojić , M. Sakać , A. Mišan, J. Kojić , C. Perussello , V. Banjac M.Pojić , B. Tiwari, Kinetic modelling of ultrasound-assisted extraction of phenolics fromcereal brans. *Ultrasonics Sonochemistry*, 2021; 79: 105761. <doi.org/10.1016/j.ultsonch.2021.105761>.
 26. S.W. Chan, C.Y. Lee, C.F. Yap, A.W.M. Wan, and C.W. Ho, Optimisation of extraction conditions for phenolic compounds from *limau purut* (*Citrus hystrix*) peels.*International Food Research Journal*, 2009; 16: 203-213.
 27. D. Franco, J. Sineiro, M. Pinelo, M.J. Nunez, Ethanolic extraction of *Rosa rubiginosa* soluble substances: oil solubility equilibria and kinetics studies. *Journal of Food Engineering*, 2007; 79(1): 150–157. <https://doi.org/10.1016/j.foodeng.2006.01.047>.
 28. F. Sharififar, G. Dehghn-Nudeh, and M. Mirtajaldini, Major flavonoids with antioxidant activity from *Teucrium polium* L. *Food Chemistry*, 2009; 112(4): 885–888. <http://dx.doi.org/10.1016/j.foodchem.2008.06.064>.
 29. Y. Jun, L. Wang, R.L. Walzem, E.G. Miller, L.M. Pike, B.S. Patil, Antioxidant activity of Citrus limonoids, flavonoids, and coumarins. *Journal of Agricultural and Food Chemistry*, Mar 23, 2005; 53(6): 2009-14 <doi:10.1021/jf04846322>
 30. D. Hamdan, M.Z. El-Readi, A. Tahrani, F. Herrmann, D. Kaufmann, N. Farrag, A. El- Shazly, M.Wink, Chemical composition and biological activity of *Citrus jambhiri* Lush. *Food Chemistry*, 2011; 127: 394–403. <https://dx.doi.org/10.1016/j.foodchem.2010.12.129>
 31. R. Gualdani, M.M. Cavalluzzi., G. Lentini, S. Habtemariam, The Chemistry and Pharmacology of *Citrus* Limonoids. *Molecules*, 2016; 21: 1-39. <doi:10.3390/molecules21111530>
 32. O. Nana, J. Momeni, T.R. Nzangué, M.B. Ngassoum, Phytochemical screening,antioxydant and antiplasmodial activities of extracts from *Trichilia roka* and *Sapium ellipticum*. *Journal of Phytopharmacology*, 2013; 2(4): 22-29.