APPLICATIONS OF ARTIFICIAL NEURAL NETWORKS IN SUPERCRITICAL FLUID EXTRACTION MODELLING AND SIMULATION

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ABSTRACT

In this work, the artificial neural networks (ANN) technology was applied to the simulation of supercritical fluid extraction (SCFE) process of vegetable oil. For this technology, a 3-layer BP network structure is applied, and the operation factors such as pressure, temperature and extraction time are used as input variables of the network, whereas the oil yield of extraction is treated as output values of the network. Optimization of the topological structure of the net, 6 neurals of middle hidden layer had been proved to be the optimum value.

With the normalization pretreatment of the initial input data, not only the convergent speed and accuracy has been improved greatly, but also the problem of derivative at zero point has been solved. Therefore, the method is an improved model based on Fullana [1]. In addition, we investigated the impact of the output variables on the net properties, and found that it is very convenient for the data correction when the extraction rate-time curve is set as training sample.

An ANN-SCFE simulation system has been programmed. For the first time, the simulation for the SCFE procees of Hippophae Rhamnoides L. seed oil has been made, and the results show that the average absolute relative deviations (AARD) are lower than 6%.

INTRODUCTION

Compared with the traditional grinding technique and solvent extraction, supercritical fluid extraction using CO_2 has gained more and more attention due to the high purity of the product oil and free from solvent contamination. From the mathematical point of view, the models previously proposed are generally based on differential mass balances integration. Bulley et al.[2], Lee et al.[3] and Fattori et al.[4] assumed that mass transfer resistance occurred only in the solvent phase. In other models an internal mass transfer resistance was considered (Roy, [5]). A sigmoidal equilibrium curve was adopted by Perrut et al. [6] to fit the experimental data. King et al. [7] used a shrinking core model to describe a variable external resistance where the solute balance on the solid phase determines the thickness of the mass transfer layer in the external part of the particles. However, more reliable models can be produced when the vegetable structure of the seed is considered in the equations. From this respect, Sovova et al. [8] proposed a model based on the hypothesis that the internal part of seeds is formed by specialized structures that contain the oil. Other literatures can see [9-12].

In general, however, the above models require experimental data and parameter fitting. Although a few parameters can be obtained from correlations, the intrinsic rate and fluid-solid

equilibrium data should have a rather firm experimental basis (i.e., from adsorption isotherms, rate coefficients, equilibrium solubility, etc.). The above critique on semi-theoretical or mechanistic models would suggest not to put too much stress on the validity of the detailed external and intraparticle mass transport coefficients, but instead it would be wise to use alternative types of rate formulations. Fullana et al. [1] proposed a model based on ANN, it can simulate the SCFE process very well. However, their ANN system will not be able to train very well when there is a zero point input data. So near the original point of the coordinate, they still have to use the data fitting method. The aim of this work is to investigate the ANN simulation method for SCFE process, and to improve the ANN model by means of modification the initial sample data.

I - MATERIALS AND METHODS

The experiments were carried out in an apparatus designed by our laboratory. The apparatus is composed of a high-pressure vessel with a volume of 1L and two separators with a volume of 1L and 0.3L, respectively. The high-pressure vessel contains an extraction basket with a volume of 300mL. To reduce the dead space and to allow the uniform distribution of solvent, a layer of SS beads with diameters of 4mm was placed on the bottom of the extractor basket. CO_2 with a purity of 99% was employed as solvent. Hippophae Rhamnoides L seed (produced in eastern area of Inner Mongolia, China) was milled and sieved to appropriate particle size before use. Temperature was controlled by use of the electric heating belt. When the operating temperature was raised to preset value, CO_2 was pressurized, preheated and enter into the extractor. After the extraction, the CO_2 was depressurized, separated and allowed to vent. The seed oil was recovered from the separator after a certain period and weighed. The flow rate was measured by the flow meter.

II - MATHEMATICAL MODELS

Since the scale-up of the equipment and the evaluation of the cost of a process cannot be done without mass transfer rate data in a convenient form, several methods have been proposed for modeling the supercritical extraction process. In this article, the modeling is based on the differential mass balance equation along the axial direction of the extractor bed.

III -I EMPIRICAL MODEL

The modeling of the extraction process is based on the following hypotheses.

1. We suppose that the behavior of all compounds extracted is similar and can be described by a single pseudo-component with respect to the mass transfer phenomena.
2. Concentration gradients in the fluid phase develop at larger scales than the particle size.
3. The solvent flow rate, with superficial velocity *u*, is uniformly distributed in all the sections of the extractor.
4. The volume fraction of the fluid, ε , is not affected by the reduction of the solid mass during extraction.

Neglecting radial mass dispersion effects, the conservation equation of a solute in a packed bed can be written as

$$\rho\varepsilon\frac{\partial C}{\partial t}+\rho u_0\frac{\partial C}{\partial z}-\rho\varepsilon D_{ax}\frac{\partial^2 C}{\partial z^2}=J(C) \qquad (1)$$

In which axial mass dispersion is considered. In Eq.(1), *J* represents the rate of solute mass transfer from the outer surfaces of the seeds to the fluid per unit of bed volume. Assuming that the fluid travels in plug flow at bed entrance and is free from solute, the initial and

boundary conditions for the above problem are

$$C\big|_{z,t=0} = 0 \qquad (2a)$$

$$-reD_{ax}\left(\frac{\partial C}{\partial z}\right)_{z=0^+,t} + ru_0 C\big|_{z=0^+,t} = 0 \qquad (2b)$$

$$\left(\frac{\partial C}{\partial z}\right)_{z=L,t} = 0 \qquad (2c)$$

The mass transfer rate, *J*, will be written in terms of a product of the overall mass transfer coefficient, *U*, and the specific mass transfer area, *A*. So, *UA* will be the mass transfer rate per unit driving force, expressed as kg of solute/(m3 bed s). The driving force is C^*-*C*, thence *J* can be written as

$$J=UA(C^*-C) \qquad (3)$$

In Eq. (3), C^* is the mass fraction of solute oil in the fluid at saturation for the system temperature (solubility).

In order to use neural computing, a net trained with the differential data is available in terms of extraction yield, pressure, temperature and time. The vertical packed-bed extractor is divided into horizontal slices or layers each containing approximately the same amount of seeds as in the thin beds used in differential runs. If the amount of extracted oil from a layer is known, at given pressure and temperature, then the time elapsed after the start of extraction for any layer in the pile of layers will be calculated iteratively. The quantity of oil extracted is calculated from the equation

$$q_{ex_j}^{k} = q_{ex_j}^{k-1} + (C_i^k - C_{i-1}^k)Q\Delta t \qquad (4)$$

By knowing the amount of seeds and the fluid flowrate in a differential bed, it is possible to infer the concentration of the solute in the fluid in contact with the layer *i*. The expression for the hybrid neural concentration will be written as:

$$C_i^{NN} = \frac{(\frac{dq}{dt})_{t_i} \cdot q_s}{Q} \qquad (5)$$

here, *q* is the amount of oil extracted (kg/kg seeds), and *dq/dt* is the rate of oil extraction at time *t*.

Finally, in order to integrate Eq. (1), it is necessary to have a value of the mass transfer rate given by Eq. (3). This will be so provided that fluid velocity is the same in the integral bed and in the laboratory extraction runs. The value used for *UA* for a layer *i* in the calculations is obtained from the following relationships:

$$C_i^{NN} Q = J_i^{NN} V \qquad (6)$$

$$(UA)_i = \frac{Q/V}{(C^*/C_i^{NN} - 1)} \qquad (7)$$

In the programs, the following partial calculations or modules were implemented: The axial dispersion coefficient was calculated from a correlation (Catchpole et al. [13]). The solubility of oil in SCF was calculated with del Valle method [14]. The integration of Eq.(1) was made by using the method of lines in combination with the Crank-Nicolson algorithm. Fluid density was estimated using the P-R EOS. Neural net computations were obtained from differential bed modeling. In this article, the ANN model was referenced from Fullana [1]. For this technology, a 3-layer BP network structure is applied, and the operation factors such as

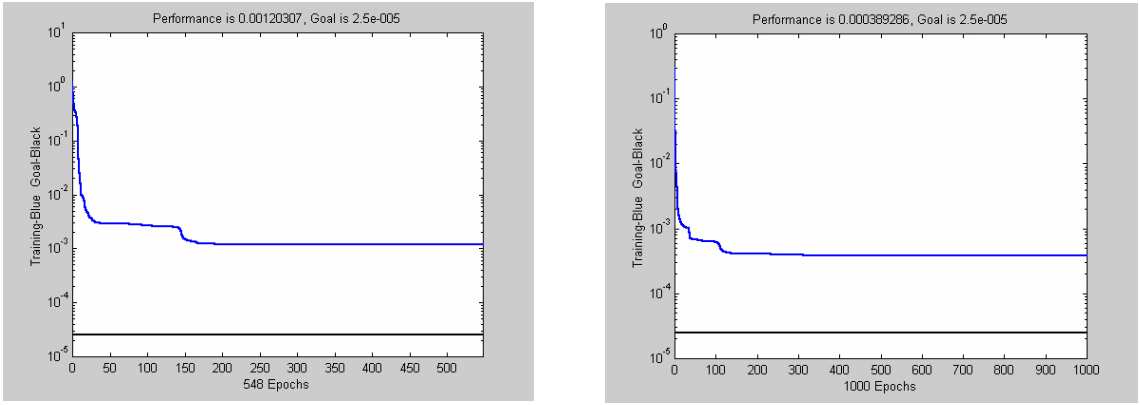
pressure, temperature and extraction time are used as input variables of the network, whereas the oil yield of extraction is treated as output values of the network. Optimization of the topological structure of the net, 6 neurals of middle hidden layer had been proved to be the optimum value.

III -II NORMALIZATION PRETREATMENT OF THE INITIAL INPUT DATA

For the SCFE process, we should use the pressure, temperature, time as the input parameters and extraction yield as the output one for the ANN system when training the ANN system. In our experimental conditions, the pressure P is about 15-30MPa, the temperature T is about 303-323K, time t is about 0-9000s and extraction yield q is about 0-5g. Comparing the amount of these four parameters, we can find out that there is an infinite difference among them. It can make the network produce a huge difference for the joint weight in every node. This input and output data could reduce the properties of the ANN system, and make the ANN training very difficult, and at the same time, it can decrease the accuracy of the ANN prediction to SCFE process. On the other hand, ANN system will not be able to train very well when there is a zero point input data. It is why Fullana et al.[1] called their method as "a neural-regressive hybrid prediction system". In this article, in order to overcome the above difficulties and make the Fullana's method improvement, we first time use the normalization method to modify the initial input and out data. The detailed formula is as follows:

$$x_i^* = 0.9\frac{x_i - x_{i\min}}{x_{i\max} - x_{i\min}} + 0.05 \qquad (8)$$

here, x_{imin} is the minimum value of the i^{th} input data, x_{imax} is the maximum value of the i^{th} input data, and x_i^* is the new value after normalized treatment. It is obviously that all data can be mapped into [0.05, 0.95] after normalized treatment. The data not only are no the huge difference, but also have no the zero point for ANN system training. From Fig.1, we can find out the prediction accuracy and convergent rate overall are very well. Fig.1 shows the situation of error (in terms of AARD%) vs. number of iterations for back-propagation with 5, 6 units in the hidden layer.



(a) 5 units (b) 6 units

Figure 1. Training and Learning Curves

IV- RESULTS AND DISCUSSION

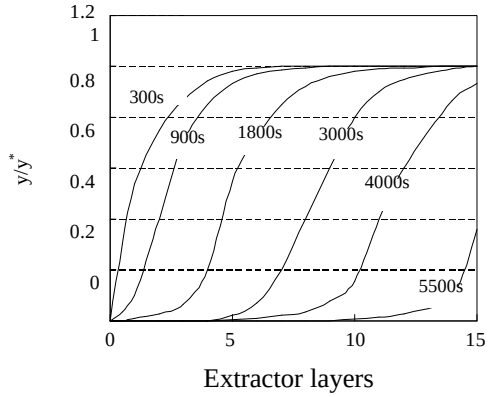
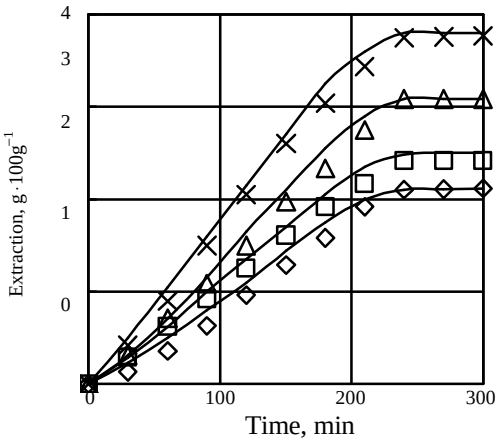


Fig.2 Concentration Curves of Beds Simulated at Different Time Interval



? 15MPa, 35? —ANN simul., 15MPa
? 20MPa, 35? —ANN simul., 20MPa
? 25MPa, 35? —ANN simul., 25MPa
× 30MPa, 35? —ANN simul., 30MPa

Fig.3 Simulation and Experiment

In this article, An ANN system was developed that is used to simulate SCFE processes based on Matlab5.3 software. For MATLAB5.3 software, it has an ANN Tool-Box, we can make our own special program very conveniently. So the kinetic model of artificial neural networks (ANN) was established based on differential mass balance of packed bed. Using a 3-layer BP network, with extraction pressure, temperature and extraction time as the input variables, the extraction yield as the single response for the net, the network was trained with experimental data. The results show that the trained network can simulate the extraction rate of the Hippophae Rhamnoides L. seed oils. Besides, the network can make a good prediction for the change of fluid concentration with the bed position and extraction time. By using this system to train and predict our experimental data (for sample data), the average absolute relative deviations (AARD) are less than 0.23% (for training group) and 0.5% (for test group) respectively. Fig.2 shows the relationship between the fluid concentration and the extraction bed layers, where the advancement of the mass transfer zone is seen for a bed of 30 cm in length. This simulates the integral bed using differential kinetic data, scaled up from laboratory values. As soon as the concentration wave reaches the bed exit, solute starts to appear in the leaving stream giving rise to the breakthrough curve. These phenomena are accordance with the theoretical analysis result for the packed bed mass transfer processes. The results of experiments and ANN simulations are also compared in the paper, shown in Fig.3. Under the conditions of 35? , 15-30MPa, and 0.2m^3/h, when time t is 5500s, we calculate the oil yield as a function of the extraction time. It is indicated that the ANN system can predict the SCFE process in very well and the maximum error of average absolute relative deviations is less than 6%.

V-CONCLUSION

In this work, the artificial neural networks (ANN) technology was applied to the simulation of SCF extraction process of vegetable oil. For this technology, a 3-layer BP network structure is applied, and the operation factors such as pressure, temperature and extraction time are used as input variables of the network, whereas the oil yield of extraction is treated as output values of the network. Optimization of the topological structure of the net, 6 neurals of middle hidden layer had been proved to be the optimum value.

With the normalization pretreatment of the initial input data, not only the convergence speed and accuracy has been improved greatly, but also the problem of derivative at zero has been solved. Therefore, the method is an improved model of Fullana [1]. In addition, we investigated the impact of the output variables on the net properties, and found that it is very convenient for the data correction when the extraction rate-time curve is set as training sample.

An ANN-SCFE simulation system has been programmed. For the first time, the simulation for the SCFE process of Hippophae Rhamnoides L. seed oil has been made, and the results show that the AARD is lower than 6%

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