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Research Article

Solubility Optimal System for Supercritical Fluid Extraction Based on a New Nonlinear Temperature-Pressure Decoupling Model Constructed with Unequal-Interval Grey Optimal Models and Peng-Robinson Models

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This paper presents a new solubility optimal system to improve the efficiency of supercritical fluid extraction (SFE). The major contribution is a nonlinear temperature-pressure decoupling model constructed with unequal-interval grey optimal models (UEIGOMs) and Peng-Robinson models (PRMs). The linear parts of temperature and pressure process can be constructed with UEIGOM, respectively. The nonlinear parts of temperature and pressure process can be described by PRMs, respectively. The whole nonlinear model cannot be input-output decoupled resulting from the singularity of decoupling matrix for PRM. This problem on input-output nondecoupling can be transformed to the problem on disturbance decoupling for a class of MIMO nonlinear systems. Therefore, the whole nonlinear coupling model can be disturbance decoupled. Furthermore, solubility optimal method is presented in the paper; it can calculate the optimal pressure according to the given temperature, namely, optimal working points, to maximize solubility for SFE process. The feasibility, effectiveness, and practicality of the proposed nonlinear temperature-pressure decoupling model constructed with UEIGOMs and PRMs are verified by SFE experiments in biphenyl. Experiments using the designed solubility optimal system are carried out to demonstrate the effectiveness in control scheme, simplicity in structure, and flexibility in implementation for the proposed solubility optimal system based on a new nonlinear temperature-pressure coupling model constructed with UEIGOMs and PRMs.

1. Introduction

Extraction of a material using a supercritical fluid is called supercritical fluid extraction (SFE). SFE, which is a contamination-free extraction technology in food science and chemical industry, is of central importance in biomaterial processing. During the separation process, the solvency of SFE can be modified by adjusting temperature, pressure, moisture contents, and so on [1–4]. Temperature and pressure play a crucial role in SFE process. The model of temperature and pressure process, which is nonlinear, is composed of linear and nonlinear parts. The linear part, which can be obtained easily, is SISO model of temperature or pressure. The nonlinear part is the coupling relationship between temperature and pressure. Numerous methods and models have been proposed to describe SFE process. Cubic equation of state (EoS) with simplified inner structure and generalized form is one of the most widely used models to describe the temperature, pressure, and time behaviors for fluid. vdW EoS, which is used in calculation of vapor-liquid equilibrium, was proposed by van der Waals in 1873 [5]. RK EoS was proposed by Redlich and Kwong in 1949 [6]. RK EoS was improved by Soave, and RKS EoS was proposed in 1979 [7]. RP EoS was proposed by Peng and Robinson in 1976 [8]. PR EoS is widely used and contrasted with the other three models. A hybrid model, which is constructed with a radial basis function (RBF) model and RP EoS, was proposed to keep all the physical information in PR model and optimize the binary interaction parameter in the PR model [9, 10]. Combined with operating cost, safety index, and yield rate of extraction calculated by hybrid model, an optimal control system can be designed. However, temperature has a momentous effect on pressure, in temperature and pressure control process, and vice versa. The coupling relationship between temperature and pressure in SFE process is not considered in the proposed optimal control system. The performance of temperature and pressure control has an effect on yield rate and solubility of extraction. Therefore, combining with PR EoS, study on modeling of temperature and pressure coupling and decoupling model has great significance with new theories and methods for improving SFE work efficiency to obtain maximal yield rate and solubility of extraction.

In this work, the linear parts of temperature and pressure models can be described through UEIGOMs with grey technology, respectively. The nonlinear parts of temperature and pressure models are modeled with PR EoS. Discussing the decoupling conditions of input-output of temperature-pressure nonlinear model, the decoupling system is given through state and output transforms. Furthermore, solubility optimal method is presented in the paper; it can calculate the optimal pressure according to the given temperature, namely, optimal working points, to maximize solubility for SFE process. The rest of the paper is organized as follows: In Section 2, temperature-pressure process and solubility modeling are discussed. In Section 3, temperature-pressure decoupling control system and solubility optimal system are designed. Computer simulation results of UEIGOMs of temperature and pressure process, temperature-pressure decoupling control, and solubility optimal system are presented and discussed, respectively, in Section 4. Finally, a conclusion regarding research and future works is made in Section 5.

2. Temperature-Pressure Process and Solubility Modeling

Due to the reaction character of SFE process, the nonlinear strong coupling relationship between temperature and pressure is the main factor affecting the efficiency of SFE. In order to improve the performance of SFE control system effectively, it is necessary to model temperature-pressure process. In this work, the temperature and the pressure processes can be modeled by grey technology, respectively, and their UEIGOMs can be given; the coupling parts can be obtained by PRMs.

2.1. UEIGOM for Temperature-Pressure Process. Grey system theory is based on fewer samples, which are made of some known and unknown information. It can work on extracting the valuable information from the fewer samples. Grey generating technology can provide intermediate information and weaken the randomness of original data; therefore, the model using grey generating technology can give the correct description of the reaction character of SFE process. In grey calculation, accumulated generating operation (AGO) and inverse accumulated generating operation (IAGO) are utilized [11–13]. They can be defined as follows.

Let $\mathbf{x}^{(0)} \in \mathbf{R}^{1\times n}$ be original series and $\mathbf{x}^{(r)} \in \mathbf{R}^{1\times n}$ be r-AGO series, if there is an accumulated generated matrix $\mathbf{A}_1 \in \mathbf{R}^{n\times n}$ that satisfies (1); let $\mathbf{x}^{(r)} \in \mathbf{R}^{1\times n}$ be r-IAGO series, if there is an inverse accumulated generated matrix $\mathbf{A}_2 \in \mathbf{R}^{n\times n}$ that satisfies (2).

$$\mathbf{x}^{(r)} = \mathbf{x}^{(r-1)}\mathbf{A}_1, \tag{1}$$

$$\mathbf{x}^{(r-1)} = \mathbf{x}^{(r)}\mathbf{A}_2, \tag{2}$$

where

$$\begin{gathered}
\mathbf{x}^{(0)} = \left[x^{(0)}(1), x^{(0)}(2), \dots, x^{(0)}(n)\right]; \\
\mathbf{x}^{(r)} = \left[x^{(r)}(1), x^{(r)}(2), \dots, x^{(r)}(n)\right]; \\
\mathbf{A}_1 = \begin{bmatrix} 1 & 1 & \cdots & 1 \\ 0 & 1 & \cdots & 1 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & 1 \end{bmatrix}; \\
\mathbf{A}_2 = \begin{bmatrix} 1 & -1 & 0 & \cdots & 0 \\ 0 & 1 & -1 & \cdots & 0 \\ 0 & 0 & 1 & \cdots & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & 0 & \cdots & 1 \end{bmatrix}.
\end{gathered} \tag{3}$$

2.1.1. Transforming from Unequal-Interval Series to Equal-Interval Series. AGO and IAGO operations can all aim at equal-interval series; however, actual process signals are always unequal-interval series. Therefore, it is necessary to transform to equal-interval series before AGO or IAGO operation.

Let $\mathbf{x}_1^{(0)} = [x_1^{(0)}(t_1), x_1^{(0)}(t_2), \dots, x_1^{(0)}(t_n)]$, $i = 1, 2, \dots, n$, be an unequal-interval series; then its average interval can be calculated as follows:

$$\Delta t = \frac{\sum_{i=2}^{n} \Delta t_i}{n-1} = \frac{t_n - t_2}{n-1}, \quad i = 2, 3, \dots, n. \tag{4}$$

The coefficient between every interval and average interval is given.

$$\mu(t_i) = \frac{t_i - (i-1)\Delta t}{\Delta t}, \quad i = 2, 3, \dots, n. \tag{5}$$

The total difference value of every interval can be calculated.

$$\Delta x_1^{(0)}(t_i) = \mu(t_i)\left[x_1^{(0)}(t_i) - x_1^{(0)}(t_{i-1})\right], \quad i = 2, 3, \dots, n. \tag{6}$$

Above all, the equal-interval series $\overline{\mathbf{x}}_2^{(0)} = [\overline{x}_2^{(0)}(1), \overline{x}_2^{(0)}(2), \dots, \overline{x}_2^{(0)}(i)]$, $i = 1, 2, \dots, n$, can be obtained, where

$$\begin{gathered}
\overline{x}_2^{(0)}(1) = x_1^{(0)}(t_1), \quad i = 1, \\
\overline{x}_2^{(0)}(i) = x_1^{(0)}(t_i) - \Delta x_1^{(0)}(t_i), \quad i = 2, 3, \dots, n.
\end{gathered} \tag{7}$$

2.1.2. Grey Optimal Model. Series $\overline{\mathbf{x}}_2^{(1)}$ is obtained by 1-AGO with series $\overline{\mathbf{x}}_2^{(0)}$; namely, $\overline{\mathbf{x}}_2^{(1)} = \overline{\mathbf{x}}_2^{(0)}\mathbf{A}_1$. Define $\mathbf{Z}^{(1)} = [z^{(1)}(2), z^{(1)}(3), \ldots, z^{(1)}(n)]$ as the black ground value of series $\overline{\mathbf{x}}_2^{(1)}$, where $z^{(1)}(k) = \alpha\overline{x}_2^{(1)}(k-1) + (1-\alpha)\overline{x}_2^{(1)}(k)$, $k = 2, 3, \ldots, n$. Generally, $\alpha = 0.5$; then differential equation can be described as

$$\frac{d\overline{\mathbf{x}}_2^{(1)}}{dt} + a\overline{\mathbf{x}}_2^{(1)} = b, \tag{8}$$

where a is developing coefficient and b is grey input. Utilizing least square method, a and b can be obtained:

$$\widehat{\mathbf{a}} = \begin{bmatrix} a \\ b \end{bmatrix} = \left(\mathbf{B}^T\mathbf{B}\right)^{-1}\mathbf{B}^T\mathbf{y}_N, \tag{9}$$

where

$$\mathbf{B} = \begin{bmatrix} -z^{(1)}(2) & 1 \\ -z^{(1)}(3) & 1 \\ \vdots & \vdots \\ -z^{(1)}(n) & 1 \end{bmatrix}; \qquad \mathbf{y}_N = \begin{bmatrix} \overline{x}_2^{(0)}(2) \\ \overline{x}_2^{(0)}(3) \\ \vdots \\ \overline{x}_2^{(0)}(n) \end{bmatrix}. \tag{10}$$

Time response function of GOM can be expressed as follows:

$$\begin{aligned} \widehat{\overline{x}}_2^{(1)}(k+1) &= \left(\overline{x}_2^{(0)}(1) - \frac{b}{a} - c\right)e^{-ak} + \frac{b}{a} + c, \\ \widehat{\overline{x}}_2^{(0)}(k+1) &= \widehat{\overline{x}}_2^{(1)}(k+1) - \widehat{\overline{x}}_2^{(1)}(k), \end{aligned} \tag{11}$$

where c is translation value.

2.1.3. Reversion from Equal-Interval Series to Unequal-Interval Series. Considering $k+1 = t_i/\Delta t$, (11) can be transformed for unequal-interval series, namely, UEIGOM:

$$\begin{aligned} \widehat{x}_1^{(1)}(t) &= \left(x_1^{(0)}(t_1) - \frac{b}{a} - c\right)e^{-a(t/\Delta t)} + \frac{b}{a} + c, \\ \widehat{x}_1^{(0)}(t) &= \widehat{x}_1^{(1)}(t) - \widehat{x}_1^{(1)}(t - \Delta t), \end{aligned} \tag{12}$$

where $c = ((e^a + 1)/(1 - e^{-2(n-1)a}))\sum_{i=1}^{n-1} q^{(0)}(t_{i+1})e^{-a(t_i/\Delta t)}$; $q^{(0)}(t_i) = x_1^{(0)}(t_i) - \widehat{x}_1^{(0)}(t_i)$ is residual error. Therefore, the temperature and the pressure processes can be described as first-order inertia system by UEIGOM, respectively.

2.2. PRM Modeling. Peng-Robinson model is the most commonly exploited model for treating solubility in SFE. The nonlinear strong coupling relationship between temperature and pressure is reflected in the Peng-Robinson equation of state. Therefore, the coupling parts of temperature-pressure process can be embodied by PRM in this work. PRM is given as

$$p = \frac{RT}{V - d_1} - \frac{\Omega_{11}(T)}{V^2 + 2d_1V - {d_1}^2}, \tag{13}$$

where R is gas constant, T is absolute temperature, V is the molar volume of pure solvent, $\Omega_{11}(T)$ is the parameter describing attractive interactions between molecules, T_c is critical temperature, p_c is critical pressure, ω is acentric factor, T_b is normal boiling point, and d_1 is the parameter describing volume exclusion and repulsive interactions. The parameters in PR model are listed in Table 1. Subscript 1 represents the solvent and subscript 2 represents the solute. Considering that the molar volume of pure solvent is invariable, (13) can be simplified as follows:

$$p = \eta_1 T + \eta_2 T^{1/2} + \eta_3, \tag{14}$$

where

$$\begin{aligned} \eta_1 &= \frac{p_c R\left(V^2 + 2d_1V - {d_1}^2\right) - 0.457235R^2T_c\left(V - d_1\right)f^2(\omega_1)}{p_c\left(V^2 + 2d_1V - {d_1}^2\right)\left(V - d_1\right)}; \\ \eta_2 &= \frac{0.457235R^2T_c^{3/2}\left[2f^2(\omega_1) + 2f(\omega_1)\right]}{p_c\left(V^2 + 2d_1V - {d_1}^2\right)}; \\ \eta_3 &= -\frac{0.457235R^2T_c^2\left[1 + f(\omega_1)\right]^2}{p_c\left(V^2 + 2d_1V - {d_1}^2\right)}. \end{aligned} \tag{15}$$

Furthermore, the inverse function of (14) can be obtained as follows:

$$T = g^{-1}(T). \tag{16}$$

Combining UEIGOM with (14) and (16), let $x_1 = T$ and $x_2 = p$ be state variables; the temperature-pressure process model for CO_2 can be described as follows:

$$\begin{aligned} \dot{x}_1 &= a_1x_1 + b_1u_1, \\ \dot{x}_2 &= a_2x_2 + b_2u_2, \\ y_1 &= x_1 + p_1(x_2), \\ y_2 &= x_2 + p_2(x_1), \end{aligned} \tag{17}$$

where $\mathbf{x} \in \mathbf{R}^2$, $\mathbf{u} \in \mathbf{R}^2$, $\mathbf{y} \in \mathbf{R}^2$, $\mathbf{p}(\mathbf{x}) = \left[(\zeta_1\sqrt{\zeta_2x_2 + \zeta_3} + \zeta_4)^2 \quad \eta_1x_1 + \eta_2{x_1}^{1/2} + \eta_3\right]^T$, and $a_1a_2b_1b_2\eta_1\eta_2\eta_3\zeta_1\zeta_2\zeta_3\zeta_4 \neq 0$, and they are system constants; $\mathbf{x}_0 = [0 \quad 0]^T$; $\mathbf{p}_0 = \left[(\zeta_1\sqrt{\zeta_3} + \zeta_4)^2 \quad \eta_3\right]^T$. This temperature-pressure nonlinear model cannot input-output decoupled result from the singularity of decoupling matrix. Namely, input-output decoupling conditions are unsatisfied [14, 15].

Table 1: Parameters in PR model.

Parameters	Values of pure component	Values of mixture component
$\Omega(T)$	$\Omega_{11}(T) = \dfrac{0.457235R^2T_{c,1}^2}{p_{c,1}}\left[1+f(\omega_1)\left(1-T_{r,1}^{1/2}\right)\right]^2$	$\Omega_{12}(T) = \dfrac{0.457235R^2T_{c,1}T_{c,2}}{2\sqrt{p_{c,1}p_{c,2}}}\left[1+f(\omega_1)\left(1-T_{r,1}^{1/2}\right)\right]\left[1+f(\omega_2)\left(1-T_{r,2}^{1/2}\right)\right]$
$f(\omega)$	$f(\omega_1) = 0.37464 + 1.54226\omega_1 - 0.26992{\omega_1}^2$	$f(\omega_2) = 0.37464 + 1.54226\omega_2 - 0.26992{\omega_2}^2$
ω	$\omega_1 = \left(\dfrac{3}{7}\left(\dfrac{T_{b,1}}{T_{c,1}-T_{b,1}}\right)\log p_{c,1} - 1\right)$	$\omega_2 = \left(\dfrac{3}{7}\left(\dfrac{T_{b,2}}{T_{c,2}-T_{b,2}}\right)\log p_{c,2} - 1\right)$
T_r	$T_{r,1} = \dfrac{T}{T_{c,1}}$	$T_{r,2} = \dfrac{T}{T_{c,2}}$
d	$d_1 = \dfrac{0.07780RT_{c,1}}{p_{c,1}}$	$d_2 = \dfrac{0.07780RT_{c,2}}{p_{c,2}}$

Let $\overline{\mathbf{x}} = \mathbf{x} + \mathbf{p}(\mathbf{x}) - \mathbf{p}_0$ and $\overline{\mathbf{y}} = \mathbf{y} - \mathbf{p}_0$; system (17) can be represented as follows:

$$\begin{aligned}
\dot{\overline{x}}_1 &= a_1\overline{x}_1 + b_1u_1 + a_1\left[-p_1\left(x_2\right) + p_1\left(x_{20}\right)\right], \\
\dot{\overline{x}}_2 &= a_2\overline{x}_2 + b_2u_2 + a_2\left[-p_2\left(x_1\right) + p_2\left(x_{10}\right)\right], \\
\overline{y}_1 &= \overline{x}_1, \\
\overline{y}_2 &= \overline{x}_2,
\end{aligned} \tag{18}$$

where

$$\begin{aligned}
\mathbf{f}\left(\overline{\mathbf{x}}\right) &= \left[a_1\overline{x}_1 \quad a_2\overline{x}_2\right]^T; \\
h_1\left(\overline{\mathbf{x}}\right) &= \overline{x}_1; \\
h_2\left(\overline{\mathbf{x}}\right) &= \overline{x}_2; \\
\mathbf{g}_1\left(\overline{\mathbf{x}}\right) &= \left[b_1 \quad 0\right]^T; \\
\mathbf{g}_2\left(\overline{\mathbf{x}}\right) &= \left[0 \quad b_2\right]^T; \\
\mathbf{e}_1\left(\mathbf{x}\right) &= a_1\left[-p_1\left(x_2\right) + p_1\left(x_{20}\right) \quad 0\right]^T; \\
\mathbf{e}_2\left(\mathbf{x}\right) &= a_2\left[0 \quad -p_2\left(x_1\right) + p_2\left(x_{10}\right)\right]^T; \\
\overline{\mathbf{x}}_0 &= \left[\left(\zeta_1\sqrt{\zeta_3} + \zeta_4\right)^2 \quad \eta_3\right]^T.
\end{aligned} \tag{19}$$

Definition 1 (see [14, 15]). System (18) is said to have a vector relative degree $\boldsymbol{\rho} = [\rho_1, \dots, \rho_m](u \in \mathbf{R}^m)$ for inputs on the initial state $\overline{\mathbf{x}}_0$ if

(i) $L_{\mathbf{g}_j}L_{\mathbf{f}}^k h_i(\overline{\mathbf{x}}) = 0$ for all $\overline{\mathbf{x}}$ in the field $\overline{\mathbf{x}}_0$, $i, j = 1, \dots, m$, $k < \rho_i - 1$, where

$$\begin{aligned}
L_{\mathbf{f}}^k h_i\left(\overline{\mathbf{x}}\right) &:= \frac{\partial\left(L_{\mathbf{f}}^{k-1}h_i\left(\overline{x}\right)\right)}{\partial\overline{\mathbf{x}}}\mathbf{f}\left(\overline{\mathbf{x}}\right); \\
L_{\mathbf{g}}L_{\mathbf{f}}h_i\left(\overline{\mathbf{x}}\right) &:= \frac{\partial\left(L_{\mathbf{f}}h_i\left(\overline{x}\right)\right)}{\partial\overline{\mathbf{x}}}\mathbf{g}\left(\overline{\mathbf{x}}\right),
\end{aligned} \tag{20}$$

(ii) decoupling matrix

$$\mathbf{A}\left(\overline{x}\right) = \begin{bmatrix} L_{\mathbf{g}_1}L_{\mathbf{f}}^{r_1-1}h_1\left(\overline{\mathbf{x}}\right) & \cdots & L_{\mathbf{g}_m}L_{\mathbf{f}}^{r_1-1}h_1\left(\overline{\mathbf{x}}\right) \\ \vdots & \cdots & \vdots \\ L_{\mathbf{g}_1}L_{\mathbf{f}}^{r_m-1}h_m\left(\overline{\mathbf{x}}\right) & \cdots & L_{\mathbf{g}_m}L_{\mathbf{f}}^{r_1-1}h_m\left(\overline{\mathbf{x}}\right) \end{bmatrix}_{m\times m} \tag{21}$$

is nonsingular on the initial state $\overline{\mathbf{x}}_0$.

According to Definition 1, system (18) has a vector relative degree $\boldsymbol{\rho} = [1, 1]$ for inputs.

Proposition 2 (see [14, 15]). *If system (18) has a vector relative degree, namely, decoupling matrix $\mathbf{A}(\overline{x})$ is nonsingular on the $\overline{\mathbf{x}}_0$, then the inputs and outputs of system can be decoupled near $\overline{\mathbf{x}}_0$ through a static state feedback and vice versa.*

Therefore, system (18) can be decoupled through a static state feedback. Consider a feedback control law:

$$u_i = \alpha_i\left(\overline{\mathbf{x}}\right) + \beta_{i1}\left(\overline{\mathbf{x}}\right)v_1 + \beta_{i2}\left(\overline{\mathbf{x}}\right)v_2, \quad m = 2, \tag{22}$$

where $\alpha(\overline{\mathbf{x}}) := -\mathbf{A}^{-1}(\overline{\mathbf{x}})\mathbf{b}(\overline{\mathbf{x}})$ is an analysis function vector, $\beta(\overline{\mathbf{x}}) = \mathbf{A}^{-1}(\overline{\mathbf{x}})$ is a nonsingular matrix, $\mathbf{b}(\overline{\mathbf{x}}) = [L_{\mathbf{f}}^{\rho_1}h_1(\overline{\mathbf{x}}) \ \cdots \ L_{\mathbf{f}}^{\rho_m}h_m(\overline{\mathbf{x}})]^T$, and $\mathbf{v} \in \mathbf{R}^m$ are new input variables. Substituting (22) into (18), the close-loop system can be obtained:

$$\begin{aligned}
\dot{\overline{x}}_1 &= \left[a_1\overline{x}_1 + b_1\alpha_1\left(\overline{\mathbf{x}}\right)\right] + b_1\left[\beta_{11}\left(\overline{\mathbf{x}}\right)v_1 + \beta_{12}\left(\overline{\mathbf{x}}\right)v_2\right] \\
&\quad + a_1\left[-p_1\left(x_2\right) + p_1\left(x_{20}\right)\right], \\
\dot{\overline{x}}_2 &= \left[a_2\overline{x}_2 + b_2\alpha_2\left(\overline{\mathbf{x}}\right)\right] + b_2\left[\beta_{21}\left(\overline{\mathbf{x}}\right)v_1 + \beta_{22}\left(\overline{\mathbf{x}}\right)v_2\right] \\
&\quad + a_2\left[-p_2\left(x_1\right) + p_2\left(x_{10}\right)\right], \\
\overline{y}_1 &= \overline{x}_1, \\
\overline{y}_2 &= \overline{x}_2,
\end{aligned} \tag{23}$$

where

$$\begin{aligned}
\widetilde{\mathbf{f}}\left(\overline{\mathbf{x}}\right) &= \left[a_1\overline{x}_1 + b_1\alpha_1\left(\overline{\mathbf{x}}\right) \quad a_2\overline{x}_2 + b_2\alpha_2\left(\overline{\mathbf{x}}\right)\right]^T; \\
\widetilde{h}_1\left(\overline{\mathbf{x}}\right) &= \overline{x}_1; \\
\widetilde{h}_2\left(\overline{\mathbf{x}}\right) &= \overline{x}_2; \\
\widetilde{\mathbf{g}}_1\left(\overline{\mathbf{x}}\right) &= \left[b_1\beta_{11}\left(\overline{\mathbf{x}}\right) \quad b_2\beta_{21}\left(\overline{\mathbf{x}}\right)\right]^T; \\
\widetilde{\mathbf{g}}_2\left(\overline{\mathbf{x}}\right) &= \left[b_1\beta_{12}\left(\overline{\mathbf{x}}\right) \quad b_2\beta_{22}\left(\overline{\mathbf{x}}\right)\right]^T.
\end{aligned} \tag{24}$$

Theorem 3 (see [14]). *MIMO nonlinear system (18) has the same vector relative degree as MIMO nonlinear system (23) with a static state feedback for inputs; namely,*

$$\boldsymbol{\rho} = \overline{\boldsymbol{\rho}}. \tag{25}$$

Definition 4 (see [14, 15]). System (18) is said to have a vector relative degree $\boldsymbol{\sigma} = [\sigma_1, \dots, \sigma_m](u \in \mathbf{R}^m)$ for disturbance on the initial state $\overline{\mathbf{x}}_0$ if $L_{\mathbf{e}_j}L_{\mathbf{f}}^k h_i(\overline{\mathbf{x}}) = 0$ for all $\overline{\mathbf{x}}$ in the field $\overline{\mathbf{x}}_0$, $i, j = 1, \dots, m$, $k < \sigma_i - 1$; and $L_{\mathbf{e}_j}L_{\mathbf{f}}^{\sigma_i-1}h_i(\overline{\mathbf{x}}) \neq 0$.

According to Definition 1, system (23) has a vector relative degree $\boldsymbol{\sigma} = [\infty, \infty]$ for disturbance.

Theorem 5. *The nonlinear system (18) with $\boldsymbol{\rho}$ and $\boldsymbol{\sigma}$ is said to be disturbance decoupled on the initial state $\overline{\mathbf{x}}_0$ if and only if*

$$\rho_i < \sigma_i, \quad i = 1, \dots, m. \tag{26}$$

According to Theorem 5, $\rho_1 < \sigma_1$, $\rho_2 < \sigma_2$, system (18) can be disturbance decoupled; namely,

$$\begin{aligned}
z_1^1 &= \overline{x}_1, \\
\dot{z}_1^1 &= v_1,
\end{aligned}$$

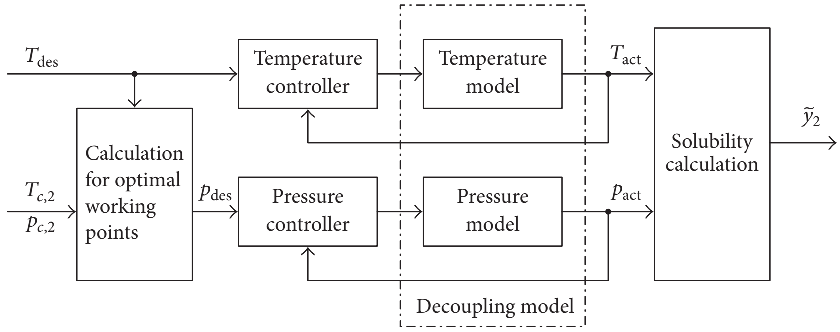


FIGURE 1: Schematic diagram of the proposed solubility optimal system for SFE.

$$\begin{aligned} z_1^2 &= \overline{x}_2, \\ \dot{z}_1^2 &= v_2, \\ \overline{y}_1 &= z_1^1, \\ \overline{y}_2 &= z_1^2. \end{aligned} \tag{27}$$

2.3. Solubility Modeling. The Peng-Robinson equation for the mole fraction, $\tilde{y}_2$, at saturation of a solute of low volatility in SFE can be written as [2]

$$\ln \tilde{y}_2 = \ln \frac{p_v(T)}{p} + \frac{pV_m}{RT} - \ln \phi_2, \tag{28}$$

where $p_v(T)$ is the vapors pressure of the solute, V_m is the volume of the pure solute, and ϕ_2 is the fugacity coefficient, which can be calculated as

$$\begin{aligned} \ln \phi_2 = &-\frac{\Omega_{11}(T)}{2\sqrt{2}RTd_1}\left(\frac{2\Omega_{12}(T)}{\Omega_{11}(T)} - \frac{d_2}{d_1}\right) \\ &\cdot \ln \frac{V + \left(1+\sqrt{2}\right)d_1}{V + \left(1-\sqrt{2}\right)d_1} + \ln \frac{RT}{p\left(V - d_1\right)} \\ &+ \frac{d_2}{d_1}\left(\frac{pV}{RT} - 1\right), \end{aligned} \tag{29}$$

where $\Omega_{11}(T)$, $\Omega_{12}(T)$, d_1, and d_2, which are listed in Table 1, can be calculated by combining rules and are given in [2]. $\Omega_{12}(T)$ can be given with a parameter k_{12} in [2], which describes the mixture. It can be calculated by RBF neural network. However, $k_{12} = 0.5$ is set in this work. The solubility optimal control can be carried out through adjusting temperature and pressure parameters, rather than the parameter in PR model.

3. Temperature-Pressure Decoupling Control and Solubility Optimal Control

A solubility optimal system for SFE is presented, and its effectiveness is evaluated through simulation experiments. The overall control scheme is shown in Figure 1.

3.1. Temperature-Pressure Decoupling Control. Temperature and pressure control are playing an increasingly important role in SFE process. In this work, proportional-integral-derivative (PID) controller is chosen as the temperature and pressure controllers. The transfer function of the PID controller is given by

$$G_{PID}(s) = K_P\left(1 + \frac{1}{T_I s} + T_D s\right), \tag{30}$$

where K_P is proportional gain, T_I is integral time constant, and T_D is derivative time constant.

3.2. Solubility Optimal Calculation. The objective of solubility optimization is to find the optimal working points, namely, the optimal temperature and pressure, to maximize solubility for SFE process. Combining (27) with (28), the yield rate of extraction process can be represented with $\lambda(T, p)$; namely,

$$\tilde{y}_2 = e^{\lambda(T,p)}, \tag{31}$$

where $\lambda(T, p) = \ln(p_v(T)/p + pV_m/RT) - \ln \phi_2$. Hence the optimal calculation is given as

$$\begin{aligned} \frac{\partial \tilde{y}_2}{\partial T} &= \frac{\partial \lambda(T,p)}{\partial T} = 0, \\ \frac{\partial \tilde{y}_2}{\partial p} &= \frac{\partial \lambda(T,p)}{\partial p} = 0, \\ T_l \le T \le T_h&, \quad p_l \le p \le p_h, \end{aligned} \tag{32}$$

where T_l and T_h are the lowest and highest temperature, respectively, and p_l and p_h are the lowest and highest pressure, respectively. Equation (32) can obtain the relationship between temperature and pressure in optimal work points and the optimal molar volume of pure solute. Therefore, the operating parameters can comply with (32).

4. Simulation and Experiment Results and Analysis

Study works are based on SFE optimal system, which is shown in Figure 2. It consists of SFE control equipment and SFE monitoring system. Temperature and pressure control

Table 2: Experimental parameters of biphenyl in supercritical CO_2.

Time (min)	0	43	97	147	183	210
Temperature (K)	313.05	316.75	318.45	322.85	327.95	332.85
Pressure (MPa)	8.00	8.40	9.10	9.64	10.20	10.72
Temperature UEIGOM	313.4697	316.6856	320.7714	324.6015	327.3875	329.4927
	$C = 0.2713$, $P = 100\%$					
Pressure UEIGOM	7.8164	8.3336	9.0318	9.7304	10.2666	10.6880
	$C = 0.0958$, $P = 100\%$					

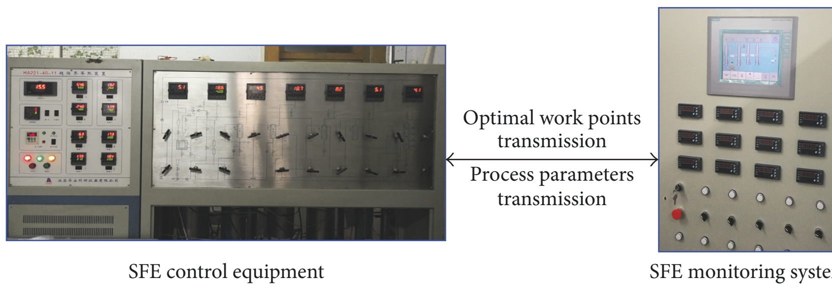


Figure 2: Schematic diagram of the proposed solubility optimal system for SFE.

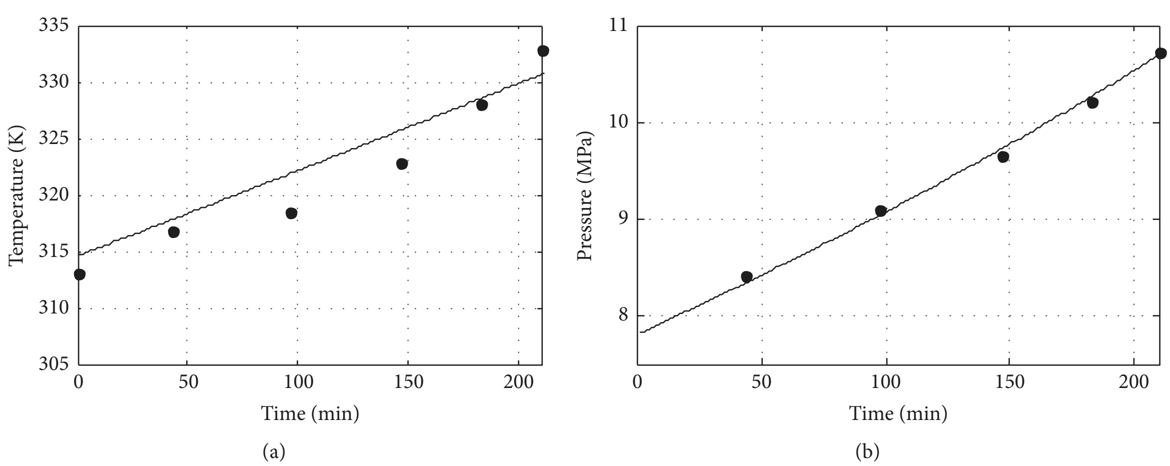


Figure 3: UEIGOM of temperature and pressure process. (a) Temperature process. (b) Pressure process.

can be implemented by SFE control equipment, which is improved SFE equipment (HA21-40-11), for SFE process. In SFE monitoring system, the process parameters can be monitored, and the optimal work points of temperature and pressure to maximize solubility can be calculated and sent to SFE control equipment.

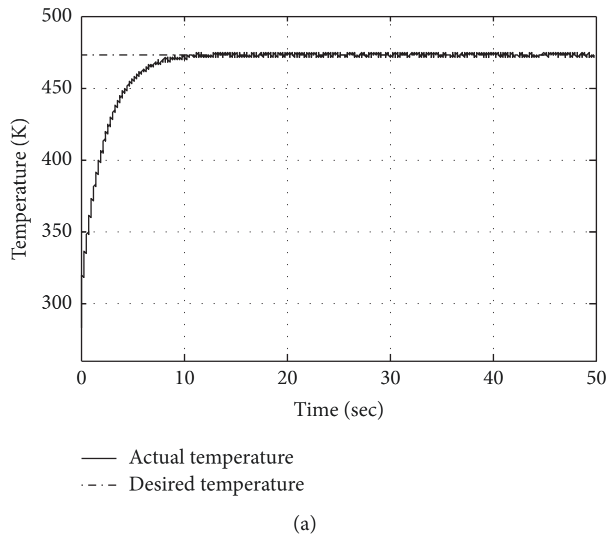
4.1. Temperature-Pressure Process Modeling and Decoupling Control. According to SFE experiments in biphenyl, the experimental parameters are listed in Table 2. Define C as proportionality and P as error frequency, which are related to the quality of UEIGOM. C, the smaller the better, generally requires its maximum to not be more than 0.65. P generally requires more than 0.95 and not less than 0.7. It can be seen with data that the temperature and pressure process can be all described with the first order of inertia, completely. The state equations of temperature and pressure process can be obtained by UEIGOM, which can be shown in Figure 3, as follows:

$$\begin{bmatrix}\dot{x}_1\\ \dot{x}_2\end{bmatrix} = \begin{bmatrix}-4201.7 & 0\\ 0 & -666.7\end{bmatrix}\begin{bmatrix}x_1\\ x_2\end{bmatrix} + \begin{bmatrix}1321800 & 0\\ 0 & 5210.7\end{bmatrix}\begin{bmatrix}u_1\\ u_2\end{bmatrix}, \quad (33)$$

$$\begin{bmatrix}y_1\\ y_2\end{bmatrix} = \begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix}\begin{bmatrix}x_1\\ x_2\end{bmatrix}.$$

Combining (13) with (16), the temperature and pressure coupling model, namely, input-output coupling model, can be described as follows:

$$\begin{bmatrix}\dot{x}_1\\ \dot{x}_2\end{bmatrix}$$



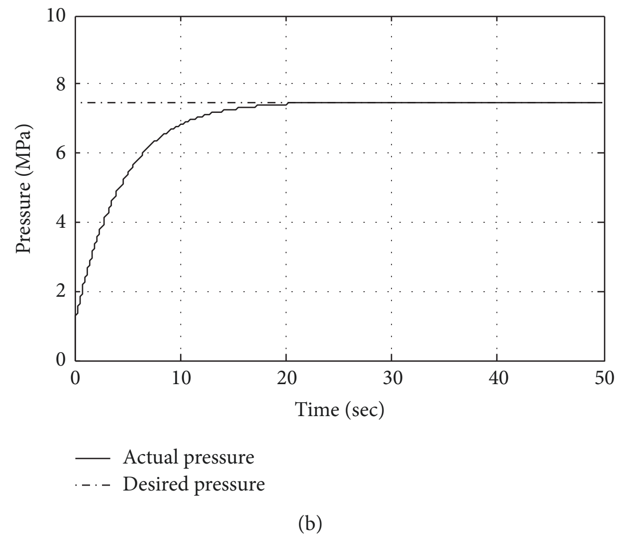


Figure 4: Temperature and pressure decoupling close-loop control. (a) Temperature control. (b) Pressure control.

$$= \begin{bmatrix} -4201.7 & 0 \\ 0 & -666.7 \end{bmatrix} \begin{bmatrix} x_1 \\ x_2 \end{bmatrix} + \begin{bmatrix} 1321800 & 0 \\ 0 & 5210.7 \end{bmatrix} \begin{bmatrix} u_1 \\ u_2 \end{bmatrix},$$

$$\begin{bmatrix} y_1 \\ y_2 \end{bmatrix} = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} x_1 \\ x_2 \end{bmatrix} + \begin{bmatrix} \left(0.0089158\sqrt{2243.2x_2 + 177211} + 16.422\right)^2 \\ 560.8004x_1 - 18419\sqrt{x_1} + 151160 \end{bmatrix}. \tag{34}$$

According to (27), state equation (34), which can be disturbance decoupled, is decoupled as follows:

$$\begin{bmatrix} \dot{\overline{x}}_1 \\ \dot{\overline{x}}_2 \end{bmatrix} = \begin{bmatrix} -4201.7 & 0 \\ 0 & -666.7 \end{bmatrix} \begin{bmatrix} \overline{x}_1 \\ \overline{x}_2 \end{bmatrix} + \begin{bmatrix} 1321800 & 0 \\ 0 & 5210.7 \end{bmatrix} \begin{bmatrix} u_1 \\ u_2 \end{bmatrix}, \tag{35}$$

$$\begin{bmatrix} \overline{y}_1 \\ \overline{y}_2 \end{bmatrix} = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} \overline{x}_1 \\ \overline{x}_2 \end{bmatrix}.$$

Therefore, the temperature and the pressure processes can be controlled independently. In order to verify the feasibility of decoupling models, close-loop control simulations are conducted. The desired temperature is set as 473 K, and the desired pressure is set as 7.5 MPa. The controllers of temperature and pressure process are PID. The simulation results are given in Figure 4. As seen in Figure 4, actual temperature can well track the desired temperature, the same with pressure. The response curve of temperature has slight fluctuation near the desired temperature resulting from the small inertial time constant, but it cannot have an effect on control performance.

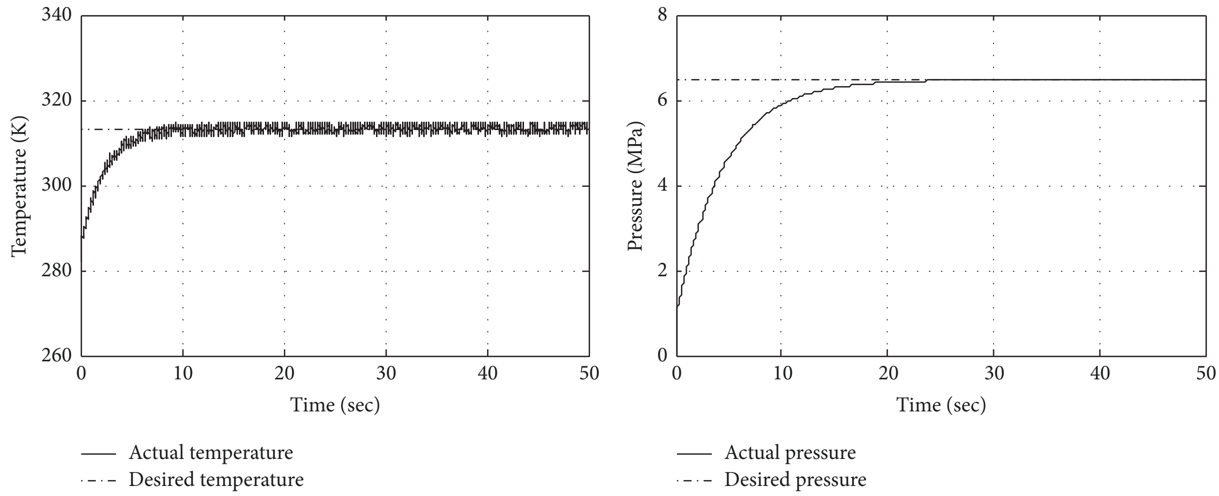
Table 3: Experimental data of biphenyl in supercritical CO_2.

Temperature (K)	Pressure (MPa)	Solubility (10^{-4})
313.16	6.4727	3.1290
318.25	7.5386	3.9730
323.25	8.5773	4.3240
328.35	9.6285	7.6790
333.15	10.6100	12.0570

4.2. Solubility Optimal. The biphenyl solubility data is listed in Table 3 in pressures 8.0 MPa~10.0 MPa, at temperatures 308 K~338 K. As shown in Table 3, the optimal pressure work points can be changed as the desired temperature changes; at the same time, the solubility in optimal work points can be calculated. The control performance of temperature and pressure is shown in Figures 5–9.

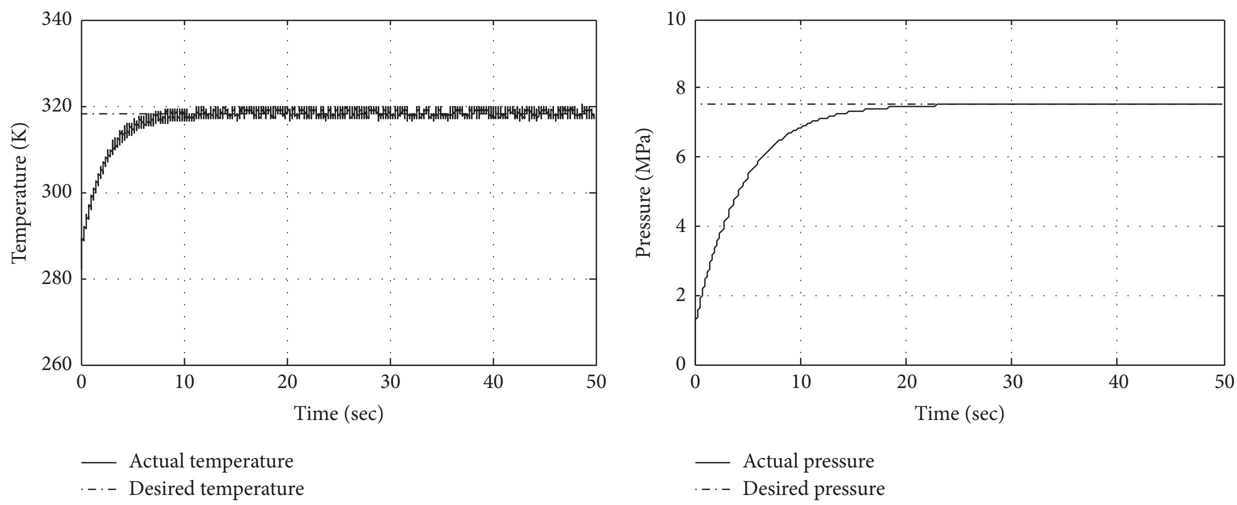
5. Conclusion and Future Works

This paper has presented a new control scheme of solubility optimal system based on a new nonlinear temperature-pressure decoupling model constructed with unequal-interval grey optimal models and Peng-Robinson models. Its performance has been verified through SFE experiments in biphenyl. Conclusions are as follows: Firstly, UEIGOMs of temperature and pressure process are modeled utilizing grey technology, which can extract valuable information from the fewer samples and weaken the randomness of original data. Therefore, UEIGOMs can give the correct description of the reaction character of temperature and



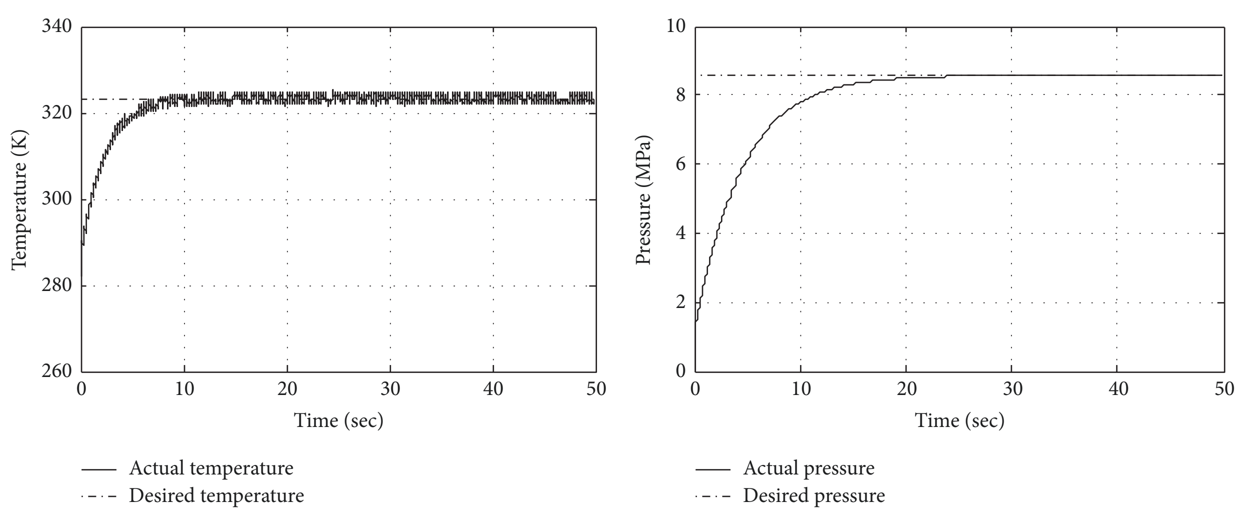
(a) (b)

Figure 5: Temperature and pressure decoupling close-loop control with $T = 313.16$ K and $p = 6.4727$ MPa. (a) Temperature control. (b) Pressure control.



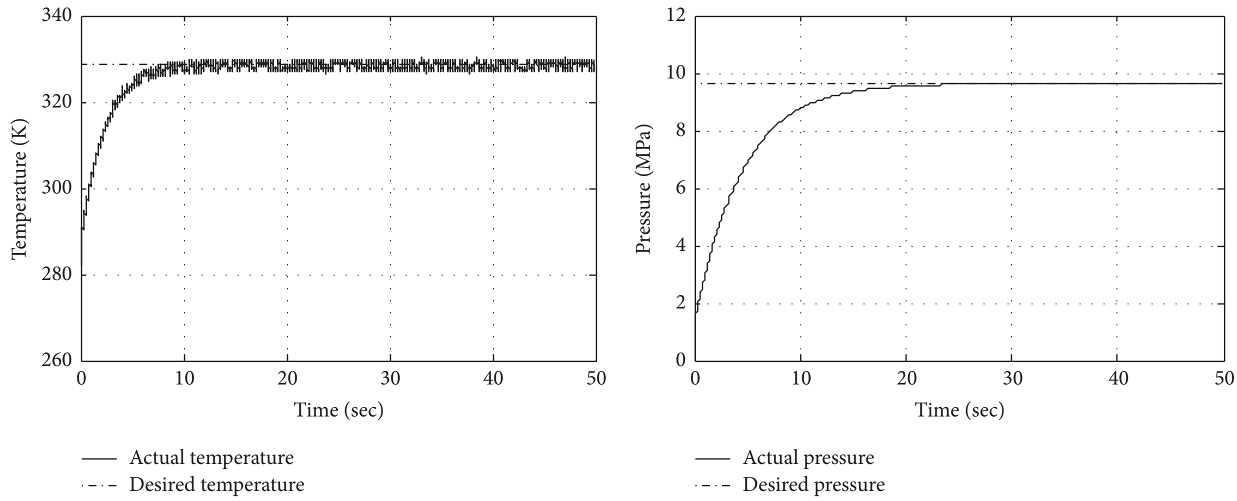
(a) (b)

Figure 6: Temperature and pressure decoupling close-loop control with $T = 318.25$ K and $p = 7.5386$ MPa. (a) Temperature control. (b) Pressure control.



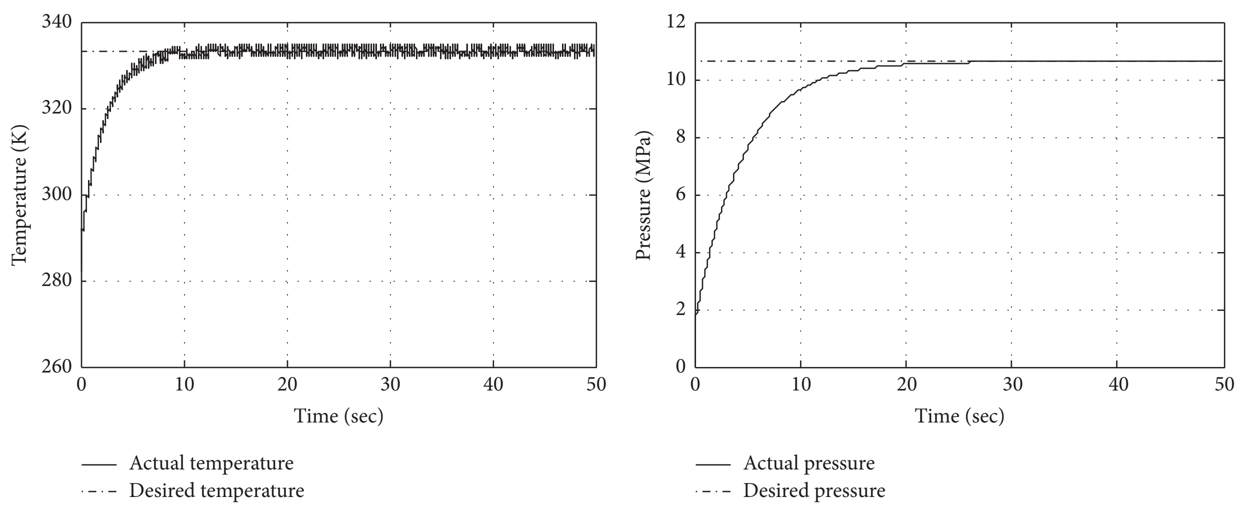
(a) (b)

Figure 7: Temperature and pressure decoupling close-loop control with $T = 323.25$ K and $p = 8.5773$ MPa. (a) Temperature control. (b) Pressure control.



(a) (b)

Figure 8: Temperature and pressure decoupling close-loop control with $T = 328.35$ K and $p = 9.6285$ MPa. (a) Temperature control. (b) Pressure control.



(a) (b)

Figure 9: Temperature and pressure decoupling close-loop control with $T = 333.15$ K and $p = 10.6100$ MPa. (a) Temperature control. (b) Pressure control.

pressure process in SFE. Secondly, based on the relative degree concept of differential geometry theory on nonlinear system, the nondecoupling problem is studied for a class of MIMO nonlinear systems. The whole nonlinear model cannot satisfy input-output decoupling conditions, namely, singularity of decoupling matrix. By transformation of outputs and state variables, it can be disturbance decoupled. Finally, a solubility optimal method is presented in the work; it can calculate the optimal pressure according to the given temperature, namely, optimal working points, to maximize solubility for SFE process.

In addition, the solubility optimal system needs to be further improved resulting from complex processing technology and the solute species. Hence, processing time, energy consumption, solubility, and so forth are all considered to design a composite performance index for optimizing SFE process. This work needs further research and further improvement.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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