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SCIENTIFIC PAPER

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# NEW METHOD BASED ON NEURO-FUZZY SYSTEM AND PSO ALGORITHM FOR ESTIMATING PHASE EQUILIBRIA PROPERTIES

**Article Highlights**

- Phase equilibria modeling is the key to the development and design of the separation process
- New method based on ANFIS and PSO algorithm for estimating the solubility of solids in $scCO_2$
- The ANFISi approach is used to estimate the critical properties from the solubility data
- A comparative study between the most used optimization algorithm where PSO gives the best results

**Abstract**

*The subject of this work is to propose a new method based on the ANFIS system and PSO algorithm to conceive a model for estimating the solubility of solid drugs in supercritical $CO_2$ (sc-$CO_2$). The high nonlinear process was modeled by the neuro-fuzzy approach (NFS). The PSO algorithm was used for two purposes: replacing the standard backpropagation in training the NFS and optimizing the process. The validation strategy has been carried out using a linear regression analysis of the predicted versus experimental outputs. The ANFIS approach is compared to the ANN in terms of accuracy. Statistical analysis of the predictability of the optimized model trained with a PSO algorithm (ANFIS-PSO) shows a better agreement with the reference data than the ANN method. Furthermore, the comparison in terms of the AARD deviation (%) between the predicted results, the results predicted by the density-based models, and a set of equations of state demonstrates that the ANFIS-PSO model correlates far better with the solubility of the solid drugs in $scCO_2$. A control strategy was also developed for the first time in the field of phase equilibrium by using the neuro-fuzzy inverse approach (ANFISi) to estimate pure component properties from the solubility data without passing through the GCM methods.*

*Keywords: modeling, ANFIS, artificial neural networks, critical properties, particle swarm optimization.*

Supercritical fluid technology is one of the most promising technologies to replace conventional techniques with plenty of advantages. Non-toxicity, low cost, availability, and the facility of separation are the major advantages of using this technology compared to the conventional one.

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The knowledge with detail and accuracy of the equilibrium solubility is the key to the development and design of the separation process. With the various experimental data relative to the solubility of solid solutes in supercritical $CO_2$ ($scCO_2$) being published every year, the modeling of phase equilibria becomes of the primordial importance for the design and optimization issues, which leads to a gainful high selective process.

Artificial intelligence (AI) has been widely used in recent years in many fields of chemical engineering [1-3], renewable energy [4,5], and other areas [6-8] because of their good capacity of modeling and representing the different studied phenomena. Artificial

neural network (ANN) among all techniques becomes very popular in modeling different engineering problems because of their ability to extract very complex relationships from serious nonlinear problems. However, some drawbacks can be accompanied by the use of ANN (the learning process is slow, and their optimized parameters are difficult to analyze where ANN is considered as a black-box tool [9]). To surpass these disadvantages, ANN can be combined with other intelligent techniques called "hybrid systems." In a previous work [10], a hybrid method based on the ANN and PSO algorithm was applied successfully to estimate the solubility of solid solutes in $scCO_2$. The ANN inverse method was used to predict the critical condition properties without using group contribution methods. In the same work, the advantage of considering the particle swarm optimization algorithm (PSO) compared to other optimization techniques (genetic algorithm and ant colony optimization (ACO)) and the importance of the supercritical technology compared to other dissolution techniques (ionic liquids and hydrotropes) were highlighted and well discussed.

In this paper, a new hybrid method based on the adaptive neuro-fuzzy inference system (ANFIS) in combination with a PSO algorithm is applied for the first time in the phase equilibrium area to evaluate the ability of this approach for estimating the solubility of solid drugs in $scCO_2$, which includes: four methoxybenzoic acid isomers (naphthalene, 2-methoxybenzoic acid, 3-methoxybenzoic acid, 4-methoxybenzoic acid) [11], cholesterol [12], five phenol derivatives (p-nitrophenol, m-nitrophenol, 2,4 dinitrophenol, 2,5 dinitrophenol, and picric acid) [13], eight pharmaceutical drugs (ibuprofen, 5-fluorouracil, azodicarbonamide, thymidine, 2-phenyl-4H-1,3-benzoxazin-one, naproxen, taxol, and acetaminophen) [14], and penicillin [15]. Also, a control strategy is adopted and tested for the first time by using the inverse of the ANFIS method for estimating the critical properties of pure solid components. It is to mention that the reason for considering the ANFIS approach will be discussed in the next section.

### *Neuro-fuzzy system*

Hybrid methods are widely used in many fields because of their high ability to adapt to various real-world problems and the possibility of combining more techniques in the modeling and optimization process.

ANFIS is an artificial intelligence method (AI) that combines artificial neural network networks and fuzzy interference systems (FIS). This method was first introduced by Jang [16].

As it was mentioned in the introduction section, ANN has two main problems:

The learning process is slow.

Analysis of their optimized parameters is complex.

To correct the second problem, the fuzzy logic, which is good in explaining their behavior because fuzzy rules can be used successfully (where ANN is weak), but their capacity to acquire the knowledge is complex (where ANN is strong). This can make a neuro-fuzzy system a high predictive approach that takes advantage of both ANN and fuzzy logic. Also, the PSO algorithm, known as a good optimization tool, can surpass the first problem and enhance the learning ability of the ANFIS model.

This work applies the ANFIS technique with PSO in training drugs to estimate solid drugs' solubility in $scCO_2$. The choice of these solids is justified by the availability of the experimental solubility data in the literature and their biological and pharmaceutical interest.

### *Adaptive neuro-fuzzy inference system (ANFIS)*

The ANFIS is Jang's hybrid neuro-fuzzy system developed in 1993 [R10]. ANFIS combines the fuzzification technique of fuzzy logic with the learning capability of ANN to facilitate the hybrid learning procedure [17]. The ANFIS architecture consists of five layers: fuzzified layer, artifact layer, standardized layer, de-fuzzified layer, and output layer. Each layer consists of a number of nodes that perform different operations according to the internal node function [18]. Based on a simple structure that considers two inputs and one output, the ANFIS with Takagi-Sugeno type is represented following Fig. 1.

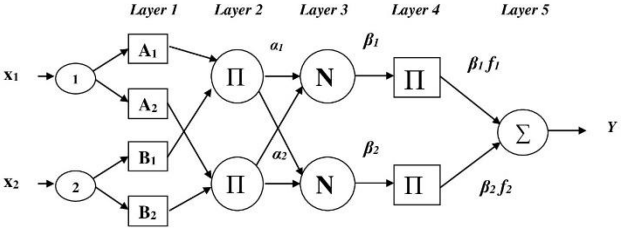


*Figure 1. ANFIS architecture with two fuzzy rules.*

A common two fuzzy if-then rules of the first-order Sugeno's type are as follows:

Rule 1: if $x_1 = A_1$ and $x_2 = B_1$ then $f_1(x_1, x_2) = a_{11}x_1 + a_{12}x_2 + b_1$

Rule 2: if $x_2 = A_2$ and $x_2 = B_2$ then $f_2(x_1, x_2) = a_{21}x_1 + a_{22}x_2 + b_2$

Layer 1: the primary purpose of the *f* layer is to map input variables into the fuzzy sets [18]. To represent the linguistic terms, the Gaussian member-

ship function is usually used [19]:

$$A_i(x) = \exp\left[-\frac{1}{2}\left(\frac{x-a_{i1}}{b_{i1}}\right)^2\right] \quad (1)$$

$$B_i(x) = \exp\left[-\frac{1}{2}\left(\frac{x-a_{i2}}{b_{i2}}\right)^2\right] \quad (2)$$

where, $a_{i1}$, $b_{i1}$, $a_{i2}$, and $b_{i2}$ are the parameter set.

Layer 2: the output of each node in this layer is the product of all the incoming signals:

$$\alpha_1 = A_1(x_1) \times B_1(x_2) \quad (3)$$

$$\alpha_2 = A_2(x_1) \times B_2(x_2) \quad (4)$$

Layer 3: the label N in this layer indicates the normalization of the firing levels. The outputs of two neurons are the normalized firing level:

$$\beta_1 = \frac{\alpha_1}{\alpha_1 + \alpha_2} \quad (5)$$

$$\beta_2 = \frac{\alpha_2}{\alpha_1 + \alpha_2} \quad (6)$$

Layer 4: The output of this layer is the product of the normalized firing level and the individual rule output:

$$\beta_1 f_1 = \beta_1(p_1 x_1 + q_1 x_2 + r_1) \quad (7)$$

$$\beta_2 f_2 = \beta_2(p_2 x_1 + q_2 x_2 + r_2) \quad (8)$$

Layer 5: The single node in this layer computes the overall system output as the sum of all incoming signals:

$$Y = \beta_1 f_1 + \beta_2 f_2 \quad (9)$$

*Solubility modeling using ANFIS-PSO*

The modeling of the phase behavior of $CO_2$ (1)-solid drugs (2) binaries is performed using the ANFIS approach with the PSO algorithm for the training. The ANFIS was built as five inputs (the equilibrium temperature, $T$, the equilibrium pressure, $P$) and three pure component properties to differentiate between the solubility of different solid drugs (critical temperature, $T_c$, critical pressure, $P_c$, and the acentric factor, $\omega$) and the solubility of the solid drugs in the SCF phase ($y_2$) as the output with five Gaussian membership functions for each input and five linear membership functions for the output. The first-order Sugeno fuzzy model was issued for generating the fuzzy rules. Figure 1 shows the ANFIS structure used in this study based on five lagged terms.

The experimental data used for developing the ANFIS-PSO model is divided into two sets: the first set is considered for training (it contains 66% of total data),

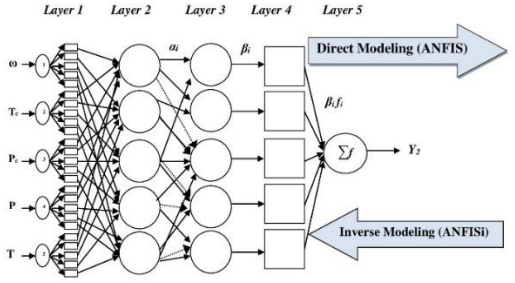


*Figure 2. Direct and Inverse ANFIS structure for the prediction of the solubility of solid drugs.*

*Table 1. Critical properties of solid components used in this work*

| Components | Tc (K) | Pc (MPa) | ω | Ref. |
|---|---|---|---|---|
| Naphthalene | 711.47[a] | 3.897[a] | 0.345 | This work |
| 2-Methoxybenzoic acid | 808.95[a] | 3.90[a] | 0.764[c] | This work |
| 3-Methoxybenzoic acid | 808.65[a] | 3.89[a] | 0.763[c] | This work |
| 4-Methoxybenzoic acid | 808.35[a] | 3.92[a] | 0.762[c] | This work |
| Cholesterol | 1151.55[a] | 1.11[a] | 0.967[c] | This work |
| Ibuprofen | 749.7 | 2.33 | 0.819 | [20] |
| 5-Fluorouracil | 807.42 | 6.22 | 0.64 | This work |
| Azodicarbonamide | 895.58 | 4.88 | 0.895 | This work |
| Thymidine | 924.21 | 3.64 | 0.886 | This work |
| 2-Phenyl-4H-1,3-benzoxazin-one | 1009.69 | 3.64 | 0.746 | This work |
| Naproxen | 807 | 2.45 | 0.904 | [20] |
| Taxol | 1023.03 | 1.01 | 1.33 | This work |
| Acetaminophen | 817.72 | 4.43 | 1.11 | This work |
| penicillin | 902.78 | 2.355 | 1.325 | [15] |
| m-nitrophenol | 896.35 | 5.53 | 0.662 | This work |
| p-nitrophenol | 896.35 | 5.53 | 0.662 | This work |
| 2,4-dinitrophenol | 914.1 | 4.91 | 0.827 | This work |
| 2,5-dinitrophenol | 914.1 | 4.91 | 0.827 | This work |
| Picricacid | 998.08 | 4.8 | 0.982 | This work |

and the second set is used for the test and validation (it has 34% of total data). This distribution is the most efficient for good ANFIS-PSO training. Table 1 shows the pure component properties of the solid components considered in this work. It is to mention that these properties for Naproxen and Ibuprofen are taken from the literature [20].

For solid drugs, where no parameters are available in the literature, the Lee-Kesler correlation was applied by using the PE software [21] to determine the Pitzer's acentric factor of solutes. The critical temperature and critical pressure were estimated by the Nannoolal method [22] and the Gani group contribution method [23].

The calculation strategy is based on the variation of the training algorithms (considered a parameter to optimize with other parameters of the ANFIS model (the topology of the ANFIS model, membership function, the number of rules). The optimization of the ANFIS model parameters is performed by minimizing the objective function, which is an average absolute relative deviation (AARD %) defined as:

$$OF = AARD(\%) = \frac{100}{N}\sum_{1}^{n}\frac{\left|y^{calc} - y^{\exp}\right|}{y^{\exp}} \quad (10)$$

## RESULTS AND DISCUSSION

The high nonlinearity is the characteristic of modeling the phase equilibrium. In this work, a comparative study was carried out between the new approach used in modeling solid solutes in $scCO_2$, i.e., the ANFIS trained with the PSO algorithm and the artificial neural network (ANN) trained with the PSO algorithm (trainpso), the Levenberg-Marquardt algorithm (trainlm), and the basien radial algorithm (trainbr).

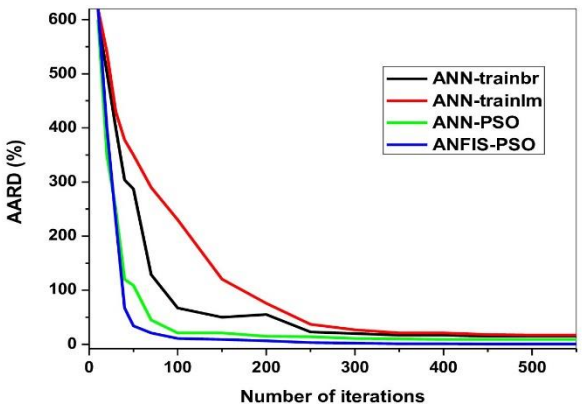


*Figure 3. Comparison in terms of AARD % between training algorithms for training ANFIS and ANN models.*

Figure 3 shows the superiority of the new approach in terms of the accuracy (lowest AARD) and the rapidity of the optimization process (lowest numbers of iterations) over the ANN model trained with the classical algorithms. Also, this study shows that the PSO training algorithm (trainpso) can be used for optimizing the ANFIS model with more accuracy than the other selected training algorithms (train-GA, trainlm, and trainbr).

Based on the study mentioned above, the ANFIS-PSO model has proved its ability to estimate the solubility of solids in $scCO_2$ with more advantages than the ANN method. Table 2 gives a detailed comparison of the AARD% between the solubility calculated by the ANFIS-PSO model and those estimated by the different models reported in the literature to show the superiority of the new approach proposed for estimating solid solubility. The comparison shows that the ANFIS-PSO model predicts the solubility of solid components in $scCO_2$ with more accuracy than those obtained by the EOS and density-based models (global AARD% = 0.99).

Figure 4 gives the solubility curves as a function of pressure for Taxol in $scCO_2$. This graphical comparison allows concluding that the proposed model is suitable for modeling and representing the solid-$scCO_2$ binary equilibrium. Also, this figure shows a good agreement between the experimental and the predicted solubility.

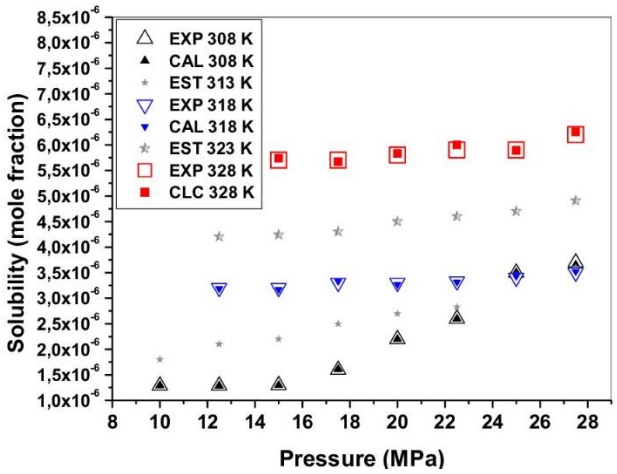


*Figure 4. Experimental solubility of taxol in $scCO_2$ and that predicted by ANFIS-PSO model at various temperatures.*

### Determination of importance of each input variable

To evaluate the effect of each input variable on the output variable, the relative importance (Ij) was calculated using the Garson expression [24]. Figure 5 gives the relative importance of five inputs where the equilibrium temperature and pressure substantially af-

*Table 2. Comparison of the AARD% of the predicted solubility of the solid drugs in $scCO_2$ obtained by ANFIS-PSO model and the literature results by some EOS and density-based models*

| SYSTEM | AARD% | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | ANFIS-PSO | Chrastil | VR-SAFT | Bartle | MT | PR | DVA | SRK |
| Naphthalene | 1.25 | NR | NR | NR | NR | 13.4 | NR | 14.8 |
| 2-Methoxybenzoic acid | 0.97 | 8.20 | NR | NR | NR | NR | NR | NR |
| 3-Methoxybenzoic acid | 1.13 | 2.67 | NR | NR | NR | NR | NR | NR |
| 4-Methoxybenzoic acid | 1.11 | 6.97 | NR | NR | NR | NR | NR | NR |
| Cholesterol | 1.05 | 4.37 | 10.40 | 6.3 | 3.44 | NR | 4.54 | NR |
| Ibuprofen | 1.23 | 6.72 | 8.80 | 8.9 | 3.99 | NR | 8.30 | NR |
| 5-Fluorouracil | 0.54 | 6.50 | NR | NR | NR | NR | NR | NR |
| Azodicarbonamide | 1.22 | 10.21 | NR | NR | NR | NR | NR | NR |
| Thymidine | 0.98 | 11.45 | NR | NR | NR | NR | NR | NR |
| 2-Phenyl-4H-1,3-benzoxazin-one | 1.00 | 5.50 | NR | NR | NR | NR | NR | NR |
| Naproxen | 0.40 | 3.50 | NR | NR | NR | 12.1 | NR | 11.5 |
| Taxol | 0.58 | 4.55 | NR | NR | NR | NR | NR | NR |
| Acetaminophen | 1.22 | 4.03 | NR | NR | NR | NR | NR | NR |
| Penicillin | 0.45 | 32.4 | NR | 22.9 | NR | NR | 32.4 | NR |
| m-nitrophenol | 1.40 | 9.42 | NR | NR | NR | NR | NR | NR |
| p-nitrophenol | 1.12 | 11.36 | NR | NR | NR | NR | NR | NR |
| 2,4-dinitrophenol | 0.40 | 13.40 | NR | NR | NR | NR | NR | NR |
| 2,5-dinitrophenol | 1.30 | 10.52 | NR | NR | NR | NR | NR | NR |
| Picric acid | 1.53 | 6.70 | NR | NR | NR | NR | NR | NR |
| Total | 0.99 | 8.80 | 9.60 | 12.7 | 3.71 | 12.75 | 15.08 | 13.15 |

[a] NR= Not Reported.

fect the solid solubility value with importance equal to 31% and 28%, respectively

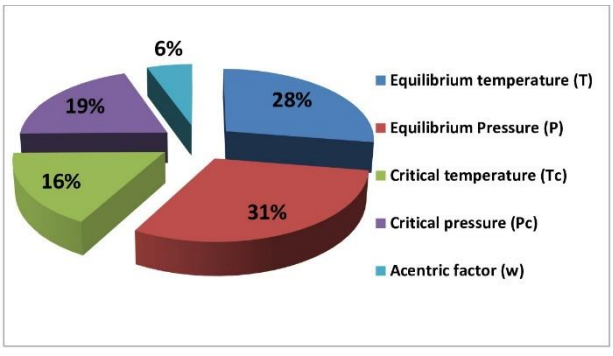


*Figure 5. Relative importance (%) of input variables on the value of the calculated solubility ($y_2$).*

## Optimal performance by mean of ANFISi

According to the ANFIS model, it is possible to simulate the system performance when the input parameters are well known, and the model parameters are well optimized. The mathematical formula that connects the selected input variables to the output (solid solubility) in the optimized ANFIS-PSO model is given as follow:

$$y_2 = \sum_{i=1}^{k} y_i(x)\left(a_i x_i + b_i\right) \tag{11}$$

with:

$$y_i(x) = \frac{\prod_{j=1}^{p} \exp\left(-\frac{(xj - cij)^2}{2\delta_{ij}^2}\right)}{\sum_{i=1}^{k}\prod_{j=1}^{p} \exp\left(-\frac{(x_j - c_{ij})^2}{2\delta_{ij}^2}\right)} \tag{12}$$

where, $p$ = 1.. .5, and $j$ = 1...5.

The INFISi approach is a method that can be used for estimating the input parameters from the output. Some of those parameters (critical properties) are not available in the literature, or there are no predictive methods to calculate them with acceptable accuracy. At

this step, the optimized ANFIS model provides the nonlinear equation:

$$Fun(x) = y_2 - \sum_{i=1}^{k} \frac{\prod_{j=1}^{p} \exp\left(-\frac{(x_j - c_{ij})^2}{2\delta_{ij}^2}\right)}{\sum_{i=1}^{k}\prod_{j=1}^{p} \exp\left(-\frac{(x_j - c_{ij})^2}{2\delta_{ij}^2}\right)} (a_i x_i + b_i) \quad (13)$$

This equation has to be minimized at zero to get the optimal input parameters where the optimization of equation (13) is classified as a constrained multivariable nonlinear optimization problem.

A set of the parameters are available for the inputs estimation process. When we introduce the value of the optimized ANFIS model, equilibrium temperature, and pressure, the other unknown parameters (critical temperature, critical pressure, and acentric factor) will be estimated using the optimization process.

Following the same strategy applied in section (4), the ability of major optimization algorithms was tested in estimating the ANFISI model, where a comparative study was carried out among a set of optimization algorithms.

The evaluation in terms of the AARD shows that the most efficient algorithm for the input parameters estimation is the PSO algorithm (Figure 6).

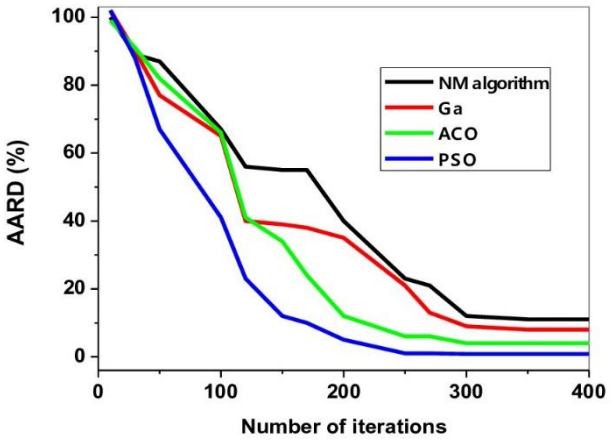


*Figure 6. Comparison in terms of AARD % between optimization algorithms (ACO, PSO, Ga and Nelder-Mead) for estimating input parameters using ANFISi.*

The comparison between the experimental and calculated values of the input parameters using ANFISi is performed in terms of $AARD_x$:

$$AARD_x(\%) = \frac{100}{n} \frac{\sum_{i}^{n} \left(x_i^{calc} - x_i^{\exp}\right)}{x_i^{\exp}} \quad (14)$$

**Estimation of critical properties by the interpolating method**

The ANFISI approach developed has reproduced the critical properties from the experimental data with good precision (the AARD (%) calculated are 0.85, 0.75, and 0.60 for the critical temperature, critical pressure, and acentric factor, respectively).

In this part, an extrapolating test is carried out to predict the acentric factor, critical temperature, and pressure from the experimental solubility data of the solid components found in the literature, which were not used in the development of the ANFIS model. Because this work has a point to evaluate the promising heuristic techniques' ability to represent the phase equilibria reliability, the ANFISi was compared to the ANNi method developed previously in terms of the average relative deviation (AARD%) for estimating the properties of the pure components. Figure 7 shows that the ANFISi can estimate both critical temperature and pressure far away than the ANNi.

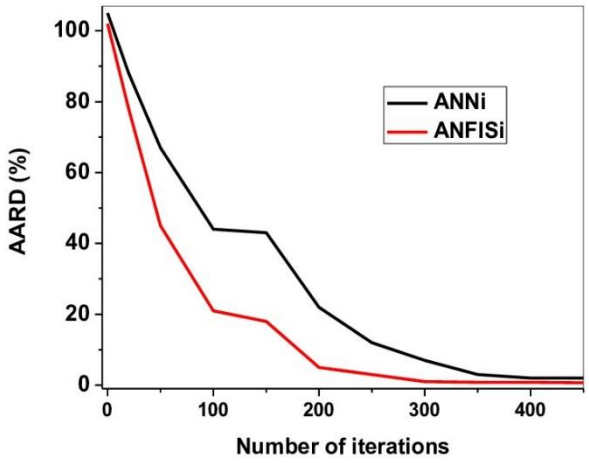


*Figure 7. Comparison in terms of AARD between ANNi and ANFISi methods for estimation critical pressure of solid solutes.*

Using the PSO algorithm for the multivariable optimization process, the estimated values of the properties mentioned above for other classes of solids are shown in Table 3. This contains a comparison between the estimated properties with the new method and those calculated with the classical GCM method found in the literature [25-38]. Table 3 shows that the critical properties and the acentric factor calculated with the inverse ANFIS approach have the same orders of magnitude, which suggests a good extrapolating ability of the ANFISi. In addition, this approach is more advantageous than the group contribution methods, which needed the ebullition temperature and the structure of molecules for calculating such properties. The complicated process used with these methods may

add a factor of inaccuracy for the calculated critical properties [39].

Table 4 gives the AARD (%) values evaluated between the experimental data and the solubility calcu lated by the PR, SRK, and Pazuki equations of state using critical properties estimated in this work using the ANFISi and those calculated with the same equations of state reported in the literature.

*Table 3. Comparison between critical properties estimated by ANFISi approach and those found in literature*

| Component | Critical properties | | | | | | Ref. |
|---|---|---|---|---|---|---|---|
| | This work | | | Literature | | | |
| | $T_c$ | $p_c$ | $w$ | $T_c$ | $p_c$ | $w$ | |
| Amiodarone hydrochloride | 989.6 | 10.50 | 0.41 | 1040.4 | 11.75 | 0.430 | [25] |
| Curcumin | 432.5 | 22.20 | 1.35 | 419.9 | 22.50 | 1.551 | [26] |
| Anthraquinone | 977.3 | 31.76 | 1.03 | 987.05 | 31.28 | 1.015 | [27] |
| 1,4 bisethylamino (Anthraquinone) | 955.2 | 22.40 | 1.11 | 945.34 | 21.03 | 1.142 | [27] |
| 1-Amino4-hydroxyanthraquinone | 933.2 | 33.20 | 0.93 | 921.01 | 30.68 | 0.982 | [27] |
| 1-Hydroxy 4-nitro anthraquinone | 919.7 | 28.5 | 1.01 | 913.95 | 27.74 | 1.046 | [27] |
| 1-Amino anthraquinone | 901.2 | 30.33 | 0.85 | 928.10 | 31.42 | 0.853 | [28] |
| 1-nitro anthraquinone | 933.3 | 28.40 | 0.90 | 916.60 | 28.10 | 0.921 | [28] |
| Photochomicdye | 785.1 | 24.20 | 0.89 | - | - | - | [29] |
| Ibuprofen | 763.5 | 22.90 | 0.89 | 749.70 | 23.30 | 0.819 | [30] |
| Ferulic acid | 846.5 | 36.78 | 1.10 | 854.60 | 36.40 | 1.194 | [31] |
| Pyridin 4-amine | 887.2 | 23.50 | 0.95 | - | - | - | [32] |
| Fluvaxamine maleate | 747.3 | 55.20 | 0.41 | - | - | - | [33] |
| Flavanone | 867.8 | 32.57 | 0.74 | 879.90 | 32.80 | 0.728 | [34] |
| Tangeritin | 1034.2 | 26.89 | 0.98 | 1139.1 | 26.66 | 1.107 | [34] |
| Nobiletin | 1178.4 | 25.94 | 1.09 | 1256 | 26.60 | 1.228 | [34] |
| 6-Hydroxyflavanone | 998.5 | 33.50 | 0.88 | 1062.5 | 35.40 | 0.914 | [34] |
| 7-Hydroxyflavanone | 1000.1 | 37.59 | 0.98 | 1074.2 | 37.81 | 0.952 | [34] |
| Protocatechuic | 861.3 | 54.50 | 0.89 | 869.29 | 55.33 | 0.984 | [35] |
| Crysin | 954.7 | 30.20 | 0.98 | 966.90 | 31.15 | 1.175 | [35] |
| Sinapic acid | 904.5 | 27.80 | 0.91 | 927.62 | 28.06 | 0.973 | [35] |
| Menadione | 624.4 | 45.88 | 0.69 | 639.58 | 46.53 | 0.623 | [36] |
| Dichlone | 744.2 | 65.04 | 0.67 | 731.96 | 64.10 | 0.639 | [36] |
| Aprepitant | 888.9 | 21.50 | 0.90 | 895.60 | 20.18 | 0.810 | [37] |
| A-Tocopherol | 888.5 | 45.30 | 0.525 | - | - | - | [37] |
| dM2B | 989.9 | 23.45 | 0.88 | 1144.11 | 24.46 | 1.015 | [38] |
| dM3P | 995.4 | 21.9 | 1.10 | 1161.46 | 22.29 | 1.051 | [38] |

*Table 4. AARD evaluated between experimental solubility and that calculated with different models*

| Component | AARD (%)[a] | | | AARD (%)[b] | | |
|---|---|---|---|---|---|---|
| | PR-EOS | SRK-EOS | Paz-EOS | PR-EOS | SRK-EOS | Paz-EOS |
| Amidarone | 4.20 | 4.35 | 3.46 | 6.03 | NR | 8.88 |

*Table 4. AARD evaluated between experimental solubility and that calculated with different models (Continued)*

| Component | AARD (%)[a] | | | AARD (%)[b] | | |
|---|---|---|---|---|---|---|
| | PR-EOS | SRK-EOS | Paz-EOS | PR-EOS | SRK-EOS | Paz-EOS |
| Curcumin | 7.33 | 7.87 | 5.34 | 50.5 | NR | NR |
| Anthraquinone | 5.25 | 5.97 | 3.67 | 7.60 | 8.40 | NR |
| 1,4 bisethylelim | 3.44 | 4.42 | 5.60 | 13.10 | NR | NR |
| Photochomicdye | 6.50 | 7.39 | 5.33 | 12.30 | 13.1 | NR |
| Ibuprofen | 7.29 | 6.50 | 3.30 | 9.70 | 9.95 | NR |
| Ferulic acid | 8.11 | 7.14 | 6.47 | 50.9 | NR | NR |
| Pyridin 4-amine | 5.55 | 4.67 | 4.95 | NR | NR | NR |
| Fluvaxamine maleate | 5.30 | 5.02 | 6.11 | 63.43 | NR | NR |
| Flavanone | 0.99 | 1.33 | 1.56 | 1.90 | NR | NR |
| Tangeritin | 1.32 | 5.44 | 4.65 | 1.77 | 9.50 | NR |
| Nobiletin | 1.20 | 1.01 | 3.45 | 0.98 | NR | NR |
| 6-Hydroxyflavanone | 2.11 | 1.32 | 1.22 | 1.72 | NR | NR |
| 7-Hydroxyflavanone | 3.77 | 2.25 | 3.34 | 1.14 | NR | NR |
| Protocatechuic | 5.70 | 5.93 | 2.12 | 5.60 | NR | NR |
| Crysin | 2.00 | 1.87 | 2.22 | 2.30 | NR | NR |
| Sinapic acid | 6.80 | 4.41 | 3.78 | 18.50 | NR | NR |
| Menadione | 4.33 | 3.89 | 5.42 | 8.27 | NR | NR |
| Dichlone | 4.20 | 4.43 | 6.43 | 9.03 | NR | NR |
| Aprepitant | 5.43 | 6.16 | 6.66 | 9.08 | 9.23 | NR |
| A-Tocopherol | 5.55 | 4.78 | 5.09 | NR | NR | NR |
| 1-Amino4-hydroxyanthraquinone | 6.73 | 7.88 | 5.44 | 13.60 | NR | NR |
| 1-Hydroxy4-nitro anthraquinone | 9.88 | 8.55 | 5.54 | 8.20 | NR | NR |
| 1-Amino anthraquinone | 3.45 | 4.44 | 6.12 | 10.50 | NR | NR |
| 1-nitro anthraquinone | 3.23 | 3.87 | 4.44 | 13.10 | NR | NR |
| dM2B | 5.21 | 4.98 | 3.77 | 9.85 | NR | NR |
| dM3P | 3.85 | 3.44 | 2.43 | 7.33 | NR | NR |

[a] AARD (%) evaluated between experimental data and solubility calculated by PR, SRK and Pazuki equations of state using critical properties estimated in this work (ANFISi). [b] AARD (%) evaluated between experimental data and solubility calculated by PR, SRK and Pazuki equations of state reported in the literature. NR= Not Reported.

## CONCLUSION

In this work, a new method that combines ANN and FIS has been used for developing a model to predict the solubility of solids in $scCO_2$. The estimation results show that the ANFIS-PSO model can predict the solid solubility far better than the ANN-PSO model and the classical models with the AARD value equal to 0.99 %. Also, the comparative study shows that the PSO algorithm was more advantageous in training, test, validation, and optimization problems. Therefore, the new approach shows the high predictive and interpolating abilities at temperatures where no experimental data was found in the literature.

The second valuable contribution was developing an efficient method for reproducing the input parameters by an inverse FIS. The ANFISi was used first to reproduce the critical properties of the solids used for developing the ANFIS-PSO model to test the validity of this approach and then estimate the critical parameters for another set of solid drugs (extrapolation test). The results show that the ANFISi method can be a promising technique and a good alternative to the GCM method in estimating the critical properties of solid drugs. Also, ANFISi has facilitated the perfor-

mance of estimating the solubility of the solid using the EOS equations compared to the experimental data.

## List of symbols

| | |
|---|---|
| AARD | Average absolute relative deviation |
| ACO | Ant colony optimization |
| AI | Artificial intelligence |
| ANN | Artificial neural network |
| ANNi | Inverse artificial neural network |
| ANFIS | Adaptive neuro-fuzzy inference system |
| ANFISi | |
| $a_{ij}$, $b_{ij}$ | Gaussian membership function parameters |
| br | Bazian regularization |
| DVA | Del Var Aguilera equation of state |
| FIS | Fuzzy interference system |
| I | Relative importance |
| GA | Genetic algorithm |
| GCM | Group contribution method |
| lm | Levenberg_Marquardt |
| NM | Nelder_Mead optimization algorithm |
| T | Equilibrium temperature (K) |
| $T_c$ | Critical temperature (K) |
| P | Pressure (MPa) |
| $P_c$ | Critical pressure (MPa) |
| PE | Phase equilibria |
| PR | Peng_Robinson |
| PSO | Particle swarm optimization |
| SRK | Soave_Redlich_Kwong |
| VR-SAFT | Variable ranged statistical associating fluid theory |
| α | Firing strength |
| β | Normalized firing level |
| ω | Acentric factor |
| $y_2$ | Solubility of solid drugs |

**Superscripts**

| | |
|---|---|
| calc | Calculated property |
| exp | Experimental property |
| o | Output |

**Subscripts**

| | |
|---|---|
| 2 | Solute (solid) |
| c | critical property |

## REFERENCES


[1] J.C. Rojas-Thomas, M. Mora, S. Santos, Neural Comput. Appl.31 (2019) 2311-2327.

[2] S.K. Ashan, M.A. Behnajady, N. Ziaeifar, Neural Comput. Appl. 29 (2018) 969-979.

[3] M. Khayet, C. Cojocaru, Desalination 308 (2013) 102-110.

[4] B. Gülçin, G. Sezin, Energy 123 (2017) 149-163.

[5] M. Laidi, S. Hanini, Rezrazi A, M.R. Yaiche, A. Abdallah el Hadj, F. Chellali, Theor. Appl. Climatol. 128 (2017) 439-451.

[6] A. Abdallah El Hadj, C. Si-Moussa, S. Hanini, M. Laidi, Chem. Ind. Chem. Eng. Q. 19 (2013) 449-460.

[7] M. Velibor, Chem. Ind. Chem. Eng. Q. 26 (2020)309–319.

[8] Y. Jewajinda, P. Chongstitvatana, Neural Comput. Appl. 22 (2013)1609–1626.

[9] R. Fuller, H.J. Zimmermann, in Proceedings of 2nd International Workshop on Current Issues in Fuzzy Technologies, University of Trento, Trento, May 28-30 (1993) 45-54.

[10] A. Abdallah El Hadj, M. Laidi, C. Simoussa, S. Hanini, Neural Comput Appl. 28 (2017) 87-99.

[11] J.W. Chen, F.N. Tsai, Fluid Phase. Equilib. 107 (1995) 189-200.

[12] F.E. Wubbolts, O.S.L. Bruinsma, G.M. Van Rosmalen, J. Supercrit. Fluids 32 (2004) 79-87.

[13] M. Shammsipur, F. Reza, Y. Yamini, A.R. Ghiasvand, J. Supercrit. Fluids 23 (2002) 225-231.

[14] S. David, L.A. Estévez, J.C. Pulido, J.E. Garcia, M. Carmen, J. Chem. Eng. Data 50 (2005) 1234-1241.

[15] M.D. Gordillo, M.A. Blanco, A. Molero, E. Martinez de la Ossa, J. Supercrit. Fluids 15 (1999) 183-190.

[16] J.S.R. Jang, IEEE Trans. Syst. Man. Cybern. 23 (1993) 665-685.

[17] A.R. Fallahpour, A.R. Moghassem, J. Eng. Fibers Fabr. 8 (2013) 6-18.

[18] R. Kamali, A.R. Binesh, Microfluid. Nanofluid. 14 (2013) 575-581.

[19] R.Babuska, Neuro-Fuzzy Methods for Modeling, In Recent Advances in Intelligent Paradigms and Applications, A. Abraham, L.C. Jain, J. Kacprzyk, Springer-Verlag, Heidelberg (2002), pp 161-186.

[20] P. Coimbra, C.M. Duarte, H.C. de Sousa, Fluid Phase Equilib. 239 (2006) 188-199.

[21] O. Pfohl, S. Petkov, G. Brunner, High-pressure fluid-phase equilibria containing supercritical fluids, In 8th International Conference on properties and Phase Equilibria for Product and Process Design, Noordwijkerhout, Netherlands, April 26-May (1998).

[22] Y. Nannoolal, J. Rarey, D. Ramjugernath, Fluid Phase Equilib. 269 (2008) 117-133.

[23] J. Marrero, R. Gani, Fluid Phase Equilib. (183-184) (2001) 183-208.

[24] G.D. Garson, Interpreting Neural Network Connections weights, AI Expert: Miller Freeman, Inc. San Francisco (1991), p. 46.

[25] G. Sodeifian, S.A. Sajadian, F. Razmimanesh, Fluid Phase Equilib. 25 (2017) 149-159.

[26] P.C. Larissa, M.C. Acosta, C. Turner, J. Supercrit. Fluids

130 (2017) 381-388.

[27] K. Tamura, R.S. Alwi, Dyes Pigm. 113 (2015) 351-356.

[28] R.S. Alwi, T. Tanaka, K. Tamura, J. Chem. Thermodyn. 74 (2014) 119-125.

[29] P. Coimbra, M.H. Gil, C.M.M. Duarte, B.M Heron, H.C. de Sousa, Fluid Phase Equilib. 238 (2005) 120-128.

[30] A. Mehdi, M. Mehrdad, Z. Fatemeh, Chin. J. Chem. Eng. 22 (2014) 549-558.

[31] G.R. Bitencourt, F.A. Cabral, A. Meirelles, J. Chem. Thermodyn. 103 (2016) 285-291.

[32] C. Chun-Ta, L. Chen-An, T. Muoi, Y.P. Chen, J. $CO_2$ Util. 18 (2017) 173-180.

[33] Y. Khayyat, S.M. Kashkouli, F. Esmaeilzadeh, Fluid Phase Equilib. 399 (2015) 98-104.

[34] M. Ota, M. Sato, Y. Sato, L.S.J. Richard, H. Inomata, J. Supercrit. Fluids 128 (2017) 166-172.

[35] J.P. Paulaa, I.M.O. Sousab, M. Foglioc, F. Cabral, J. Supercrit. Fluids 112 (2016) 89-94.

[36] A.G. Reveco-Chilla, A.L. Cabrera, J.C. de la Fuente, F.C. Zacconi, J.M. del Valle, L.M. Valenzuela, Fluid Phase Equilib.42 (2016) 84-92.

[37] G. Sodeifian, S. Sajadian, N.S. Ardestani, J. Supercrit. Fluids128 (2017) 102-111.

[38] F.C. Zacconi, O.N. Nuñez, A.L. Cabrera, L.M. Valenzuela, J.M. del Valle, J.C. de la Fuente, J. Chem. Thermodyn. 103 (2016) 325-332.

[39] E. Potrich, F.A.P. Voll, V.F. Cabral, L. Cardozo Filho, Chem. Ind. Chem. Eng. Q. 25 (2019) 153–162.

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NAUČNI RAD

# NOVA METODA ZASNOVANA NA NEURO-FAZI SISTEMU I PSO ALGORITMU ZA PROCENU KARAKTERISTIKA FAZNE RAVNOTEŽE

*Predmet ovog rada je predlaganje nove metode zasnovane na ANFIS sistemu i PSO algoritmu za osmišljavanje modela za procenu rastvorljivosti čvrstih lekova u natkritičnom $CO_2$. Visoki nelinearni proces je modelovan neuro-fazi pristupom (NFS). PSO algoritam je korišćen u dve svrhe: za zamenu standardne propagacije unazad u obuci NFS-a i optimizacija procesa. Strategija validacije je sprovedena korišćenjem analize linearne regresije i upoređenjem predviđenih sa eksperimentalnim podacima. ANFIS pristup je upoređivan sa ANN u smislu tačnosti. Statistička analiza predvidljivosti optimizovanog modela obučenog PSO algoritmom (ANFIS-PSO) pokazuje bolje slaganje sa referentnim podacima od ANN metode. Štaviše, poređenje u smislu AARD devijacije (%) između predviđenih rezultata, rezultata predviđenih modelima zasnovanim na gustini i skupa jednačina stanja pokazuje da ANFIS-PSO model daleko bolje korelira rastvorljivost čvrstih lekova u natkritičnom $CO_2$. Takođe, po prvi put je razvijena kontrolna strategija u oblasti fazne ravnoteže korišćenjem neuro-fazi inverznog pristupa (ANFISi) za procenu svojstava čistih komponenti iz podataka o rastvorljivosti bez prolaska kroz GCM metode.*

*Ključne reči: modeliovanje, ANFIS, veštačke neuronske mreže, kritična svojstva, optimizacija rojem čestica.*