MODELLING OF MULTISTAGE EXTRACTION KINETICS
FOR *NICOTIANA TABACUM L.* – WATER SYSTEM

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ABSTRACT

The extraction of active substances from vegetable raw materials provokes significant interest, because of the wide use of the extracts obtained in pharmaceutics, cosmetics and food industries. Numerous studies for increasing the precision of process description and figuring, and assuring of better degree of extraction of active substance, are carried out. In the present study, a model to describe the solid-liquid extraction kinetics at three stage cross-flow process design is proposed. The time dependence of the concentration profiles in the particle is determined, as well as the bulk kinetics. The influence of the model parameters as: initial concentration in the vegetable raw material particle, effective diffusivity, particle porosity is investigated. The model adequacy is verified with experimental data obtained during three stage cross-flow extraction of Nicotiana tabacum L. with water. The comparison of experimental and numerical data shows very good coincidence, the incertitude being 0,7-6,5%.

Keywords: modelling, extraction, kinetics.

INTRODUCTION

The solid-liquid extraction is a widely used process in pharmaceutics, cosmetics, tobacco and food industries and has also its application for environmental purposes. This interest provokes numerous scientific studies focused on the process kinetics, the diffusivities, the yield of extraction or the process design. The modelling reveals a powerful mean for the optimization of the equipment, simulation, design and control of the solid-liquid extraction, allowing theoretical description of the process and evaluation of the mass transfer coefficients [1-13]. Yet, the process description is very difficult because of the influence of a large number of parameters: solid phase structure, particle shape and size distribution, initial concentration of active substance, temperature, solid-solvent ratio, variation of the kinetic coefficients during the extraction. Generally, two different approaches exist to describe the process, based on experimental data: development of mathematical model set on the differential mass transfer equations and the methods of the standard, kinetic and characteristic functions. In the present study, a mathematical description of the extraction kinetics is made for the *Nikotiana tabacum* leaves – water system, aimed to determine different parameters' influence that give the possibility to control and optimize the process performance.

EXPERIMENTAL

A mathematical model is developed to describe the solid-liquid extraction from vegetable raw materials. We accepted that equations of type (1) and (2) present very correct description of the solid-liquid ex-

traction experimental data [3, 7, 8, 10, 13]. Assuming constant diffusivity and plate particle shape, we obtain for the concentration in the particle:

$$\varepsilon_{\mathrm{p}} \frac{\partial C_p}{\partial t} = D_e \left. \frac{\partial^2 C_p}{\partial r^2} \right|_{r=0}^{r=R} \tag{1}$$

with the following boundary conditions:
- no transfer in the symmetry surface of the particle:

$$r = 0 \qquad \frac{\partial C_p}{\partial r} = 0 \tag{1a}$$

- equality of diffusion and convection transfer on the solid-liquid interface:

$$r = R \qquad D_e \frac{\partial C_p}{\partial r} = \kappa \left(C_p \big|_{r=R} - C_{liq} \right), \tag{1b}$$

where:

ε_p - is the porosity of the solid particle, m^3/m^3;
C_p - is the concentration in the solid particle, kg/m^3;
t - is the time, s;
D_e - is the effective diffusivity, m^2/s;
r – is characteristic dimension, plate thickness, m;
κ – is local mass transfer coefficient, m/s.

The variation of the concentration in the liquid phase is:

$$\frac{\partial C_{liq}}{\partial t} = \frac{\varepsilon_p (1 - \varepsilon_l)}{R \varepsilon_l} D_e \left. \frac{\partial C_p}{\partial r} \right|_{r=R} \tag{2}$$

where:

C_{liq} - is the concentration in the liquid phase, kg/m^3;
ε_l – is liquid phase porosity, m^3/m^3.

To solve the differential equations of the model we chose one of the mostly used in the practice method, those of the finite differences [14, 15]. This is a problem with rigid boundaries and tridiagonal matrix, in which only the coefficients of the three diagonals are different from zero. Efficient for solving a system of linear equations with such matrix is Thomas method.

The transformed equations are presented below:

$$\frac{C_{liq}^j - C_{liq}^{j-1}}{\Delta t} = M \frac{C_{p\,Ikr}^j - C_{p\,Ikr-1}^j}{\Delta r} \tag{3}$$

where

$$M = D_e \left. \frac{\varepsilon_p (1 - \varepsilon_l)}{r \varepsilon_l} \right|_{r=R} ;$$

and for the concentration in the solvent we obtain:

$$C_{liq}^j = M \frac{\Delta t}{\Delta r} \left[C_{p\,Ikr}^j - C_{p\,Ikr-1}^j \right] + C_{liq}^{j-1} \tag{4}$$

The variation of the concentration is the particle:

$$\varepsilon_{\mathrm{p}} \frac{C_{p_i}^{j+1} - C_{p_i}^j}{\Delta t} = D_e \frac{C_{p_{i+1}}^{j+1} - 2C_{p_i}^{j+1} - C_{p_{i-1}}^{j+1}}{\Delta r^2}$$

$$\frac{\varepsilon_p}{\Delta t} C_{p_i}^{j+1} - \frac{\varepsilon_p}{\Delta t} C_{p_i}^{j-1} =$$

$$= \frac{D_e}{\Delta r^2} C_{p_{i+1}}^{j+1} - \frac{2D_e}{\Delta r^2} C_{p_i}^{j+1} + \frac{D_e}{\Delta r^2} C_{p_{i-1}}^{j+1} \tag{5}$$

From equation (5) we are able to determine the coefficients of the matrix:

$$A_i C_{p_{i+1}}^{j+1} + B_i C_{p_i}^{j+1} + S_i C_{p_{i-1}}^{j+1} = F_i^j$$

$$A_i = -\frac{D_e}{\Delta r^2} ,$$

$$B_i = \frac{\varepsilon_p}{\Delta t} + \frac{2D_e}{\Delta r^2} ,$$

$$S_i = -\frac{D_e}{\Delta r^2} , \quad F_i^j = \frac{\varepsilon_p}{\Delta t} C_{p_i}^{j-1} \tag{6}$$

RESULTS AND DISCUSSION

All the parameters of the model are determined experimentally [6, 9, 10]: the particle porosity ε_p=0,7;
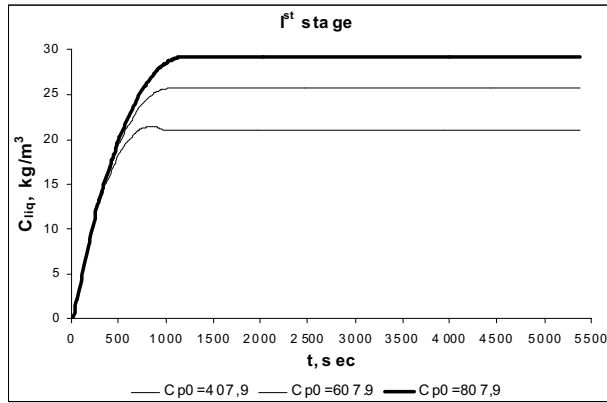
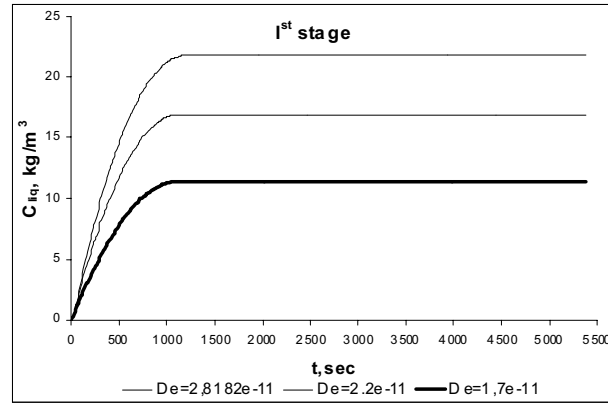Fig. 1a. Influence of the initial concentration on the first stage yield.

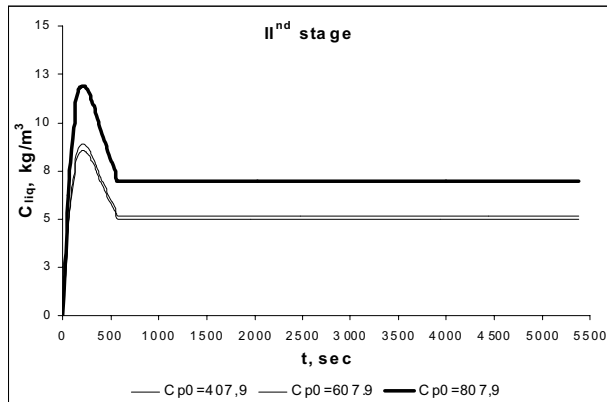Fig.2a. Influence of the effective diffusivity on the first stage yield.

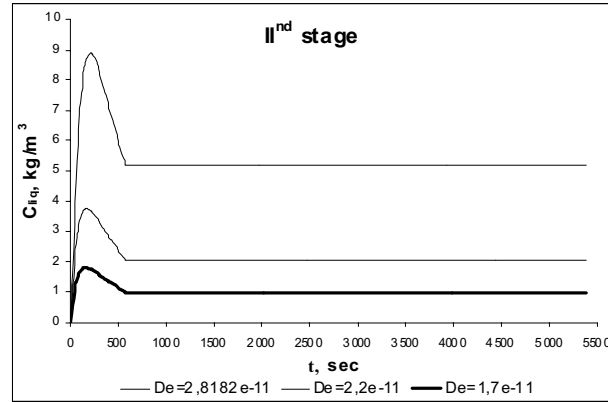Fig. 1b. Influence of the initial concentration on the second stage yield.

Fig. 2b. Influence of the effective diffusivity on the second stage yield.

local mass transfer coefficient, $\kappa= 1,5.10^{-6}$m/s; the initial concentration of active substances in the solid phase $C_{p0}=607,9$ kg/m³; the effective diffusivity $D_e=1,7.10^{-11}$ m²/s and $D_e=2,8182.10^{-11}$ m²/s (the effective diffusivity was evaluated using two different approaches); the plate thickness $R=1,8.10^{-4}$m; the experiment duration $\tau=5400$s. Numerical experiments are made changing some of these parameters influencing the extraction yield. The modelled process is three stage cross flow extraction, the hydromodule being $\xi=0,02$, n=7s^{-1}: it has been shown that mixing at this angular velocity ensures that the systems are in the purely internal-diffusion-controlled regime.

Initial concentration, C_{p0}. First the influence of the initial concentration of active substances in the solid phase on the yield is studied. C_{p0} is varied ±30% of the nominal value. The results for the first and second stage are presented in Fig.1a and 1b.

As shown in the figures, the initial concentration of tobacco concrete in the solid phase influence mostly the first stage, because of its more important concentration in the solvent. The pattern of the curves is similar, wherever C_{p0} is, as well as the time for reaching equilibrium. As expected, the yield in the liquid phase is proportional to the initial concentration in the particle. The third stage curves present the same result.

Effective diffusivity D_e. The effective diffusivity is varied from $1,7.10^{-11}$m²/s – value obtained using the regular regime method and $2,8182.10^{-11}$m²/s – value obtained using the standard function method [9]. The numerical results are presented in Fig.2a and 2b.

We observe that increasing the effective diffusivity results in an increase of the yield. The curves preserve the same pattern and time for reaching equilibrium. The third stage curves present the same result.
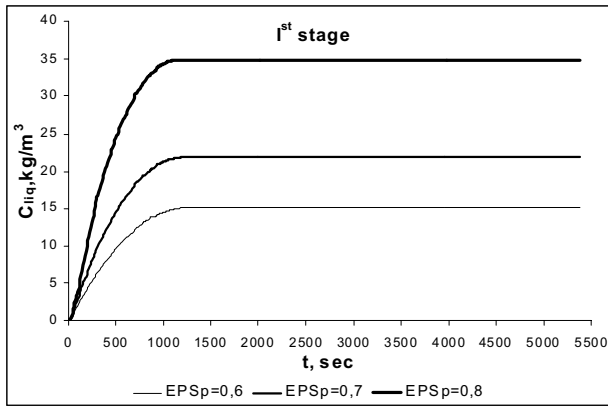
121

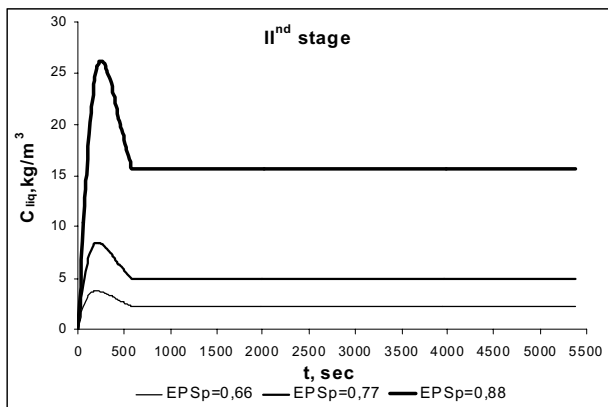Fig. 3a. Influence of the porosity on the first stage yield.

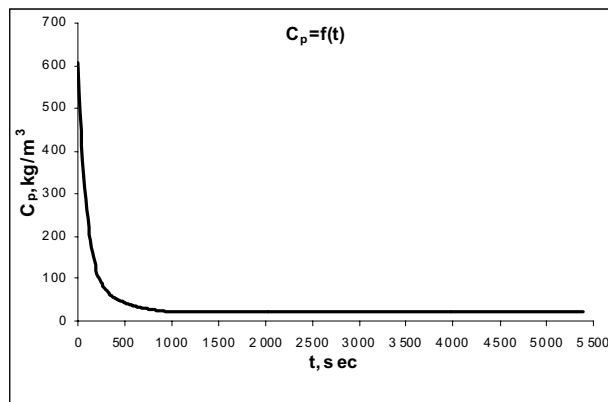Fig. 3b. Influence of the porosity on the second stage yield.

Fig. 4. Variation of the concentration at the particle surface in time.
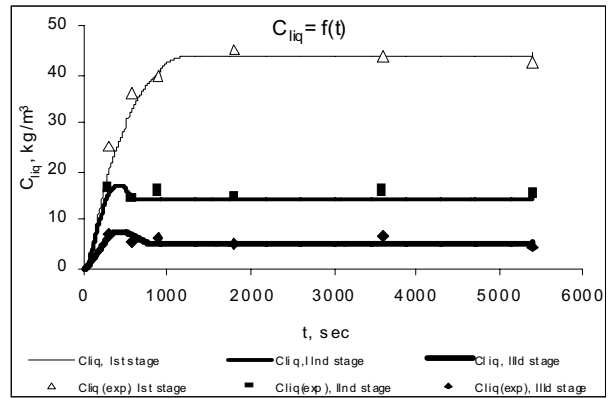

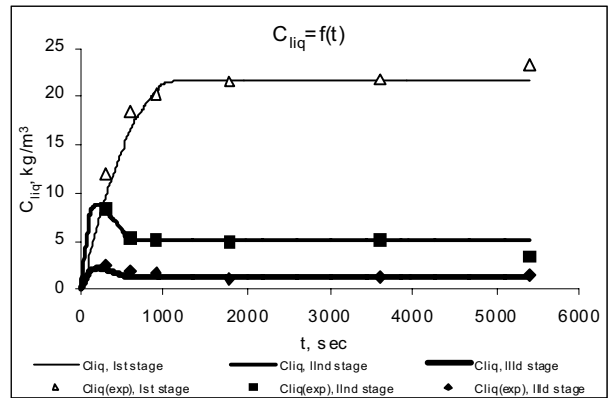Fig.5a. Experimental and numerical data comparison, $\zeta=0,01$.

Fig. 5b. Experimental and numerical data comparison, $\zeta=0,02$.
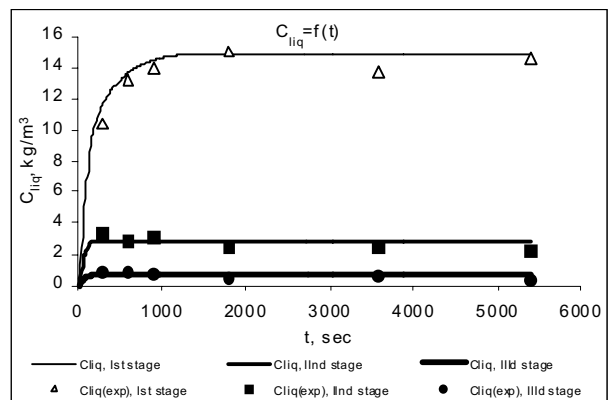
Fig. 5c. Experimental and numerical data comparison, $\zeta=0,03$.

Particle porosity, ε_p. In order to determine the influence of the particle porosity on the extraction yield, numerical experiments are run for $\varepsilon_p=0,6$; 0,7 and 0,8. The results for the first and second stage are shown in Fig.3a and 3b, those for the third stage differ only in concentration, not pattern.

Variation of the concentration in the particle, C_p. The variation of the concentration at the particle surface in time is plotted in Fig. 4. The parameters of the model are those, experimentally obtained for the first stage, cross flow extraction of *Nicotiana tabacum* leaves with water. We observe a rapid decrease of the concen-

tration in the particle at the initial period that corresponds to the rapid increase of the concentration in the solvent. After this period, the concentration stabilizes reaching the value in equilibrium with the liquid phase.

Experimental data description. The model is tested for description of the experimental kinetics data obtained for the three stage, cross flow extraction of *Nicotiana tabacum* leaves with water. The parameters of the model are: C_{p0}, ε_p, R, D_e, κ, τ. The experimental and numerical results are compared in fig.5. The average deviation of the model and experimental data is 0,7-6,5% for the three stages and for the three hydromodules, which are studied.

In order to fit better the model to the experimental data, the concentration peaks for the second and the third stage are described numerically introducing in the equation for the liquid phase an additional source term. It's supposed that this concentration peaks are related to the higher equilibrium concentration in the previous stage: the *Nicotiana tabacum* leaves immerged with this higher concentration solution arrive in the next stage. For that reason this source is proportionally related to the equilibrium concentration for the previous stage: thus for the first stage it is zero, and inverse proportionally to the time square: thus its influence became negligible after the initial period.

CONCLUSIONS

A mathematical description is proposed for a three stage cross flow extraction from vegetable raw material. The numerical solution is based on the finite difference method and Thomas method. Numerical experiments are run in order to determine the influence of the different parameters on the kinetics process. The model is tested with experimental data for three stage cross flow extraction for the system *Nikotiana tabacum L.* - water. A source term is added to fit better the model to the experimental data. The comparison of experimental and numerical data are in very good coherence, the incertitude being 0,7-6,5%.

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