

Online estimation for catalyst activity of acetylene hydrogenation reactor

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Abstract

In the ethylene industry, the high purity of the ethylene product depends on hydrogenation in acetylene hydrogenation reactor. Because the catalyst deactivation leads to the moving of the operating point, the operation scheme must be adjusted continually according to the catalyst activity. It is necessary to estimate the catalyst activity online. Based on the discrete dynamic model of the acetylene hydrogenation reactor, the extended Kalman filter (EKF) is used to build the soft sensor for catalyst activity. Considering that EKF involves the large computation costs, we propose a method that estimates the parameters of the time-varying deactivation kinetics model for the tradeoff of accuracy and complexity. The method is effective to reduce computation complexity of estimation, and simultaneously, the accuracy satisfies the process requirement.

KEYWORDS

acetylene hydrogenation reactor, catalyst activity deactivation, extended Kalman filter, soft sensor

1 | INTRODUCTION

The acetylene hydrogenation reactor is a common reactor for purifying ethylene stream in industry, where the palladium catalyst is often used.^{1,2} For simplified modeling, the one-dimensional pseudo-homogeneous model has been widely accepted for the modeling of acetylene hydrogenation reactor.³⁻⁵ Moreover, the improved bimetallic catalyst is also extensively utilized, and comprehensive research about modeling and analysis has been carried out.^{6,7} For higher modeling accuracy, the two-dimensional heterogeneous model is also used for the modeling of acetylene hydrogenation reactor,^{8,9} which is also of higher computation complexity. Based on these models, the relationship among variables is analyzed, and the nature of critical variables is elucidated.

During the running of acetylene hydrogenation reactor, the catalyst deactivation is an inevitable problem.^{10,11} As the catalyst activity slowly declines with time, the

operation must be adjusted. Modeling of catalyst deactivation has been studied by many scholars.¹²⁻¹⁶ Several catalyst deactivation models are proposed, and the majority of studies consider that the catalyst deactivation is related to temperature and catalyst poisoning.^{17,18} In practical production process, the fitted curve of industrial data is employed for temporary catalyst deactivation modeling, which is of no universality.

In industrial processes, it is necessary to obtain some key variables that cannot be measured through online estimation for better control and optimization,¹⁹⁻²¹ so the method of soft sensor is developed. In recent years, the data-driven statistical regression soft sensor has been applied in chemical processes, such as principal component analysis,²²⁻²⁴ partial least square,²⁵⁻²⁷ artificial neural network,^{28,29} support vector machine,^{30,31} and just-in-time learning.³²⁻³⁴ Considering the nonlinearities and time-varying characteristics, a soft sensor development based on supervised ensemble learning with

estimation. Kalman filter is available in practical application of the robust design algorithm proposed.⁴⁷ To handle discrete-time nonlinear model is established as a new method for robust EKF.⁴⁸ It is proved that discrete EKF is more accurate and use.⁴⁹⁻⁵¹

In existing work, the catalyst activity is regarded as a one-dimensional distribution parameter. The activity after a long period of operation is calculated by the existing kinetic model without considering the model mismatch factors.⁵² Thus, the main differences between the existing work are shown as follows: the catalyst activity is regarded as a two-dimensional parameter for estimation and control.

on reactor. Removing concentration steam by In the process, the reactor is gradually increasing time. The reactor stops running and carries out This process lasts several The main products of the reaction and green oil. The content of acetylene is small, and the content of acetylene needs to meet the production. The reactor contains many cleaning device is set between the oil carried in the gas after

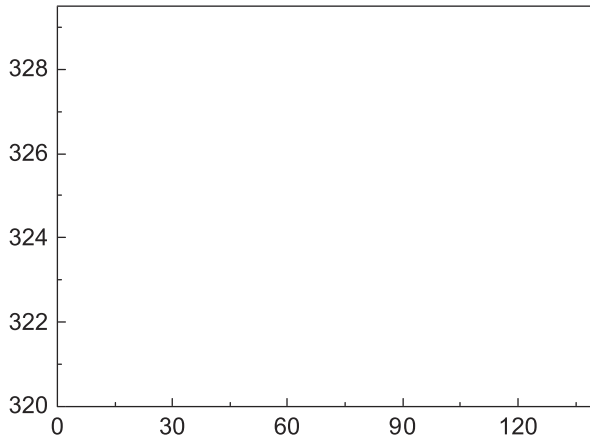
is a complex process involving the deactivation of catalyst, accumulation of reaction, and changes of model parameters. Catalyst activity, a major factor affecting the catalyst surface in the process. But the surface micropores of catalyst are affected by secondary reaction, and the activity of catalyst activity. Instead of one-dimensional distribution, the catalyst activity is a two-dimensional parameter of radical concentration. Early, the oligomer concentration in micropores by secondary reaction is a two-dimensional distribution in the radial direction. As shown in Figure 1, the activity of catalyst activity in radial direction.

The existing research of the acetylene hydrogenation reactor focus on offline operation optimization, but the optimal operating point will deviate because of decreasing catalyst activity with time. Compared with other disturbances, the decreasing catalyst activity has a significant effect on product. Thus, the two-dimensional heterogeneous deactivation model based on the intrinsic kinetics is used in this paper.

Figure 2 shows the tendency of optimal inlet temperature that is treated as one of commonly optimization variables in acetylene hydrogenation reactor. It illustrates that the optimal operation point of the actual reactor in the whole running cycle is related to the decreasing catalyst activity and the online catalyst activity estimation is necessary.

In our previous work, we realized the offline full cycle optimization of acetylene hydrogenation reactor,⁵² taking the influence of catalyst deactivation into account. However, the offline optimization is difficult to cope with the activity changes caused by the unmeasurable uncertainties in actual industrial process. To make the feasible optimal operation strategy, the online soft sensor for catalyst activity is needed. The advantage of online soft sensor is that it could estimate real-time values of the state variables which are difficult to measure directly. By the online soft sensor of the acetylene hydrogenation reactor, the real-time activity of the catalyst could be obtained accurately, and the optimality and robustness of the operation and control strategy could be improved.

In addition, there are certain requirements for accuracy and estimation time of the online soft sensor. For the accuracy, the estimated mean standard deviation should not exceed the activity difference between the last day of the operating cycle and the day before, as shown in Table 1. Considering that the temperature runaway of the reactor caused by the sudden activity change must be



avoided in the actual industrial process, the feedback of the activity in the reactor should be received regularly during operation. Hence, online calculation time of soft sensor should be controlled within 1 hr. In addition, if the reactor is controlled by an advanced process controller (APC), the online calculation time of soft sensor should be about 3 min.

3 | CATALYST ACTIVITY ESTIMATION OF EKF

In response to the above problem description, the two-dimensional heterogeneous model of the reactor is employed. In general, during operation optimization of the acetylene hydrogenation reactor, it is assumed that all the catalyst activity is equivalent in all internal sites of reactor. In this paper, considering the integrality of the acetylene hydrogenation reactor, the internal catalyst activity is treated as a two-dimensional distribution parameter. To design a soft sensor of activity, due to the long duration of the reactor (about a few months even 1 year), the model should be discretized. Considering the catalyst activity of radial and axial direction, the model has been established in our previous work.⁵² Based on that, we propose the acetylene hydrogenation reactor model for discretization. $\theta(k)$ is defined as distribution catalyst activity of radial and axial direction. Mass balance discrete equation for fluid phase is as follows:

$$p_{ik}^g(Z, R, k) = p_{ik}^g(Z, R, k-1) + u(k) \frac{\partial p_{ik}^g(Z, R, k)}{\partial Z} + f_1(Z, R, k). \quad (1)$$

Thermal balance equation for fluid phase is as follows:

$$T_k^g(Z, R, k) = T_k^g(Z, R, k-1) - u(k) \frac{\partial T_k^g(Z, R, k)}{\varepsilon \partial Z} + f_2(Z, R, k). \quad (2)$$

Mass balance equation for catalyst phase is as follows:

$$p_{ik}^s(Z, R, k) = p_{ik}^s(Z, R, k-1) + \frac{\theta(Z, R, k) r_i}{1 - \varepsilon} + f_3(Z, R, k). \quad (3)$$

Thermal balance equation for catalyst phase is as follows:

$$T_k^s(Z, R, k) = T_k^s(Z, R, k-1) + \frac{\theta(Z, R, k)}{(1 - \varepsilon) \rho_s c_{ps}} \sum_{j=1}^2 \eta_j r_j \Delta H_j + f_4(Z, R, k), \quad (4)$$

where p is gas partial pressure in the reactor and T is temperature in the reactor. u is the control matrix; r is the reaction rate of the gas; ε is the porosity of catalyst; ρ is the average density; c_p is the heat capacity; and subscript $i = a, b$, and c is acetylene, ethylene, and hydrogen, respectively. Subscript k denotes the sequence number of reactor bed with $k = 1, 2$, and 3 . Superscript s and g represent the variable of gaseous fluid phase and solid catalyst phase for the heterogeneous model. To simplify the equation, f_1 – f_4 is the part that indirectly related to the estimated catalyst activity. Model parameters are shown in Table 2.

We want to estimate the internal activity of a three-stage acetylene hydrogenation reactor operating for 180 days. The inlet operating data of the three-stage bed reactor are shown in Table 3.

TABLE 2 Model parameters⁵²

Variable	Model parameter	Value
ε	Catalyst porosity	0.35
η_1, η_2	Efficient factor of the ethylene and ethane generated reaction	$3.461, 4.26 \times 10^{-3}$
c_p	Heat capacity ($\text{J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$)	1.724
E_d	Activation energy of catalyst deactivation ($\text{kJ} \cdot \text{kmol}^{-1}$)	9.9
$\Delta H_1, \Delta H_2$	Heat of the ethylene and ethane generated reaction ($\text{J} \cdot \text{mol}^{-1}$)	174.3, 136.7
k_d, K_d	Catalyst deactivation coefficients	$6.73 \times 10^{-3}, 3.09 \times 10^{-3}$
R_g	Gas constant ($\text{J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$)	8.314

TABLE 3 Operational data for acetylene hydrogenation reactors

Operating variable	Value
$p_{c1}^g(0, R, k)/\text{kPa}$	29.55
$T_1^g(0, R, k)/\text{K}$	320.80
$p_{a1}^g(0, R, k)/\text{kPa}$	29.55
$p_{b1}^g(0, R, k)/\text{kPa}$	1684.35
$p_{c2}^g(0, R, k)/\text{kPa}$	14
$T_2^g(0, R, k)/\text{K}$	321.80
$p_{c3}^g(0, R, k)/\text{kPa}$	0.79
$T_3^g(0, R, k)/\text{K}$	322.80
k/day	180

Kalman filter (KF) is the most widely used state estimation method that has been widely applied in communication, navigation, guidance and control, and other fields.⁵³ KF has many different forms. The most common application of the KF to nonlinear systems is in the form of EKF. Exploiting the assumption that all transformations are quasi-linear, the EKF simply linearizes all nonlinear transformations and substitutes Jacobian matrices for the linear transformations in the KF equations.⁵⁴ Considering that the acetylene hydrogenation reactor is a complex nonlinear system, EKF is adopted for online estimation of catalyst activity.

Based on acetylene hydrogenation reactor discrete model, catalyst activity discrete dynamic model is shown in Equations (5)–(7).

$$\theta(k) = H(k)\theta(k-1), \quad (5)$$

$$y(k) = h(\theta(k), u(k), v(k)), \quad (6)$$

$$v(k) \sim (0, R_0), \quad (7)$$

where h is acetylene hydrogenation reactor discrete dynamic model, v is the associated measurement error that obeys normal distribution for catalyst activity, and $H(k)$ is linearization model based on catalyst deactivation kinetics equations, as shown in Equation (8).

$$H(k) = k_d \left(e^{-\frac{E_d}{R_g T^s(k-1)}} \sum_{j=1}^{k-1} \frac{p_a^{s2}(j) p_c^s(j)}{(1 + K_d p_a^s(j))^3} - e^{-\frac{E_d}{R_g T^s(k)}} \sum_{j=1}^k \frac{p_a^{s2}(j) p_c^s(j)}{(1 + K_d p_a^s(j))^3} \right) \quad (8)$$

Estimation error covariance of the catalyst activity $\theta(k)$ is defined as $P(k) = E \left\{ [\theta(k) - \hat{\theta}(k)] [\theta(k) - \hat{\theta}(k)]^T \right\}$. Prior estimation and posteriori estimation of $\theta(k)$ is

respectively defined as $\hat{\theta}^-(k)$ and $\hat{\theta}^+(k)$. EKF equations for $\theta(k)$ is shown in Equations (9)–(14):

$$\hat{\theta}^-(k) = H(k)\hat{\theta}^+(k-1), \quad (9)$$

acetylene hydrogenation reactor, the improved EKF based soft sensor could accurately track the trajectory of catalyst activity.

Catalyst deactivation equation obtains three variable parameters: deactivation coefficient k_d and K_d and deactivation energy E_d . For convenience, we define $\lambda = [\lambda_1, \lambda_2, \lambda_3]$ as estimated target matrix. Then the time-varying deactivation equation and discrete dynamic model are built, as shown in Equations (15)–(18). Equation (15) is the conversion equation of catalyst

activity and deactivation parameters, and the linear model $H_1(k_1)$ can be obtained by combining Equations (15), (5), and (16). $N(k)$ and $N(k_1)$ represent the number of k and k_1 , respectively. In addition, to reduce computation complexity, the discrete interval of improved method could be several times of the previous interval, where the discrete variable is represented by k_1 .

$$\theta(\lambda(k_1), k) = \theta(\lambda(k_1), k-1) - \lambda_1(k_1) e^{-\frac{\lambda_2(k_1)}{R_0 T^2(k)}} \sum_{j=1}^k \frac{p_a^{s^2}(j) p_c^s(j)}{(1 + \lambda_3(k_1) p_a^s(j))^3} \quad (15)$$

$$\lambda(k_1) = H_1(k_1) \lambda(k_1 - 1) \quad (16)$$

$$y_1(k) = h_1(\lambda(k_1), u(k), v_1(k)) \quad (17)$$

$$v_1(k) = (0, \tilde{R}_1) \quad (18)$$

$$N(k) = nN(k_1) \quad n \in N^+ \quad (19)$$

Equations (15)–(17) are substituted into Equations (9)–(14), and the corresponding EKF equations

are established. The estimated error covariance of the catalyst deactivation parameters $\lambda(k_1)$ is defined as $P(k_1) = E\left\{[\lambda(k_1) - \hat{\lambda}(k_1)][\lambda(k_1) - \hat{\lambda}(k_1)]^T\right\}$. Prior estimation and posteriori estimation of $\lambda(k_1)$ are respectively defined as $\hat{\lambda}^+(k_1)$ and $\hat{\lambda}^-(k_1)$. Estimation equations of EKF for $\lambda(k_1)$ are shown in Equations (20)–(25):

$$\hat{\lambda}^-(k_1) = H_1(k_1)\hat{\lambda}^+(k_1 - 1), \quad (20)$$

$$P^-(k_1) = H_1(k_1)P^+(k_1 - 1)H_1(k_1)^T. \quad (21)$$

The Jacobian matrices of partial derivative of \mathbf{h}_1 with respect to the catalyst deactivation parameters and the associated measurement error are respectively represented as \mathbf{Q} and \mathbf{M} .

$$Q(k_1) = \left. \frac{\partial h_1}{\partial \lambda} \right|_{\hat{\lambda}^-(k_1)}, M(k_1) = \left. \frac{\partial h_1}{\partial v_1} \right|_{\hat{\lambda}^-(k_1)} \quad (22)$$

\mathbf{K} is derived as follows:

$$K(k_1) = P^-(k_1)Q^T(k_1)[Q(k_1)P^-(k_1)Q^T(k_1) + M(k_1)R_1M^T(k_1)]^{-1}. \quad (23)$$

Combing the prior estimate, it is feasible to obtain an update equation for the estimate of the catalyst deactivation parameters:

$$\hat{\lambda}^+(k_1) = \hat{\lambda}^-(k_1) + K(k_1)\{y(k_1) - h_1[\hat{\lambda}^-(k_1), u(k), 0]\}. \quad (24)$$

The error covariance update equation is derived as follows:

$$P^+(k_1)K(k_1) = [I - K(k_1)Q(k_1)]P^-(k_1)[I - K(k_1)Q(k_1)]^T + K(k_1)M(k_1)R_1K^T(k_1)M^T(k_1). \quad (25)$$

In addition, considering that measurement matrix $y_1(k)$ and control matrix $u(k)$ are mismatching to the estimated catalyst activity matrix $\hat{\lambda}(k_1)$, $y_1(nk_1)$, and $u(nk_1)$

are treated as the measurement matrix and control matrix instead of $y_1(k)$ and $u(k)$. The improved soft sensor structure is shown in Figure 5. $\hat{\lambda}(k_1)$ is obtained using EKF and substituted into catalyst deactivation model for obtained online activity estimation $\theta(k)$.

Based on above structure and equations, the improved soft sensor is implemented at 180-day operation. As shown in Figure 6, compared with Figure 4, the estimated standard deviations of the reactor inlet are slightly higher. However, the estimated standard deviations of the reactor center and outlet are slightly lower than the previous estimation result.

On the whole, there is not much difference between the two methods in view of the estimation error. However, for computation complexity, according to the performance comparisons in Table 4, the time consumption of the improved method is much less than previous estimation. The estimated time of the improved EKF meets the requirements of safe operation of the acetylene hydrogenation reactor. However, the estimated time should be further shortened when applied to the reactor with APC. The running environment of the soft sensor is Intel Core i7-8750H CPU.

In addition, the discrete interval and number of estimation parameters influence the performance of activity estimation. According to the performance comparisons in Table 4, under the condition of $\lambda = [\lambda_1, \lambda_2, \lambda_3]$, the discrete interval increasing leads the estimated time decreasing but the estimated standard deviations increasing. To the contrary, under the condition of $N(k) = 7N(k_1)$, the number decreasing of estimation parameters leads to the estimated time decreasing but the estimated standard deviations increasing. According to estimation performance in Table 5, the estimated standard deviations of $n = 2$ and 7 have deviation much less than $n = 7$ and 12, in consideration of estimated time, $n = 7$ should be appropriate. However, the estimated standard deviations of $\lambda = [\lambda_1]$ and $\lambda = [\lambda_1, \lambda_2]$ have large deviation compared with $\lambda = [\lambda_1, \lambda_2, \lambda_3]$. Thus, the deactivation parameters are indispensable for the soft sensor.

Although a good result is obtained in Table 5, the above analysis is considered poor sample number and unpersuasive discussion. For further researching of optimal option within different discrete intervals, the Pareto set is gained by average standard deviation of the

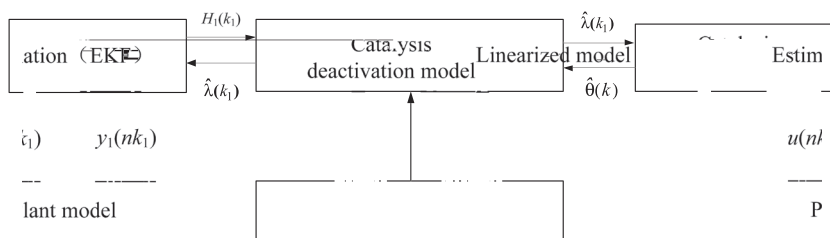
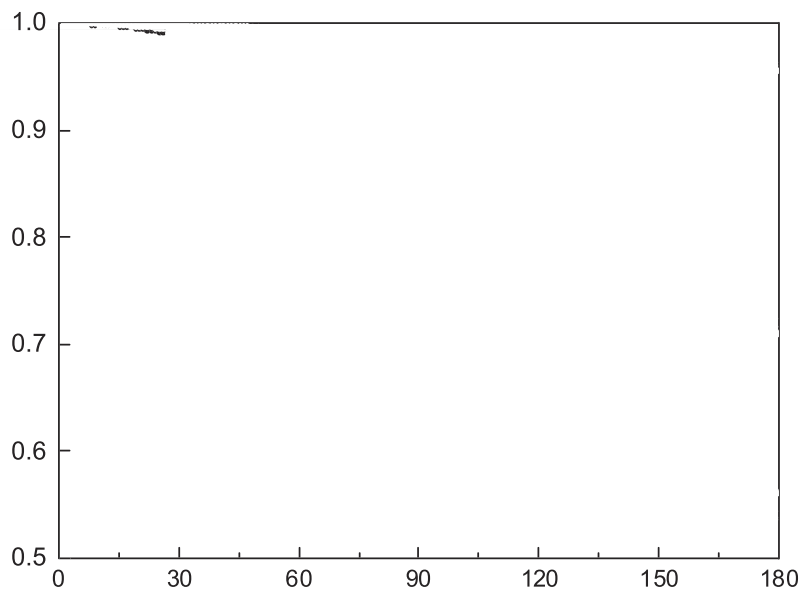


FIGURE 5 The improved soft sensor for activity. EKF, extended Kalman filter



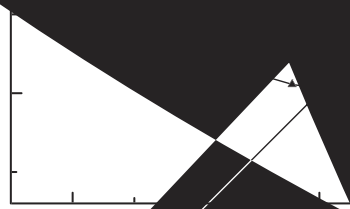
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50



of the relative importance degree of the deactivation coefficient k_d , K_d , and deactivation energy E_d for catalyst deactivation.

Moreover, considering that the above two schemes are difficult to meet APC on estimation error or time, the tradeoff of estimation schemes is proposed. Similarly, to discuss the improved method of two estimation parameters, Pareto curve of $\lambda = [\lambda_1, \lambda_2], [\lambda_2, \lambda_3], [\lambda_1, \lambda_3]$ is gained and shown in Figure 9.

As shown in Figure 9, for estimation with $\lambda = [\lambda_1, \lambda_2]$, the point of $n = 9$ is the optimal point derived by considering the estimated time and average estimated standard deviation. But for estimation with $\lambda = [\lambda_2, \lambda_3]$ and $\lambda = [\lambda_1, \lambda_3]$, the point of $n = 6$ and $n = 7$ is the optimal point. In consideration of that estimation with $\lambda = [\lambda_1, \lambda_2]$ is better than $\lambda = [\lambda_2, \lambda_3]$ or $\lambda = [\lambda_1, \lambda_3]$ on average estimated standard deviation and the estimated time difference is not significant, it is considered for optimal option to estimate catalyst online. The scheme can meet the requirements of safe operation and advanced control in terms of estimated time and accuracy.

5 | CONCLUSIONS

For optimal operation with the reactor running for a long time, the paper establishes the EKF-based soft sensor for tracking tendency of the catalyst activity that is the dominant factor of operation point deviation. Although the estimated error of the EKF is available, it is difficult to be applied in practice because of its high computation complexity. Hence, the paper proposes an improved EKF estimator for more efficient estimation. The improved method builds variable parameter estimator based on catalyst deactivation model, efficiently reducing computation complexity simultaneously ensuring minor estimated error. In addition, considering that estimation performance and estimated time are affected by discrete interval and number of estimation parameters, the paper discusses the optimal option for balance of accuracy and computational complexity. Furthermore, the Pareto set is constructed by multiple estimation taking different parameter estimation combinations and estimated time intervals, so as to find the optimal estimation scheme that comprehensively considers the average estimated standard deviation and estimated time.

The improved EKF method could also be applied to most of the estimation of some parameters that cannot be directly measured and substantially changes the system, such as the heat transfer coefficient of the heat exchangers and the catalyst activity of various fixed-bed reactors. If these parameters are accurately and real-time estimated, it contributes to more optimal operational

strategies and achieves better control and optimization effects. Considering that the improved EKF method relies on accurate mathematical models, the estimation accuracy of the method will be greatly reduced if structure changes of the model and substantial model mismatches occur. It can be considered to add the data-driven statistical regression soft sensor to verify EKF estimation results, such as principal component analysis, support vector machine and even deep learning. Timely verification could improve the robustness of the online soft sensor.

NOMENCLATURE

c_p	heat capacity($J \cdot kg^{-1} \cdot K^{-1}$)
E_d	activation energy of catalyst deactivation ($kJ \cdot kmol^{-1}$)
H, H_1	linearization model based on catalyst deactivation kinetics equations of EKF and improved EKF
h, h_1	acetylene hydrogenation reactor discrete dynamic model of EKF and improved EKF
$\Delta H_1, \Delta H_2$	heat of the ethylene and ethane generated reaction ($J \cdot mol^{-1}$)
K	Kalman gain
k, k_1	discrete variable of EKF and improved EKF
k_d, K_d	catalyst deactivation coefficients
M	The Jacobian matrices of partial derivative of h with respect to the associated measurement error
N	the number of discrete variables
P	estimation error covariance of the catalyst activity
p_a, p_b, p_c	acetylene, ethylene, and hydrogen pressure in the reactor (kPa)
p^s, p^g	the catalyst and the gas phase average gas pressure in the reactor (kPa)
p_{i1}, p_{i2}, p_{i3}	gas pressure in the 1-3 stage reactor (kPa)
Q	The Jacobian matrices of partial derivative of h with respect to catalyst activity or variable parameters
R	dimensionless radius of the reactor
R_0, R_1	covariance matrix of the associated measurement error of EKF and improved EKF
R_g	gas constant($J \cdot mol^{-1} \cdot K^{-1}$)
r_a, r_b, r_c	reaction rate of acetylene, ethylene, and hydrogen in the reactor ($mol \cdot m^{-3} \cdot s^{-1}$)
r_g	reaction rate of green oil in the reactor ($mol \cdot m^{-3} \cdot s^{-1}$)
r_1, r_2, r_3	reaction rate in the 1–3 stage reactor ($mol \cdot m^{-3} \cdot s^{-1}$)
T^s, T^g	the catalyst phase and the gas phase temperature in the reactor (K)

$T_1, T_2,$	
T_3	temperature in the 1–3 stage reactor (K)
u	operation variable
v, v_1	the associated measurement error of EKF and improved EKF
y	output of the catalyst activity discrete dynamic model
Z	dimensionless axial of the reactor
ε	catalyst porosity
η_1, η_2	efficient factor of the ethylene and ethane generated reaction
θ	catalyst activity
ρ	average density ($\text{kg}\cdot\text{m}^{-3}$)
$\lambda_1, \lambda_2, \lambda_3$	variable parameters: deactivation coefficient k_d and K_d , deactivation energy E_d

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