

工业有机污染物治理过程的多目标优化控制

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摘要: 工业有机污染物具有多相态共存、治理工艺复杂、治理成本高等特点. 因此针对有机污染物的临氧裂解/催化氧化治理工艺, 研究了一种基于改进的NSGA-II的优化控制方法. 首先, 根据治理过程的工艺要求和设备操作参数, 使用Aspen Plus建立了稳态模拟系统, 并对关键控制参数进行了灵敏度分析. 然后以最低的总能耗和最大的废水治理量为优化目标, 使用改进的NSGA-II优化处理过程的控制参数, 获得一组帕累托最优解. 最后, 基于能量平衡设计了厂级有机污染物综合处理的能量自平衡控制方案, 并进行动态模拟和实验装置测试. 模拟和实验结果表明, 该系统具有良好的动态响应性能, 所设计的实验装置稳定运行状态下能耗低, 净化效率高.

关键词: 有机污染物治理; NSGA-II; 多目标优化; 动态流程模拟

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Multi-objective optimization control of industrial organic pollutants treatment process

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Abstract: Industrial organic pollutants have the characteristics of pollutants coexistence in different phases, complicated treatment process and high treatment cost. Therefore, for the organic pollutants catalytic cracking/oxidation integrated treatment process, an optimized control method based on improved NSGA-II is studied. First of all, according to the process requirements and equipment operating parameters of the treatment process, a steady-state simulation system is established using Aspen Plus. And the sensitivity analysis is performed on key control parameters. Then, the lowest total energy consumption and the largest wastewater treatment flow are chosen as the optimization objectives. The improved NSGA-II is used to optimize the control parameters of the treatment process, and a set of Pareto optimal solutions is obtained. Finally, based on energy balance, a plant-level control scheme for organic pollutants integrated treatment is designed, and dynamic simulation and experimental device testing were carried out. The simulation and experimental results show that the system has good dynamic response performance, and the designed experimental device has low energy consumption and high purification efficiency under stable operation.

Key words: organic pollutants treatment; NSGA-II; multi-objective optimization; dynamic process simulation

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1 Introduction

With the rapid development of petroleum, chemical and other industries, the issues of organic pollutants treatment have gradually attracted the attention

of academic circles. Industrial organic pollutants have the characteristics of pollutants coexistence in different phases, high pollution concentration, high toxicity, complicated treatment process and high treatment

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cost [1–5]. At present, different methods are often adopted for organic pollutants in different phases. Therefore, problems such as complicated treatment process and difficult optimization control have appeared. The best solution is the integrated treatment of gaseous, liquid and solid organic pollutants in a set of equipment. Biological methods are generally only applicable to non-toxic and low-concentration organic pollutants [6]. At the same time, the pollutants treatment efficiency is limited by the concentration of organic pollutants. High-concentration organic pollutants, especially distillation residue, are extremely difficult to be treated by biological methods. Incineration is one of the main methods of organic pollution control, but it has some disadvantages, such as high energy consumption and the generation of toxic gases, particulate matter and fly ash [7]. Compared with incineration, the catalytic oxidation of organic pollutants can effectively reduce the temperature of the treatment process [8]. One of the research focuses of this technology is to improve the process to reduce energy consumption and secondary pollution. Common catalytic oxidation reactors mainly include: circulating fluidized bed reactors, fluidized bed reactors, stirred reactors and catalytic membrane reactors [9].

There are various treatment processes for organic pollutants, and it is difficult to conduct experiments directly by oxidation methods such as incineration and catalytic oxidation. Therefore, modelling and simulation of pollutants treatment process have gradually become an important part of pollutants treatment research. Chen et al. [10] used Aspen Plus to conduct experiments and process simulations on the characteristics of fixed-bed pyrolysis of domestic waste. And the influence of operating conditions on pyrolysis gas composition, heating value and conversion rate was obtained. Keche et al. [11] designed, simulated, optimized, and analysed fixed-bed biomass gasifiers, and accurately predicted the volume fractions of H_2 , CO , and CO_2 . Ma et al. [12] designed a rotary kiln process control system with three components including incineration, waste heat recovery and flue gas treatment. And Aspen Plus and numerical simulation method were used to simulate the rotary kiln combined with the second combustion chamber. The influence of different operating parameters on furnace temperature and flue gas concentration was analyzed.

Various optimization methods have been developed for different organic pollutants treatment processes, especially the non-dominated sorting algorithm (NSGA-II) has been widely concerned because of its speed and universality in multi-objective optimization. Magli et al. [13] used NSGA-II to optimize the proposed self-contained biogas treatment plant to achieve maximum methane recovery efficiency and lowest total equipment

cost. Qiao et al. [14–15] proposed a multi-objective optimization control scheme in the process of biological treatment of urban wastewater. The improved NSGA-II and adaptive differential evolution algorithm was adopted to optimize the treatment cost and effect, and then the adaptive neural network fuzzy control method was used to design control system. However, there is no report on multi-objective optimization and control of organic pollutants catalytic oxidation treatment process with two-stage fluidized/fixed bed.

In the previous research, a cracking-catalytic oxidation system with two-stage fluidized/fixed bed has been established, and the catalytic oxidation process has been used to treat industrial organic pollutants [16]. A large amount of distillation residue could be generated in the distillation reaction. So, the high calorific value of the distillation residue in the system could be used to compensate for the heat transferred by water vapor and air in the process of catalytic oxidation of waste gas. And it could maintain the heat balance of the reaction system.

In this article, a set of steady-state simulation system was established with Aspen Plus based on heat balance, and key operating parameters were simulated and sensitivity analyzed. Then, the improved NSGA-II was used to optimize the operating parameters of the organic pollutant treatment process. Finally, a plant-level control scheme based on heat balance was designed and dynamically simulated. The simulation results showed that the system had good dynamic response performance and anti-interference performance. And it proved the effectiveness of the plant-level optimization control system.

2 Materials and methods

2.1 Technological process

Organic pollutants in three different phases of gas, liquid and solid can react with oxygen under certain catalytic conditions to produce H_2O , CO_2 and other substances. Therefore, a new two-stage fluidized/fixed bed catalytic system is established for the integrated treatment of multiphase organic pollutants. The fluidized bed has a diameter of 1m and is filled with 300 kg of Cu/Ce catalyst, the particle size distribution of the catalyst is 110~170 μm . The fluidized bed is connected in series with a fixed bed reactor with a diameter of 0.3 m. The fixed bed catalyst is filled with 300 kg. The Cu/Ce catalyst loaded in the fixed bed has a particle size distribution of 2~4 mm. In view of safety and catalyst performance considerations, the temperature of the fluidized bed is controlled at 300~380°C during smooth operation. At the same time, due to the structure, the maximum temperature of the fixed bed cannot be higher than 420°C. The organic pollutant treatment process flow is shown in Fig. 1.

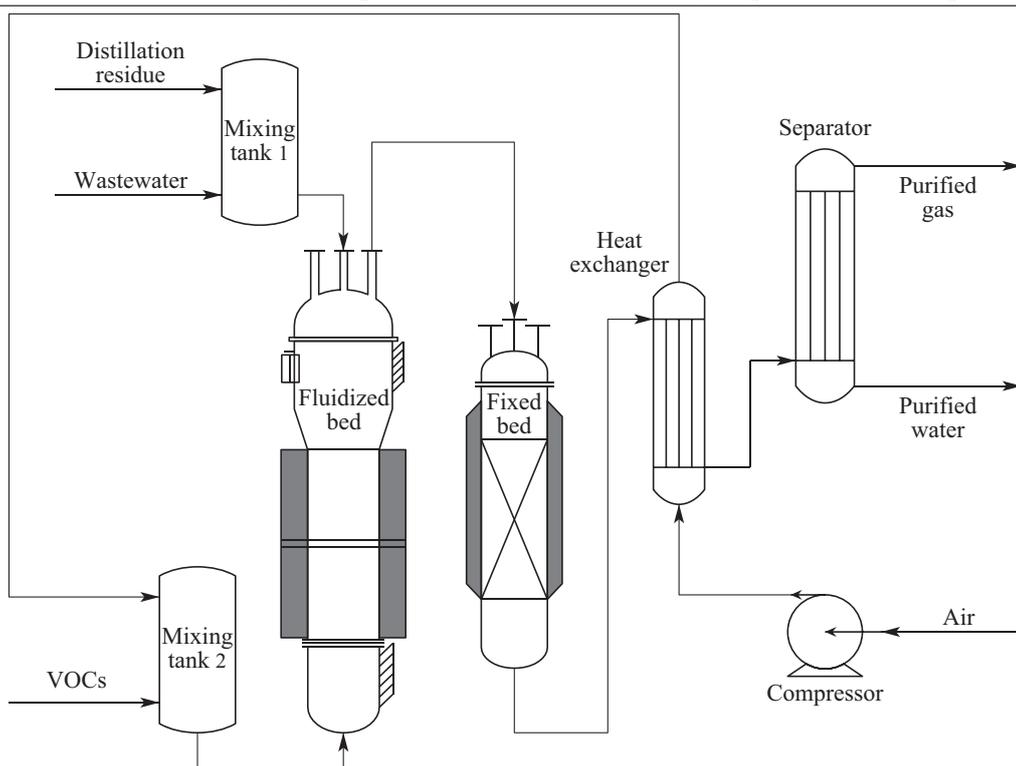


Fig. 1 Process of two-stage fluidized/fixed-bed catalytic system

The organic pollutant treatment process is mainly divided into two stages. The first stage is the heating stage, and in this stage, only air is passed into the device, and then the heating jackets of the fluidized bed and the fixed bed are used to heat themselves. After the temperature in the fluidized bed and the fixed bed reaches the set temperature, the multiphase organic pollutants are introduced according to the process flow in Fig. 1.

The second stage is the stable operation stage. The multiphase organic materials introduced mainly include organic waste gas, process wastewater and distillation residue. First, the organic waste gas is uniformly mixed with a certain amount of air blown into the air compressor, and then passed into the fluidized bed from the bottom. At the same time, the wastewater and the distillation residue are mixed and passed through the top of the fluidized bed. Under the action of the catalyst, cracking and catalytic oxidation reactions occur in the fluidized bed. Currently, the internal reaction in the fluidized bed generates a large amount of heat. And it can be used to heat the incoming organic pollutants and convert them into gaseous state self-heating balance of fluidized bed. Therefore, the self-heat balance of the fluidized bed is achieved. All the gas generated by the reaction is passed into the fixed bed, and further catalytic oxidation reaction is carried out in the fixed bed to treat the excess waste gas. The high-temperature gas discharged from the fixed bed can be used to preheat multiphase organic pollutants to recover heat, and finally pass through a condensing separator to achieve gas-liquid separation. The purified water discharged at the

bottom needs to meet the wastewater direct discharge standard of $80 \text{ mgO}_2/\text{L}$, and the purified gas discharged at the top needs to meet the exhaust gas discharge standard of 120 mg/m^3 .

2.2 Steady-state model

2.2.1 Reaction kinetics and thermodynamics

At present, a few research institutions have studied the reaction kinetics of catalytic oxidation of chemical production tail gas such as acrylonitrile and aromatic hydrocarbons. In this article, the reaction kinetics of catalytic oxidation is comprehensively referred to the research of several institutions. And the intrinsic reaction rate mechanism model takes the treatment of organic pollutants in the production of acrylonitrile as an example. The reaction formula for the catalytic oxidation of organic pollutants is as follows:



Among them, A is organic pollutant. This article assumes that the adsorbed organic pollutants react with oxygen, and the reaction of the adsorbed organic pollutants with oxygen is controlled by the reaction kinetics. The fluidized bed and fixed bed used in the experiment can be considered as ideal integrating reactors for isothermal operation. The reaction kinetic model is obtained as follows:

$$\frac{dx_A}{dW} = \frac{k_A K_A P_{\text{O}_2} P_{\text{AO}} (1 - x_A)}{N_{\text{AO}} [1 + K_A P_{\text{AO}} (1 - x_A)]}. \quad (2)$$

Original condition: $W = 0, x = 0$. The reaction rate constant k_A and adsorption equilibrium constant k_A

in formula (3) are:

$$k_A = k_0 \exp\left[-\frac{E}{RT}\right], \quad (3)$$

$$K_A = a_0 \exp\left[-\frac{Q}{RT}\right]. \quad (4)$$

The reaction kinetic model parameters for the treatment of organic pollutants in acrylonitrile production are shown in Table 1.

In addition, the thermodynamic parameters of the main organic pollutants in acrylonitrile production are shown in Table 2.

Table 1 The reaction kinetic parameters for pollutants treatment in acrylonitrile production

Component	$k_0/(\text{mol} \cdot \text{g}^{-1} \cdot \text{h}^{-1} \cdot \text{MPa}^{-1})$	$E/(\text{J} \cdot \text{mol}^{-1})$	a_0/MPa^{-1}	$Q/(\text{J} \cdot \text{mol}^{-1})$
CO	6.67×10^9	62833	8.2×10^{13}	1.25×10^5
C ₃ H ₆	2.09×10^{11}	72834	2.0×10^{10}	1.14×10^5
C ₃ H ₈	6.28×10^4	43936	1.06×10^6	5.73×10^4

Table 2 Thermodynamic parameters of main organic pollutants in acrylonitrile production

	Melting point/K	Boiling point/K	Relative density/(25°C)	Vaporization Heat/(kJ·mol ⁻¹)	Combustion Heat/(kJ·mol ⁻¹)
C ₃ H ₆	87.9	225.4	0.52	18.41	-1923
C ₃ H ₈	85.5	231.1	0.58	18.77	-2040
CO	68.1	81.7	0.79	6.04	-283
C ₂ H ₄ O ₂	16.7	118	1.049	—	-876.1
C ₃ H ₄ O ₂	14.0	140.9	1.05	—	-1536.4

2.2.2 Energy balance model

In the process of establishing the energy balance model, the main factors are considered based on the reaction mechanism. Some unimportant factors, such as the enthalpy change of H₂O and CO₂ produced by the reaction, and the heat loss of the reaction process to the outside world, are ignored. In the whole process, the energy output of the system mainly includes the endothermic heat of the phase change and the pollutant heating up to the reaction temperature. The energy input of the system mainly includes the oxidative heat release of organic pollutants, as well as additional supplementary heating by electric heaters that may exist.

$$Q_{\text{rwg}} = \sum_{i=1}^n \frac{(H_{\text{wgi}} \times V_{\text{wgi}} \times C_{\text{wgi}}) \times 1000}{V_{\text{m}}}, \quad (5)$$

$$Q_{\text{rww}} = \frac{H_{\text{ww}} \times V_{\text{ww}} \times \text{COD}}{1000}, \quad (6)$$

$$Q_{\text{rds}} = \rho_{\text{ds}} \times V_{\text{ds}} \times H_{\text{ds}}, \quad (7)$$

$$Q_{\text{r}} = Q_{\text{rwg}} + Q_{\text{rww}} + Q_{\text{rds}}, \quad (8)$$

where Q_{r} (kJ/h) is the total energy input supplied by the catalytic oxidation of organic pollutants, and Q_{rwg} , Q_{rww} , and Q_{rds} are respectively the combustion heat released by the catalytic combustion of waste gas, waste water and kettle residue in the reaction. C , V and H are the concentration, volume and standard combustion heat of each phase of organic pollutants.

Among the energy input, wastewater is the most complicated. Therefore, the influence of a small amount of organic matter in wastewater is ignored, and the heat

absorption of waste water in the system is regarded as the change of water in the system.

$$Q_{\text{aH}_2\text{O-SH-w}} = VM_{\text{H}_2\text{O}} \times 4.2 \times \frac{M_{\text{H}_2\text{O}}}{1000} \times (100 - T_0), \quad (9)$$

$$Q_{\text{aH}_2\text{O-LH}} = VM_{\text{H}_2\text{O}} \times r_{\text{H}_2\text{O}} \times \frac{M_{\text{H}_2\text{O}}}{1000}, \quad (10)$$

$$Q_{\text{aH}_2\text{O-SH-v}} = VM_{\text{H}_2\text{O}} \int_{T_0}^{T_{\text{flu}}} (29.16 + 14.49 \times 10^{-3}T - 2.022 \times 10^{-6}T^2) dT, \quad (11)$$

$$Q_{\text{aH}_2\text{O}} = Q_{\text{aH}_2\text{O-SH-w}} + Q_{\text{aH}_2\text{O-LH}} + Q_{\text{aH}_2\text{O-SH-v}}, \quad (12)$$

where $Q_{\text{aH}_2\text{O}}$ represents the energy input caused by the heating and vaporization of wastewater. $Q_{\text{aH}_2\text{O-SH-w}}$, $Q_{\text{aH}_2\text{O-LH}}$ and $Q_{\text{aH}_2\text{O-SH-v}}$ are the heat absorbed by wastewater from the initial temperature to 100°C, the heat of vaporization of wastewater at 100°C, and the heat absorbed by wastewater vapor from 100°C to fluidized bed operating temperature. $VM_{\text{H}_2\text{O}}$ is the mole flow of water, $M_{\text{H}_2\text{O}}$ is the molar mass of water, and $r_{\text{H}_2\text{O}}$ is the latent heat of vaporization of water.

$$Q_{\text{aN}_2} = VM_{\text{N}_2} \int_{T_0}^{T_{\text{flu}}} (27.32 + 6.226 \times 10^{-3}T - 0.9502 \times 10^{-6}T^2) dT, \quad (13)$$

$$Q_{\text{aO}_2} = VM_{\text{O}_2} \int_{T_0}^{T_{\text{flu}}} (28.17 + 6.297 \times 10^{-3}T -$$

$$0.7494 \times 10^{-6} T^2) dT,$$

$$Q_a = Q_{aN_2} + Q_{aO_2} + Q_{aH_2O}, \quad (15)$$

where Q_a is the total energy input generated by heating or vaporization. Q_{aN_2} and Q_{aO_2} are respectively the heat absorbed by the inert gas N_2 and the heat absorbed by O_2 from the initial temperature T_0 to the fluidized bed operating temperature T_{flu} . VM_{N_2} and VM_{O_2} are the mole flow of nitrogen and oxygen respectively.

The energy balance model is an important basis for determining the feed ratio of each phase state organic matter, designing steady-state simulation, and establishing an optimization model.

When $Q_r < Q_a$, the energy input of the reaction is low, which will cause the temperature in the fluidized bed and the fixed bed to drop. This will affect the normal progress of the reaction. At this time, the content of organic pollutants should be increased or additional electric heating should be performed.

When $Q_r > Q_a$, the temperature increase may cause damage to the catalyst or the reactor, and even affect the progress of the reaction. At this time, the flow of kettle residue should be increased or the flow of waste water and air should be reduced to reduce the temperature in the reactor.

2.3 Multi-objective optimization

2.3.1 The multi-objective optimization model

Since the flow of waste water and waste gas is usually large and the capacity of the intermediate storage tank is limited, it is often necessary to prioritize the demand for pollutant treatment in the project. And the waste gas is often proportional to the waste water flow, so the waste water flow is one of the optimization goals. After determining wastewater flow, the most important optimization goal is to achieve dynamic energy balance while meeting the boundary conditions to achieve the lowest total energy consumption. Therefore, this optimization model selects waste water flow and energy consumption as the two optimization goals, and then uses the optimization algorithm to obtain the Pareto optimal solutions. It is easy to find that the two are mutually opposed goals. The larger the flow of wastewater we need to treat, the higher the minimum total energy consumption. According to the wastewater flow to be treated, chemical engineers can select the lowest total power consumption and a set of optimal control variables corresponding to the wastewater flow in the solutions. Wastewater flow is one of the control variables and does not need to be calculated. The calculation formula of total energy consumption is as follows:

$$\sum P = P_c + P_p + P_{h1} + P_{h2}, \quad (16)$$

where $P, P_c, P_p, P_{h1}, P_{h2}$ are respectively the total energy consumption, air compressor power, water pump

power, fluidized bed heater power, fixed bed heater power.

$$P_c = \frac{Q_{c1}(F_1)}{t} \times \frac{1}{\eta_c} + \frac{Q_{c2}(F_1, K_1, K_2)}{t} \times \frac{1}{\eta_c}, \quad (17)$$

$$P_p = \frac{Q_{p1}(F_1)}{t} \times \frac{1}{\eta_p} + \frac{Q_{p2}(F_1, K_2)}{t} \times \frac{1}{\eta_p}, \quad (18)$$

$$P_{h1} = \frac{Q_{h1}(F_1, K_1, K_2, T_1, T_2)}{t} \times \frac{1}{\eta_{h1}}, \quad (19)$$

$$P_{h2} = \frac{Q_{h2}(F_1, K_1, K_2, T_1, T_2)}{t} \times \frac{1}{\eta_{h2}}, \quad (20)$$

where F_1, K_1, K_2, T_1, T_2 are respectively the wastewater flow, oxygen excess multiple, ratio of distillation residue to wastewater, fluidized bed temperature, fixed bed temperature (5 decision variables). $Q_{c1}, Q_{c2}, Q_{p1}, Q_{p2}, Q_{h1}, Q_{h2}$ are respectively the required energy of waste gas compressor, air compressor, wastewater pump, distillation residue pump, fluidized bed and fixed bed. $\eta_c, \eta_p, \eta_{h1}, \eta_{h2}$ are respectively air compressor efficiency (62.5%), pump efficiency (90%), fluidized bed thermal efficiency (85%), fixed bed thermal efficiency (60%).

The boundary range of this optimization model includes: the temperature of the fluidized bed is maintained at 300~380°C; the temperature of the fixed bed is higher than the temperature of the fluidized bed and at the same time lower than 420°C; the wastewater flow is not higher than 70 L/h; The excess oxygen multiple is not higher than 2; the ratio of distillation residue to wastewater is not higher than 0.1; the COD of purified water is lower than the wastewater discharge standard; the concentration of organic pollutants in the purified gas is lower than the exhaust emission standard. The formula for calculating emissions is as follows:

$$\text{COD} = \sum_{i=1}^n \frac{n_i \times \text{OC}_{wi} \times 32}{F_w}, \quad (21)$$

$$\text{VOC} = \sum_{i=1}^n \frac{m_i}{F_g}. \quad (22)$$

COD and VOC are respectively the COD of purified water and the concentration of organic pollutants in the purified gas.

n_i is the amount of substance per unit of time in purified water, OC_{wi} is the amount of oxygen substance corresponding to the complete oxidation of 1 mol of the organic pollutant, F_w is the wastewater flow, F_g is the waster gas flow.

Therefore, the multi-objective optimization model is as follows:

$$\begin{cases} \min f_1(F_1, K_1, K_2, T_1, T_2) = P_c + P_p + P_{h1} + P_{h2}, \\ \max f_2(F_1) = F_1 \times C_1 + \alpha \times F_1 \times C_2, \end{cases} \quad (23)$$

$$s.t. \begin{cases} 10 \leq F_1 \leq 70, \\ 0 \leq K_1 \leq 2, 0 \leq K_2 \leq 0.1, \\ 300 \leq T_1 \leq 380, 300 \leq T_2 \leq 420, T_1 \leq T_2, \\ 0 \leq COD \leq 80, 0 \leq VOC \leq 120, \end{cases} \quad (24)$$

where C_1 is the concentration of organic pollutants in the waste water, C_2 is the concentration of organic pollutants in the waste gas, and α is the ratio of the flow rate of waste gas to waste water. In our optimized problem, C_1 , C_2 and α are all idealized as constants, so the second optimization goal is equivalent to the largest wastewater flow.

2.3.2 Improved NSGA-II

An enhanced non-dominated sorting genetic algorithm with elite retention strategies (NSGA-II) is employed by Deb et al. [17]. And the main advantages of the algorithm are the rapid non-dominated sorting method, the concept of crowding and the elite retention strategy. It can solve the complex multi-objective optimization problem of strong coupling and non-linearity in the multiphase organic pollutant treatment process. Compared with methods such as simulated annealing, general genetic algorithm, particle swarm optimization, etc., this algorithm can solve multi-objective problems with good accuracy and rapidity, and is not easy to fall into local optimization. The optimization in this paper is achieved through the interaction between Aspen Plus and MATLAB. Running a steady-state simulation requires a few seconds of running time, so in the case of a high population size and number of iterations, the running time can reach tens of hours or even days. And in the organic pollutant treatment process, sometimes it is necessary to re-obtain the optimization results according to the process, productivity, and the variety or concentration of pollutants produced. In order to obtain new optimization results as soon as possible, faster optimization speed is required. Therefore, the generation and mutation methods of the initial population are improved.

1) Fast non-dominated sorting based on crowding.

For individuals in fast non-dominated sorting, there are two key parameters, one is the non-dominated rank F , and the other is the crowding degree i_d . When comparing different individuals, first determine the non-dominated level, and the higher the non-dominated rank is the better. When the non-dominated ranks are the same, the individual with lower congestion operator is better. If an individual x_1 is better than another individual x_2 in all optimization goals, x_1 dominates x_2 . All individuals which are not dominated by any other individuals are given the non-dominated level F_1 . Then the individuals which have been given the non-dominated level is removed, and in the same way, the

non-dominated levels $F_2, F_3 \dots$ are given. Finally, the individual crowding comparison operators are calculated by the following formula:

$$I(d_k) = I(d_k) + \frac{I(k+1).m - I(k-1).m}{f_m^{\max} - f_m^{\min}}. \quad (25)$$

In this way, all individuals can be sorted according to the non-dominant rank and the crowding comparison operators.

2) Initial population of Gaussian distribution.

Because the distribution of the first generation population is too scattered, the process of finding several elite solutions has consumed a lot of time, and it is also easy to cause the problem that the elite solutions are too close. However, in order not to fall into the local optimum, it is also necessary to be able to jump out of the limit when the population iterates, so the value range cannot be narrowed directly. Therefore, the initial population generation methods of the four control variables of oxygen excess multiple, distillation residue-wastewater ratio, fluidized bed temperature and fixed bed temperature are changed from random distribution to Gaussian distribution. The Box-Muller method is used to generate an improved initial population. The probability density function of the primary population is as follows:

$$x_{ki} = \frac{x_{kb} - x_{ka}}{6} \times \sin(2\pi k_a) \times \sqrt{-2 \ln(1 - k_b)} + \frac{x_{ka} + x_{kb}}{2}. \quad (26)$$

Among them, k_a and k_b are random numbers from 0 to 1, x_{ki} is the k -th decision variable of the i -th individual in the initial population, and x_{ka}, x_{kb} are the minimum and maximum values of the k -th decision variable respectively.

3) Elite retention strategy with improved mutation.

NSGA-II proposed a genetic operator composed of simulated binary crossover (SBX) and polynomial mutations to ensure the diversity of the population and the retention of elite individuals.

In the simulated binary crossover (SBX), the mathematical formula for the generation of offspring by the parent is:

$$c_{1,k} = \frac{1}{2}[(1 - \beta_k)p_{1,k} + (1 + \beta_k)p_{2,k}], \quad (27)$$

$$c_{2,k} = \frac{1}{2}[(1 + \beta_k)p_{1,k} + (1 - \beta_k)p_{2,k}], \quad (28)$$

Among them, $c_{1,k}, c_{2,k}, p_{1,k}, p_{2,k}$ are respectively the k -th components of c_1, c_2, p_1, p_2 ; β_k is a random variable which is used to produce $c_{1,k}, c_{2,k}$, probability density functions of β_k are as follow:

$$f_{\beta_k}(\beta_k) = \frac{1}{2}(\eta_c + 1)\beta_k^{\eta_c}, \quad 0 \leq \beta_k \leq 1, \quad (29)$$

$$f_{\beta_k}(\beta_k) = \frac{1}{2}(\eta_c + 1)\beta_k^{\frac{1}{\eta_c+2}}, \quad \beta_k > 1. \quad (30)$$

Then,

$$\beta_k = (2u)^{\frac{1}{\eta_c+1}}, u \leq 0.5, \quad (31)$$

$$\beta_k = \frac{1}{[2(1-u)]^{\frac{1}{\eta_c+1}}}, u > 0.5, \quad (32)$$

where u is the random number between $(0, 1)$, η_c is the cross-distribution index.

The mutation method of NSGA-II is relatively simple. When dealing with some complex optimizations, it often requires more iterations to jump out of the local optimum. Therefore, in addition to the original mutation method, a mutation method based on exponential distribution is added to the polynomial mutation. The trend of the exponential distribution is relatively gentle, and it is easier to jump out of the local optimum when using the exponential distribution mutation to improve the optimization speed.

In the polynomial mutation, the generation of offspring C by parent P is through the following variation:

$$c_k = p_k + (x_k^n - x_k^l)\delta_k, \quad (33)$$

c_k, p_k are respectively the k -th components of c, p ; δ_k

is computed with follow formula:

$$\delta_k = (4r_k)^{\frac{1}{\eta_m}} - 1, 0 \leq r_k < 0.25, \quad (34)$$

$$\delta_k = 1 - [2 - 4r_k]^{\frac{1}{\eta_m+1}}, 0.25 \leq r_k \leq 0.5, \quad (35)$$

$$\delta_k = -e^{(2-4r_k)\eta_m}, 0.5 \leq r_k < 0.75, \quad (36)$$

$$\delta_k = e^{(3-4r_k)\eta_m}, 0.75 \leq r_k < 1, \quad (37)$$

The range of r_k is $(0 \sim 1)$, and η_m is the variation distribution index.

The improved NSGA-II optimization program is written in Matlab, and then the communication between Matlab and Aspen Plus is realized through COM technology. The decision variables are constantly modified and rerun in Aspen Plus to calculate the objective function value. Taking the lowest total energy consumption and the largest wastewater flow rate as the optimization goals, the five decision variables of F_1, K_1, K_2, T_1, T_2 are searched through the iteration of the improved NSGA-II algorithm, and the Pareto optimal solutions are output. The flow chart of the improved NSGA-II algorithm for the treatment process is shown in Fig. 2.

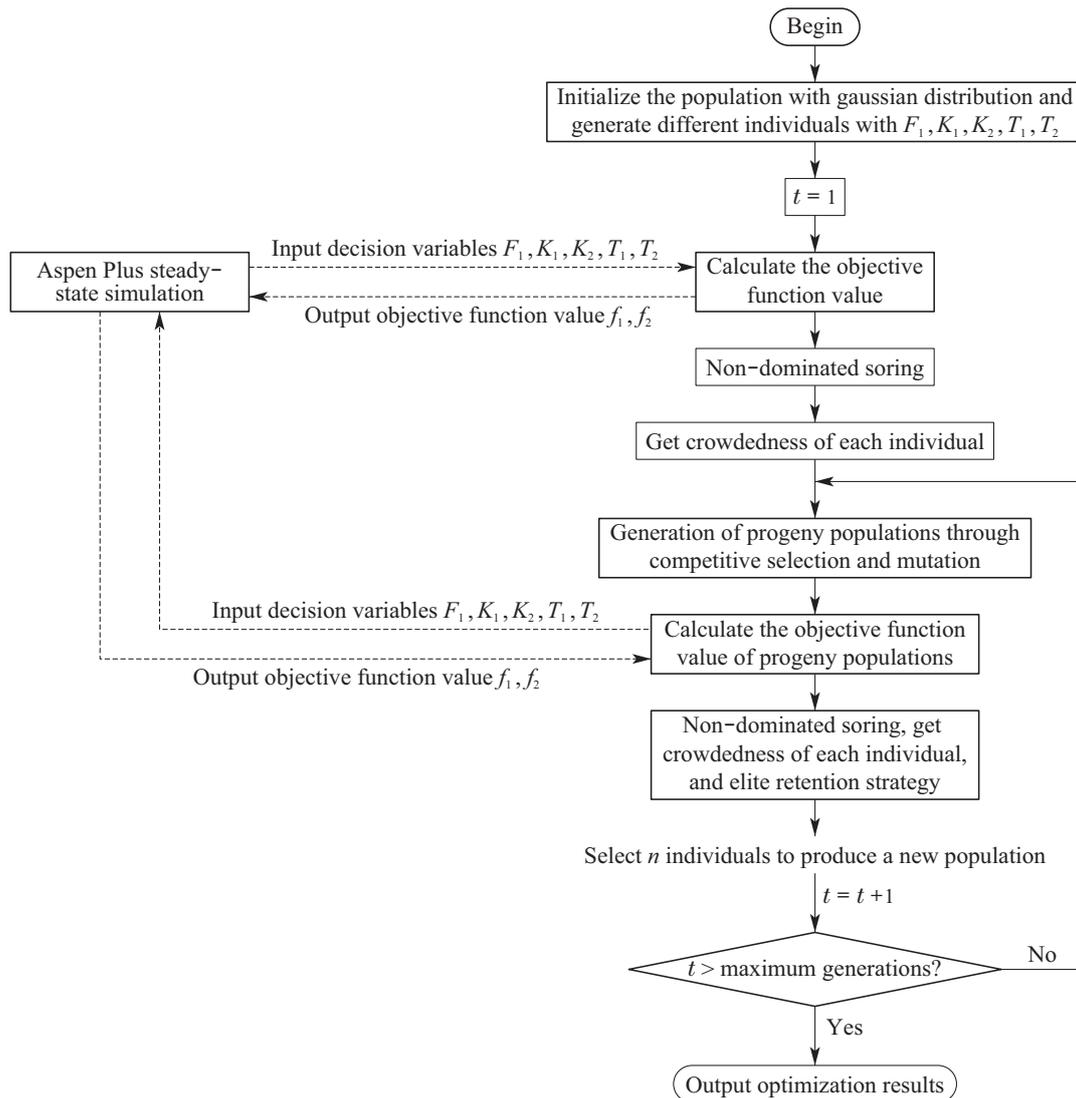


Fig. 2 Flow chart of the improved NSGA-II algorithm for the treatment process

3 Results and discussion

3.1 Simulation and sensitivity analysis

Since optimization control is based on control variables, it is necessary to simulate the treatment process and perform sensitivity analysis on these control variables to determine the boundary range of control variables during optimization control. According to the catalytic oxidation reaction energy balance model, kinetics and thermodynamics, the integrated treatment process of multiphase organic pollutants was simulated using Aspen Plus. The simulation mainly realized the feed mixing of waste gas and air, the feed mixing of wastewater and distillation residue, the fluidized bed reaction process, the fixed bed reaction process, the electric heating process and the waste heat recovery process. At the same time, the process was simplified as follows:

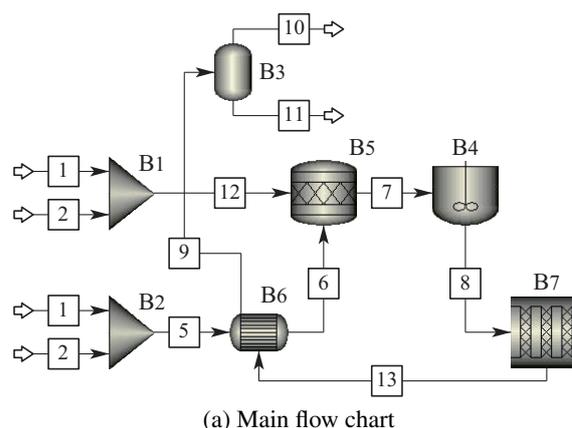
- 1) The waste gas and air are fully mixed;
- 2) Wastewater and distillation residue are fully mixed;
- 3) The reactor of the fluidized bed is divided into two parts. The first part adopts RStoic which is assumed that some macromolecular organic pollutants without kinetic equation can be completely oxidized. The second part adopts RStoic, which is assumed that the waste gas and air are fully mixed in the fluidized bed and react according to the given reaction kinetics.

The default assumption is that the room temperature is 25°C, the atmospheric pressure is 1.01 bar, the flow after the organic waste gas and air are mixed is 80,000 L/h, the concentration is 1%, the oxygen excess multiple is 1, the wastewater flow is 50 L/h, the distillation residue flow is 2 L/h, the ratio of distillation residue to wastewater is 0.04, the temperature of the fluidized bed is 370°C, and the fixed bed temperature is 10°C higher than the fluidized bed. The specific composition of waste gas, waste water, and distillation residue is mainly determined according to the pollutants produced in the acrylonitrile production. At the same time, some components with a proportion of less than 1% in each stream are ignored. The simulated components used in Aspen Plus are shown in Table 3.

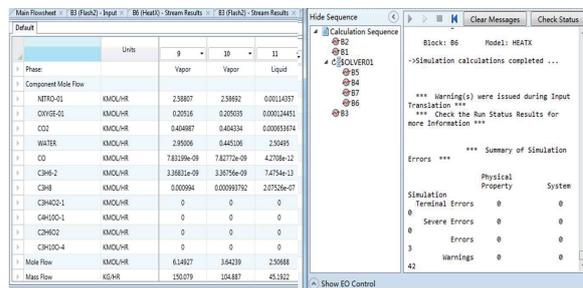
Table 3 The simulated components used in Aspen Plus

Stream name	Flow/(L·h ⁻¹)	Components	Mass/%
Air	79200	N ₂	78.0
		O ₂	22.0
		CO	40.0
Waste gas	800	C ₃ H ₆	14.4
		C ₃ H ₈	45.6
Waste water	50	H ₂ O	95.5
		C ₃ H ₄ O ₂	4.5
		C ₃ H ₄ O ₂	42.8
Distillation residue	2	C ₄ H ₁₀ O	13.9
		C ₂ H ₆ O ₂	38.7
		C ₃ H ₁₀ O ₄	4.6

Since the process includes multi-phase organic impurities but has been transformed into gaseous state during the reaction, the NRTL physical property method is selected. There are also many different calculation models for the reaction kinetics in the reactor. The reaction kinetic formula of the catalytic oxidation process in fluidized bed and fixed bed is shown in Section 2.2.1, and since the kinetic equation of adsorption and reaction is adopted in this article, the LHHW kinetic model is chosen, and the forward driving force is set to 1 and the reverse driving force is set to approach 0. The established Aspen Plus model is shown in Fig. 3.



(a) Main flow chart



(b) The result of export composition and operating status

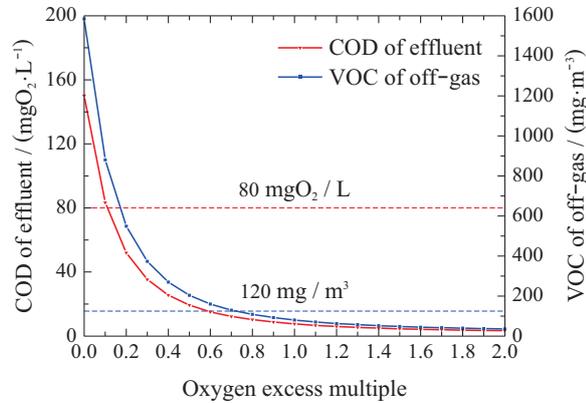
Fig. 3 Aspen Plus steady-state simulation

According to the default reaction conditions, the steady-state simulation was realized. The COD of the purified water was 7.53 mgO₂/L, and the concentration of organic pollutants in the purified gas was 79.58 mg/m³, which meets the emission requirements.

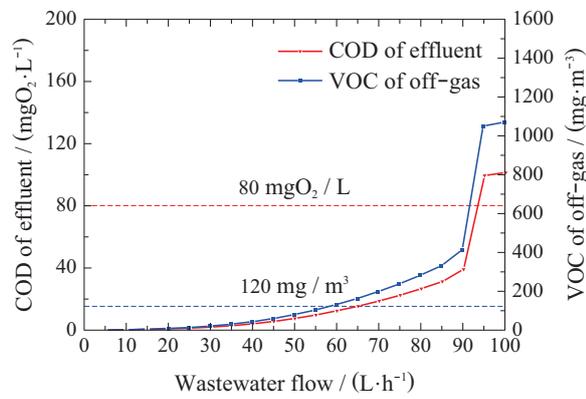
The control variables include: oxygen excess multiple K_1 (0~2); process wastewater flow F_1 (5 L/h~100 L/h); ratio of distillation residue to wastewater K_2 (0~0.1); fluidized bed temperature T_1 (300°C~380°C); Compared with the fluidized bed, the fixed bed increased temperature T_2 (0°C~40°C). Then, according to the boundary range, sensitivity analysis is carried out on the oxygen excess multiple, wastewater flow, ratio of distillation residue to wastewater, fluidized bed temperature, and fixed bed temperature. The results are shown in Fig. 4.

It is easy to be seen that the five selected decision variables have a significant impact on the degree of organic pollutant treatment, that is, the concentration of

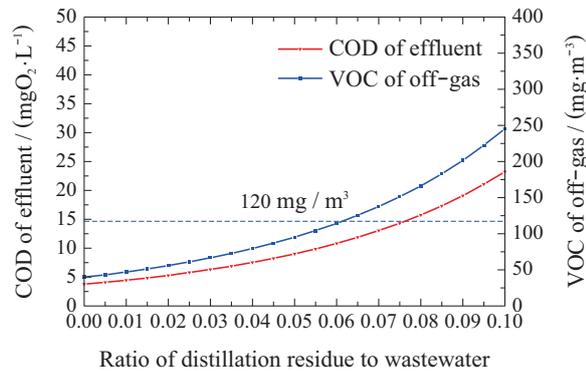
organic pollutants in the purified gas and the COD of the purified water. At the same time, the increase of any single flow or temperature variable in the above five control variables will lead to an increase on total energy consumption.



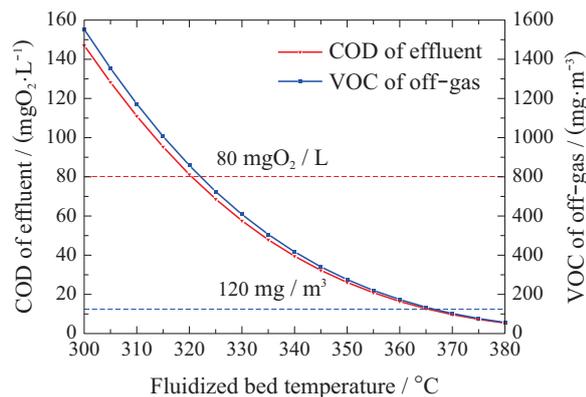
(a) Oxygen excess multiple



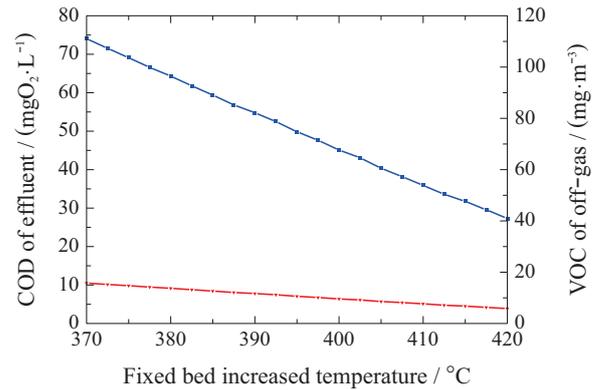
(b) Wastewater flow



(c) Ratio of distillation residue to wastewater



(d) Fluidized bed temperature



(e) Fixed bed increased temperature

Fig. 4 The results of sensitivity analysis

3.2 Optimization results

The population size, the number of maximum generations, crossover probability, mutation probability, crossover distribution index and mutation distribution index were set to 40, 120, 0.9, 0.1, 20, 20, respectively. It was found that the improved NSGA-II completed the operation after 6240.2 seconds. The final optimized operation results are shown in Fig. 5.

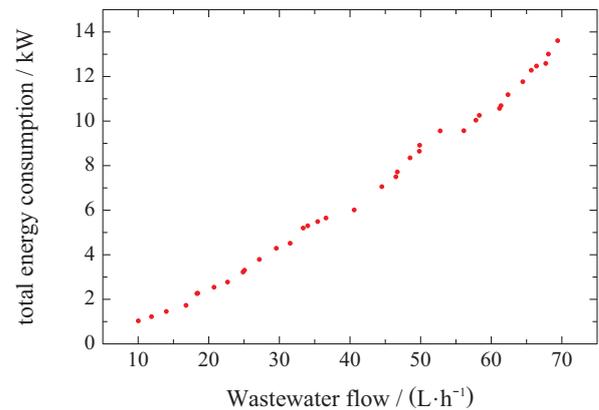


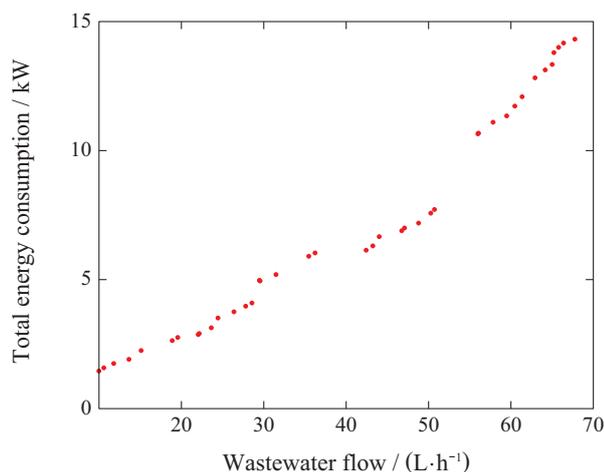
Fig. 5 The optimization results of improved NSGA-II

As the amount of wastewater treated increases, the corresponding minimum total energy consumption also increases. In addition to the 2 optimization goals, each point in the graph also corresponds to 5 control variables. Chemical engineers can select any one of Pareto optimal solutions in Fig. 5, adjust the control variables to the corresponding set values, and realize the process operation with the lowest energy consumption. Choosing the closest to the original wastewater flow, it can be found that the minimum total energy consumption has dropped from 14.34 kW to about 9.56 kW, which is a decrease of nearly 33%. Although the concentration of pollutants has increased, they are still within the range of emission standards. Based on the principles of non-dominated sorting and uniform distribution of solutions, 10 solutions are selected from the optimization results and listed in Table 4.

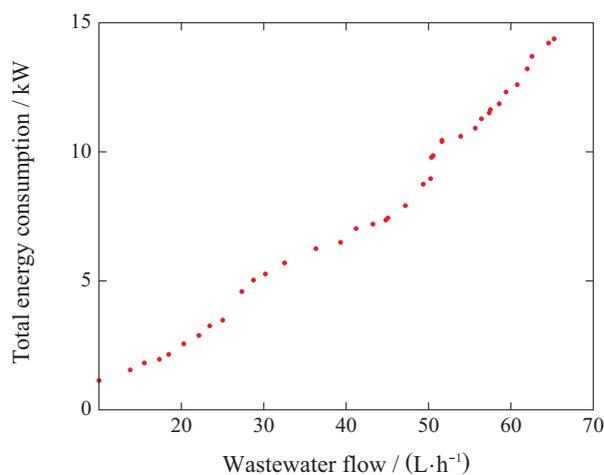
Table 4 10 Solutions in the Pareto optimal solution of the improved NSGA-II

Waste water flow/(L·h ⁻¹)	Total energy consumption/(kW)	Oxygen excess multiple	Ratio of residue -waste water	Fluidized bed temp/°C	Fixed bed increased temp/°C
10.0	1.0280	0.50	0.05245	372.308	40.0
16.775	1.7263	0.5183	0.05285	373.631	35.735
22.643	2.7726	0.6564	0.05197	370.665	35.001
29.540	4.2866	0.8231	0.05090	370.062	31.438
36.588	5.6466	1.2302	0.05577	372.938	13.890
44.501	7.0562	1.2692	0.05585	373.095	6.6348
52.748	9.5581	1.4209	0.05437	375.701	22.933
58.302	10.2525	1.6148	0.05865	379.052	25.344
62.353	11.1808	1.6311	0.05840	378.962	26.577
69.384	13.6062	1.8528	0.05943	379.859	26.444

We compared the optimization results of improved NSGA-II and original NSGA-II in the 50-th generation to test the speed of the improved optimization algorithm, as shown in Fig. 6.



(a) Original NSGA-II



(b) Improved NSGA-II

Fig. 6 Comparison between the improved NSGA-II and the original NSGA-II

According to the simulation, if you want to obtain optimization results similar to the improved algorithm, the original NSGA-II requires more genetic generation and more time. Therefore, for the multiphase organic pollutant treatment process, the improved NSGA-II can improve the optimization speed to a certain extent.

3.3 Dynamic control system

The two-stage fluidized/fixed bed process for treating multiphase organic pollutants is relatively complicated. Therefore, it is necessary to design effective plant-level control schemes using the top-down method for each part, including wastewater and distillation residue mixing, air and waste gas mixing, fluidized bed reactor, fixed bed reactor, heat exchanger and condenser. To achieve the purpose of automatic control of key control variables and safe, stable and energy-saving operation under steady-state optimized control variables. The plant-level control scheme for organic pollutant treatment is shown in Fig. 7.

The feeds need to maintain a certain ratio according to the optimized oxygen excess multiple and the ratio of distillation residue to wastewater, so a variable ratio control scheme is designed to ensure the heat balance. The fluidized bed and fixed bed need to reach the set reaction temperature under constant pressure, so the PI controllers are designed to ensure the stability of pressure and temperature.

In all control loops, the feed mixing is proportional control, and the other temperature and pressure controllers are PI controllers. The tuning of the PI controller parameters uses the Tyreus-Luyben tuning method. The closed loop gain K_c and integration time T_i is obtained using the relay feedback test. The parameters of all controllers are tuned according to their importance. The adjusted PI control parameters are shown in Table 5.

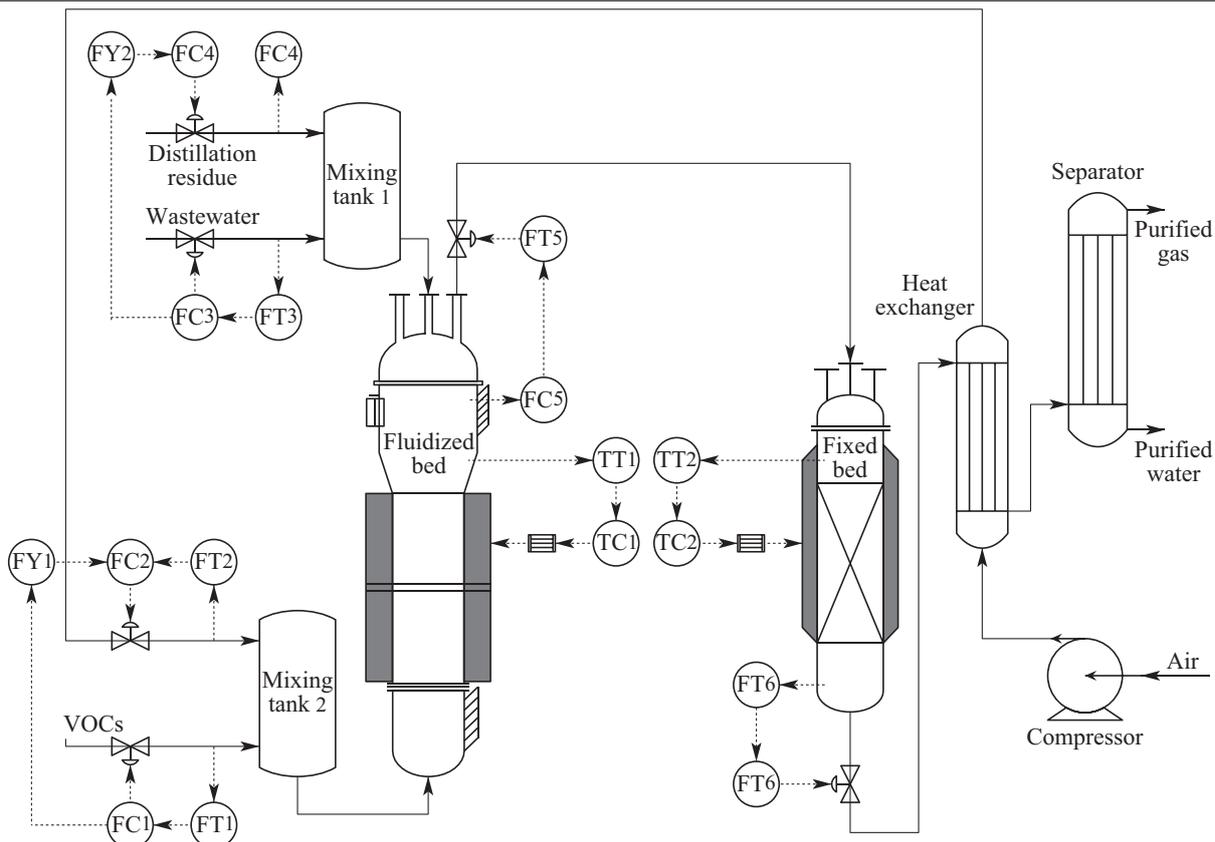
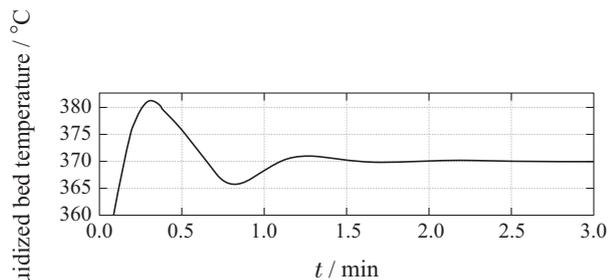
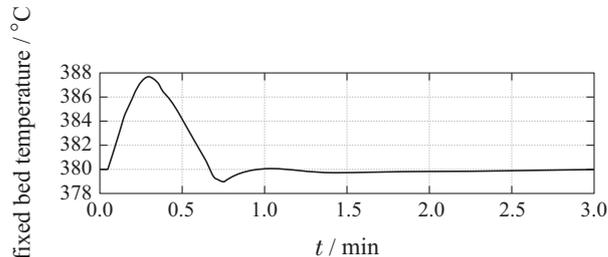


Fig. 7 The plant-level control scheme

To test the effectiveness of the optimization results and the proposed control scheme, a dynamic simulation system was established in this section based on Aspen Plus simulation. The plant-level control scheme is applied to establish the dynamic process simulation system using Aspen Dynamic model. The optimized values selected in the simulation is the control variables when the wastewater flow is 52.748 L/h. Temperature control is the core of the control system, so it is mainly tested. After 1 hour steady running with no disturbance, the dynamic system tracking ability and anti-disturbance ability were tested to verify the performance of the optimization and the control scheme. The default fluidized bed temperature is 350°C, the default fixed bed temperature is 380°C and the system is already in a steady state, then try to change the set temperature of the fluidized bed to 370°C, the tracking curve is shown in Fig. 8.



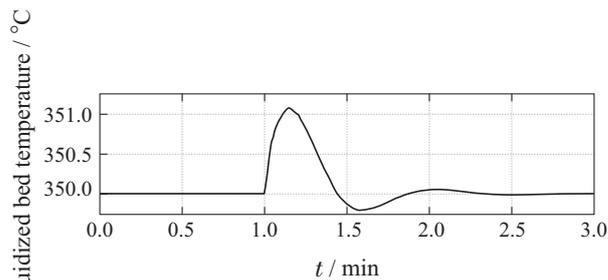
(a) Fluidized bed temperature



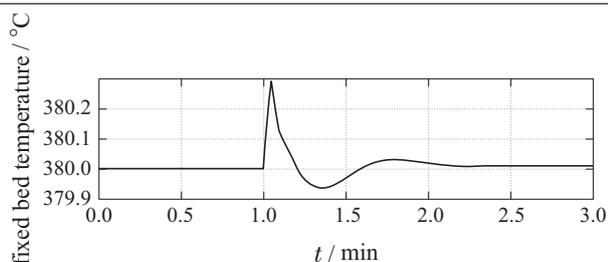
(b) Fixed bed temperature

Fig. 8 Fluidized bed temperature tracking curve

There are two main types of interference that may occur. The first is that fluctuations in the production process cause the concentration of a certain organic pollutant to rise or fall. Therefore, when the system is in a stable state, the concentration of organic pollutant $C_3H_4O_2$ in the wastewater is increased by 20%. The temperature change at this time is shown in Fig. 9.



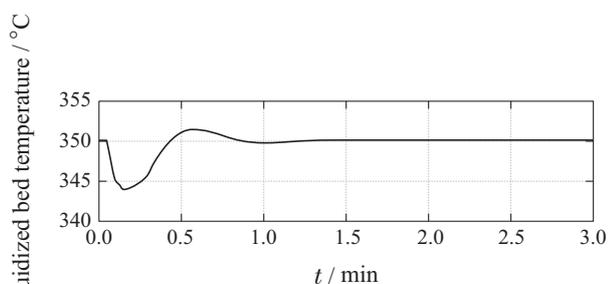
(a) Fluidized bed temperature



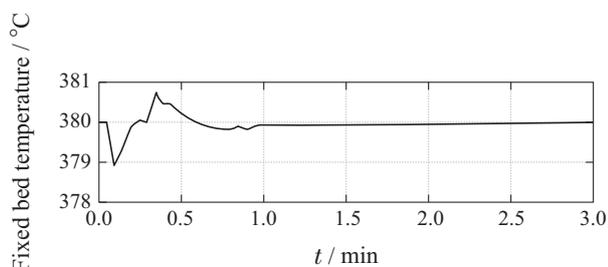
(b) Fixed bed temperature

Fig. 9 Concentration disturbance test

The second type of disturbance is mainly the change of organic pollutant flow and air flow. Due to the proportional control at the flows, the results after changes in various flows are relatively similar, so here we only consider the situation when the flow of the wastewater changes, and other flows vary with the flow of the wastewater. The waste water flow drops by 20%, and the temperature curve is shown in Fig. 10.



(a) Fluidized bed temperature



(b) Fixed bed temperature

Fig. 10 Flow disturbance test

A pilot experimental device for organic pollutant treatment has been developed based on the previously optimized operating variables and the control scheme. The design of control program includes hardware configuration, input signal reading and control scheme realization based on SIEMENS S7-200 SMART controller. According to the needs of the actual wastewater treatment volume, a set of optimal solutions with a wastewater flow rate of 29.54 L/h and a total energy consumption of 4.29 kW is selected. After setting the control parameters, it is found that the control parameters are basically stable at the set values. Then the voltage of the electric heater is detected to determine the power consumption of the heater when the device is running.

Since the noise is strong when the voltage is continuously measured, every 30 minutes, an average voltage is selected to plot. The electric heater voltage of the chemical bed and fixed bed is shown in Fig. 11.

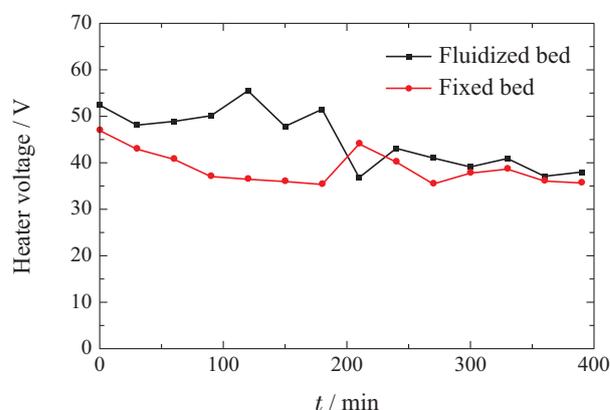


Fig. 11 Electric heater voltage for organic pollutant treatment device

According to the simulation results and experimental device, it can be seen that the designed temperature control system has good tracking performance, small overshoot and short response time. At the same time, it can better suppress the changes in fluidized bed temperature and fixed bed temperature when the concentration or flow suddenly changes. In the pollutant treatment process, the heating voltage fluctuates from 35 V to 55 V. Therefore, the fluidized bed and fixed bed electric heaters consume very little electricity during the process of pollutant power.

4 Conclusions

In this article, for the integrated treatment process of catalytic oxidation of organic pollutants in a two-stage fluidized/fixed bed system, Aspen Plus was used to establish a steady-state and dynamic simulation system. Based on this system, sensitivity analysis was performed on key control parameters. Then, the improved NSGA-II was used for multi-objective optimization of control variables. The chemical engineers could flexibly choose control variables according to the optimization results. Finally, the plant-level control scheme was designed and the dynamic performance and power consumption were tested in the simulation system and pilot experimental device. However, the optimization speed in this paper is still not fast enough, and the control scheme can still be improved. Therefore, the optimization and control are needed to be further researched.

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