Application of Hybrid Predictive Tools for Prediction and Simulation in Supercritical Fluid Extraction – An Overview

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Abstract. Supercritical fluid technology (SFT) has been applied in many areas, such as in pharmaceutical and food sectors, due to its outstanding features. SFT is an efficient technology that performs extraction and leaves no or less organic residues compared to conventional processes. Recently, the simulation and prediction of the process output from supercritical fluid extraction was determined using intelligent system predictive tools. The prediction of the set of results from supercritical fluid extraction for designing and scale up purposes is crucial because it can not only reduce the usage of extraction solvent and the energy and time of the process but it can also solve the problem that the complex mathematical model cannot solve. A neural network is considered as one of the artificial intelligent systems and is a key technology in industry 4.0. The use of hybrid predictive tools is also a developing area in the prediction and simulation of supercritical fluid extraction and therefore will be further discussed in this paper.

1. Introduction

Supercritical fluid technology is now widely used in many areas such as in the chemical processing, food and pharmaceutical sectors. Supercritical fluid technology utilizes the unique properties of a supercritical fluid, namely, the pressure dependent dissolving power, to alter the process performance. Supercritical fluid offers the exclusive behaviour of possessing the dual characteristics of a fluid, which are gas and liquid. Among all the supercritical fluid available, carbon dioxide is a commonly used fluid because it is inexpensive and safe, it is available in high purity, and it can be used under mild conditions. Moreover, carbon dioxide is a small sized molecule with a linear structure that offers fast diffusion compared to other solvents. The critical temperature and pressure for carbon dioxide are 31.3°C and 71.9 bars; these mild conditions and this non-toxic behaviour of carbon dioxide have provided benefits to many food and health-related processes [1].

To apply supercritical fluid technology, fundamental knowledge, such as solubility, is crucial to control, design and initiate a more efficient and environmental friendly extraction process. For prediction purposes, in addition to having the appropriate mathematical model, the values of the experimental data and the thermodynamic properties of the sample and the extract are required. There
are traditional mathematical modelling approaches for supercritical fluid extraction to determine solubility, as they can be developed from differential mass balance equations for the packed solid bed and, if possible, they can represent the overall extraction process. Some of the examples are hot ball diffusion, broken and intact cells, shrinking cores and desorption models. Assumptions and simplifications of a mathematical model are sometimes required, causing the model to not be reliable. However, many conventional mathematical models have been both challenging and impractical. Therefore, the prediction of the effects of temperature and pressure towards the yield and solubility of the supercritical fluid extraction process using tools is now developing because of time and cost concerns.

Predictive tools using intelligent systems are now increasingly being used for the prediction, to model and simulate a chemical process, as well as to compute, classify and optimize a process for process control. Intelligent systems apply the ability to learn, observe and memorize in a situation full of factors and important data. The main advantage of intelligent systems is that the predictions can be performed easily and quickly, as well as in an accurate manner, for systems that are difficult to simulate using physical models. Intelligent systems address imprecision, uncertainty, partial truth and approximation to achieve tractability, robustness and, more importantly, low solution cost. Examples of intelligent systems are neural networks, fuzzy systems and evolutionary algorithms. Thus, there is an increasing number of studies on the solubility of different compounds in supercritical fluid using intelligent systems. Nevertheless, this information is not well summarized. Therefore, this paper discusses several works involving the utilization of hybrid predictive tools based on intelligent systems for the simulation and prediction of the yield of plant extracts using the supercritical fluid extraction process.

2. Predictive Tools

Among all the predictive tools from intelligent systems that have been applied, the Artificial Neural Network (ANN) has been the common tool used for prediction because it is easily referenced and applied. ANN is based on the biological nervous system, which consists of a large number of neurons in an architecture inspired by the brain. Neural networks learn by example; therefore, they are trained by known examples of a problem to attain knowledge about it. After being properly trained, the network can be put to effective use in solving unknown parameters of the problem [1]. Over the years, neural network systems have evolved.

The architectures of neural networks can be identified by three types: single layer feedforward networks, multilayer feedforward networks and recurrent networks. There are various types of learning methods for neural networks; one of the most applied method is the backpropagation learning rule. Neural networks have been applied to several supercritical fluid extraction processes in predicting the yield of extraction [2]–[7], the initial slope [5] and the solubility [8]–[10].

3. Hybrid Predictive Tool Systems

Hybrid denotes the integration of two or more systems. Prediction using predictive tools with intelligent systems is a system integrating two or more technologies. The integration of technologies can effectively solved problems compared to using the technology individually.

3.1. Adaptive Neuro-Fuzzy Inference System (ANFIS)

Application of ANN individually leads to problems such as unsatisfactory extrapolation and also creating a ‘black-box’ problems. Thus, suggestion on hybrid models have been developed by combining ANN with simple models. These hybrid neural network models are supposed to perform better in terms of process identification tasks since extrapolation are confined only to the uncertain parts of the process [11].

The Adaptive Neuro-Fuzzy Inference System (ANFIS) is considered one type of hybrid model since it combines the Artificial Neural Network (ANN) and the Fuzzy Inference System (FIS) system. There are 5 layers that mapping the inputs through input membership functions (MF) and fuzzy rules
in the ANFIS architecture. Similarly, output mapping is achieved through output membership functions along with its fuzzy rules. The number of membership functions assigned to each input variable is determined by trial and error. The hybrid of a neural network and the fuzzy logic in ANFIS allows it to have both the low-level learning and the computational power of neural networks as well as the advantages of the high-level human-like thinking of fuzzy systems and they can complement each other [12]. There are several studies using ANFIS to predict the solubility [13] and the mass of the extract [14, 15]. Some studies even compare the results of the application of ANN and ANFIS to identify the best system that can represent the data and that is reliable for optimization processes [16–18], [19]. A detailed description of the previous research applying ANFIS and the hybrid of it for yield and solubility predictions is shown in Table 1.

### Table 1. Compilation of research applying ANFIS tools for predicting yield and solubility of SFE.

| Sample               | Purpose                              | Remarks                                                                 | References |
|----------------------|--------------------------------------|-------------------------------------------------------------------------|------------|
| *Pimpinella anisum* L. Seed | Prediction mass of extract            | • The input parameters: pressure, solvent mass flow rate, and extraction time | [12]       |
|                      |                                      | • The output parameter: mass of extract                                 |            |
| Pomegranate          | Simulation of oil yield              | • Comparing the data obtained between the ANN and ANFIS models          | [16]       |
| *Glycyrrhiza glabra* L. | Modelling the recovery of extraction | • Independent variables: dynamic time (t), pressure (P), temperature (T) and flow rate of SCCO\textsubscript{2} (Q)  
• Dependent variable: Recovery of extract | [17]       |
| *Rosa damascene* Mill | Modelling the recovery of extraction | • Independent variables: dynamic time (t), pressure (P), temperature (T) and flow rate of SCCO\textsubscript{2} (Q)  
• Dependent variable: Recovery of extract  
• Comparing the data obtained between ANFIS and RSM | [14]       |

#### 3.2. Mathematical Model-Neural Network Hybrid

In 2003, a group of researchers from Canada proposed a hybrid model of the Radial Basis Function-Peng Robinson (RBF-PR) model. The hybrid model was developed by first developing a simple RBF model. The simple RBF model consists of three inputs of T, p and other factors, with one output, which is yield rate. This model has no knowledge of the whole process. To overcome the black box problem of the RBF model, the model is correlated with the Peng-Robinson equation to become a hybrid RBF-PR model. In the Peng-Robinson equation of state, there is an unknown interaction parameter $k_{12}$, for a binary mixture, whereby the predicted solubility is sensitive. The $k_{12}$ can be obtained from the physical property data for the mixture, but it requires trial and error processes to obtain it. Moreover, the parameters are all temperature dependent, which does not fit the data compared to pressure. The proposed hybrid model by [20] can compensate for the black box problem. The proposed model can fit the experiment data well and can retain the physical meaning of the whole SFE process.

Ten years later, a parallel hybrid model was developed by researchers from Iran [15]. Combination of conventional mathematical modelling with ANFIS was used to predict the Epigallocatechin gallate (EGCG) recovery of supercritical extraction. The conventional model used was based on the differential mass balance in the solid and mobile phases. An analytical model, can improve the accuracy of ANFIS in non-training domains. Besides that, it can predict the unknowns of the analytical parameters. The hybrid model simulates the extraction system with any arbitrary value of adjustable parameters of mathematical modelling [21]. To date, only a few studies has approached the
application of a hybrid model between neural networks and mathematical models. As a result, it is recommended to venture into this area of study in terms of the prediction of values other than the yield and recovery of extracts.

4. Analysis of Model
In both ANN and ANFIS, the correct and optimum structures were the most important problems during training. For ANN, the performance of the network will not be satisfied when the number of the neurons in the hidden layer is small or excessive. Too many neurons leads to a long length of training, and it may be compromised by over fitting. For ANFIS, whenever the number of input membership functions is increased, the structure becomes more complex, requiring more iterations to achieve the convergence to the target error. Thus, the training process is very long. Although, in the end, the predictive tools can solve the problem; the acceptance of the model highly depends on the values of the coefficient of determination ($R^2$) and the absolute average deviation (AAD). The correlation was meaningless because the model does not contain any meaning to the process.

The hybrid of the mathematical modelling and neural network was very promising because it overcomes the above-mentioned problem. The hybrid model combines the application of the mathematical model derived from the theory with the intelligent system. The limitation of solving the complex mathematical model can be resolved by applying the predictive tools, resulting in generating a much more meaningful model to be used in the prediction of yield and solubility values in supercritical fluid extraction processes. As seen in table 1, the prediction of the recovery and yield of the extract are the top applications of ANFIS.

In addition, a hybrid of the mathematical model with the neural network was mostly applied for the prediction of the yield of the extract. In ANN application, in addition to the recovery and yield, two other crucial predictions can be made: solubility and initial slope. Therefore, the hybrid between the mathematical model and the intelligent system may be applied for predictions other than the yield and recovery; such an approach will be an interesting effort because the correlation with solubility was different from that of the extraction yield mathematical model.

5. Conclusion
Overall, the application of an intelligent system to model and simulate a chemical process was achieved in the SFE process. Intelligent system models generalize the experimental results and present process behaviour, predicting and estimating problem. The model can be applied individually or by combining two or more models, called the hybrid model, to achieve the best estimation and prediction data. The appropriate model is beneficial in the development of new products by saving experimental time and costs. However, no research has been performed on the solubility of SFE using a hybrid model; thus, a comparison between a mathematical model and an intelligent model is recommended for further research.

Acknowledgements
The authors would like to thank the Ministry of Higher Education (MOHE) and Universiti Teknologi MARA for providing financial support. Moreover, the authors would like to thank Universiti Kebangsaan Malaysia for providing research facilities.

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