

Modelling study of p-xylene oxidative side-reaction based on Bayesian filtering

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Abstract

The combustion loss model of HAC and PX was established based on Bayesian filtering. Moreover, this neural network model regarded the main adjustable process parameters (residence time, the concentration of Co, Mn, Br, reaction temperature and solvent ratio) as the independent variables and the total content of CO and CO₂ as the dependent variable. The simulation results show that the network is well performed. The effects of process operating parameters on HAC and PX combustion loss are analysed based on the model. Wherein, the effects of residence time, reaction temperature on the combustion loss are bigger, while the effects of other factors are relatively small.

Keywords: Mathematical model, PX oxidative side-reaction, Bayesian filtering, soft sensor

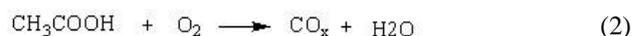
1 Introduction

PTA (PTA) is an important raw material of polyester fiber and plastic. Polyethylene terephthalate (PET) can be obtained from PTA and polyethylene glycol through a directly polycondensation process. As thermoplastic polyester, PET has extensive application in many areas. Meanwhile, as the largest output of dicarboxylic acid, terephthalic acid (TA)'s demand is great in the world. TA can be obtained from PX through a Liquid-phase catalytic oxidation action in AMOCO process, in which catalyst is Co and Mn, promoter is Br, and solvent is acetic acid (HAc) [1-2]. In addition to the main response, PX oxidative is a complex chemical reaction accompanied by a large number of side-reaction. Burning of HAc and PX is the principal side-reaction, whose main product is the carbon oxygen compounds, mainly as CO and CO₂ [3-5]. Besides the production losses of HAc and TA, by-products of HAc and PX oxidative side-reaction can lead to catalyst deactivation. Meanwhile, macromolecule side-reaction formed by the burning of PX can damage the quality of TA and PTA., and limit the mother liquor circulation. Thus not only caused a drop in the quality of products, but also caused an increase in the consumption of catalyst and promoter.

In this paper, a model of PX oxidative side-reaction was studied on the base of the plants in Yang Zi Petrochemistry Company (YPC) to provide basis for the optimization of operation parameters and the reform of production process.

2 Auth mechanism of PX oxidative side-reaction

PX oxidative reaction is in the condition of high temperature, excess oxygen and high-intensity stir. At the same time of main response, PX and HAc will partly combust to carbonic oxide (CO), carbonic dioxide (CO₂) and water according to the following reactive:



Generally, in the industrial operation condition, CO, CO₂ and methyl acetate are the by-products of the HAc's burning which losses HAc about 2% of its circulation. Carbon oxides and other by-products of PX burning make up about 2% of its feedstock. CO_x from the burning of HAc account for 60%, in which contain CO and CO₂ about 60%, while CO_x from the burning of PX is 40%, in which CO and CO₂ is 75%[6].

In the process of PX oxidation, a certain oxidation depth is requested in order to reach some conversion and absorption rate, especially reduce the concentration of 4-CBA. Over oxidation should be controlled because it can make HAc, 4-CBA and its by-products further oxidation to CO₂ and H₂O. The most important factors influent side-oxidation are: reaction temperature (x_1 , °C), solvent ratio (x_2 , Kg.HAc/Kg.PX), Co concentration (x_3 , wt%), Mn concentration (x_4 , wt%), Br concentration (x_5 , wt%) and reaction time (x_6 , s). These operation parameters not

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but control PX oxidation reaction, but also affect HAC and PX side-oxidation reaction [7-8].

3 Bayesian estimation of side-reaction

3.1 BAYESIAN ESTIMATION FIGURES AND TABLES

In this research, the content of CO and CO₂ was thought as a state estimation problem. The state-space model was utilized to describe this problem. Assume that the position state of the target is $(x_t|t=1,\dots,N)$, and the collection for each moment observation is $(z_t|t=1,\dots,N)$. Then the state of the target can be described by the motion equation and observation equation in formula 3:

$$\begin{cases} x_t = F(x_{t-1}) + \omega_t \\ z_t = H(x_t) + \nu_t \end{cases}, \quad (3)$$

where F is the impact parametric equation describing the model input;

H is the observation model, which describes the relationship between the observed quantity and model output;

ω_t is the noise, which is utilized to describe the uncertainty of movement;

ν_t is the observed noise, which is utilized to describe the uncertainty caused by outside interference and the noise of the detecting element.

Bayesian estimation is a state estimation method utilizing state priori distribution and the observation of likelihood function to determine the posterior probability distribution. For a first-order Markov process, assume that the observation of each moment is mutually independent. If the posterior distribution at time t-1 is $p(x_{t-1}|z_{t-1})$, the prior distribution at time t can be expressed as shown in formula 4:

$$p(x_t|z_{t-1}) = \int p(x_{t-1}|z_{t-1})p(x_t|x_{t-1})dx_{t-1}, \quad (4)$$

where $p(x_t|x_{t-1})$ is the transition probability density function, which is determined by F and the probability distribution of the noise $\omega_t(p(\omega_t))$ in motion equation 3. The definition is:

$$p(x_t|x_{t-1}) = \int p(\omega_t)\delta(x_t - F(x_{t-1}))d\omega_t, \quad (5)$$

wherein $\delta(\cdot)$ is the Dirac function. After the prior distribution $p(x_t|z_{t-1})$ is obtained, the state posterior distribution $p(x_t|z_t)$ could be expressed as:

$$p(x_t|z_t) = \frac{p(z_t|x_t)p(x_t|z_{t-1})}{\int p(z_t|x_t)p(x_t|z_{t-1})dx_t}, \quad (6)$$

where $p(z_t|x_t)$ is the observation likelihood function, which is determined by H and the probability distribution of the noise $\nu_t(p(\nu_t))$. The definition is:

$$p(z_t|x_t) = \int p(\nu_t)\delta(z_t - H(x_t))d\nu_t. \quad (7)$$

The above formula 4 and 5 constitute the prediction process of Bayesian estimation; formula 6 and 7 constitute the update process of Bayesian estimation. The two processes are determined by the equation and observation equation of state space model parameters in formula 3, respectively. The tracking of the posterior distribution of state x_t could be achieved through the iterative and recursive solution of above prediction and update process.

3.2 MODELING COX IN TAIL GAS OF SIDE-OXIDATION

The source of our data was the PTA production process data in a chemical plant. The model of paraxylene (PX) oxidation side reaction through soft sensor model provided the basis for production optimization of operating parameters and the transformation of the production process. The main factors that will affect the combustion side reaction are: the reaction temperature (x_1 , °C), solvent ratio (x_2 , Kg. HAC/Kg. PX), the concentration of cobalt catalyst (x_3 , wt%), the concentration of manganese catalyst (x_4 , wt%), the concentration of bromine accelerator (x_5 , wt%) and the residence time (x_6 , S). Regard these main factors after data pre-processing as the input data; the total content of CO and CO₂ (ΦCO_x) of the reactor exhaust gas as the output data y. Generally, 250 sets of data were obtained. 170 sets of data are the training data, and the remaining 80 sets of data are the current running data in the process, which were utilized to test the generalization capability of the model.

Further simulation analysis on the training results and the linear regression analysis of the output and target output of network simulation are shown in figure 1. The figure shows that the correlation coefficient is 0.963, which means that the performance of the network is satisfying.

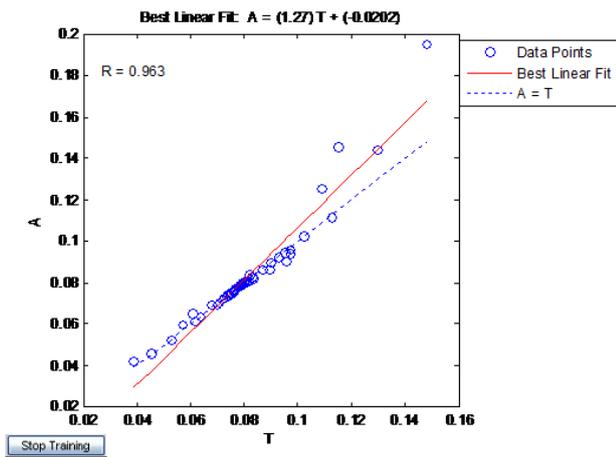


FIGURE 1 The analysis result of network output

3.3 THE COMBUSTION LOSS MODEL OF ACETIC ACID AND XYLENE

Establish the combustion loss model of acetic acid and PX based on the determined generation model of CO_x. The combustion loss model of acetic acid:

$$m_{HAc}^{consume} = \frac{m_{gas} \times x_{CO_x} \times \frac{x_{HAc}}{100} \times \frac{60}{1000}}{2 \times \frac{m_{HAc}^{CO_x}}{100} \times m_{CTA}}$$

The combustion loss model of PX is:

$$m_{PX}^{consume} = \frac{m_{gas} \times x_{CO_x} \times \frac{x_{PX}}{100} \times \frac{106}{1000}}{8 \times \frac{m_{PX}^{CO_x}}{100} \times m_{CTA}}$$

x_{CO_x} : the total content of generated CO and CO₂ of the exhaust gas

x_{HAc} : the percentage of CO_x generated by acetic acid (%)

$m_{HAc}^{CO_x}$: the percentage of CO_x accounting for the combustion product of acetic acid side reaction (%)

m_{gas} : the flow of exhaust gas from the reactor (ft³/hr)

m_{CTA} : CTA product yield (tons / Hr)

The molecular weight of acetic acid: 60 (g / mol)

$m_{HAc}^{consume}$: the amount of acetic acid loss (Kg / ton. CTA)

x_{PX} : the percentage of CO_x generated by PX (%)

$m_{PX}^{CO_x}$: the percentage of CO_x accounting for the combustion product of PX side reaction (%)

The molecular weight of PX: 106 (g / mol)

$m_{PX}^{consume}$: the amount of PX loss (Kg / ton CTA)

From the above text, the value of x_{HAc} , $m_{HAc}^{CO_x}$, x_{PX} and $m_{PX}^{CO_x}$ are known as 60%, 75%, 40% and 60%, respectively; m_{gas} and m_{CTA} were obtained through the actual production data.

4 Impact to side-oxidation of operating parameters

An impact model of operating parameters and side-oxidation of HAc and PX is presented. The impact of each operating parameters (Co, Mn, Br, reaction time, reaction temperature etc.) to side-oxidation is analysed in the model. Shows in Fig2~Fig6).

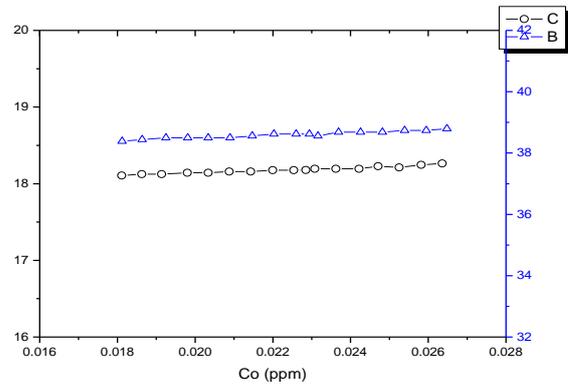


FIGURE 2 Effect of Co concentration on oxidative side-reaction

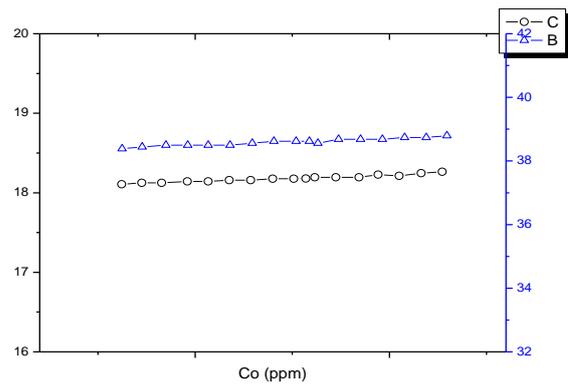


FIGURE 3 Effect of Mn concentration on oxidative side-reaction

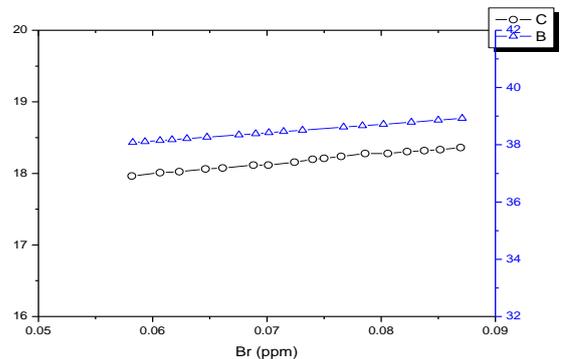


FIGURE 4 Effect of Br concentration on oxidative side-reaction

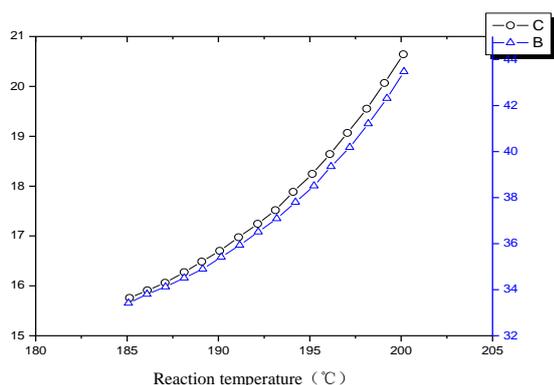


FIGURE 5 Effecton of reaction temperature on oxidative side-reaction

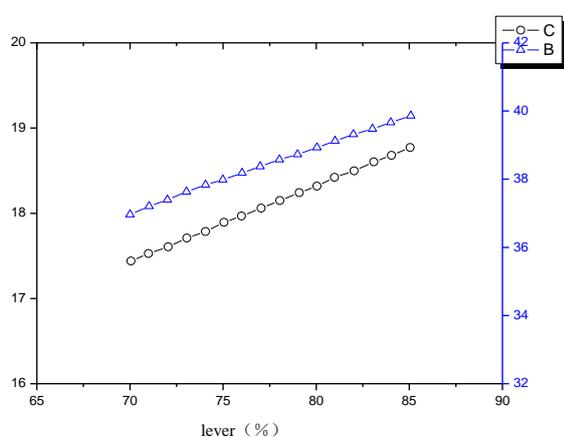


FIGURE 6 Effecton of lever on oxidative side-reaction

From the Figs, it can be seen that the impacton of reaction temperature is greatest, reaction time is greater, and others is less. Quantitative analysis of the impacton can be obtained from the figs, to guide the operating parameters' adjustment and optimization.

5 Conclusions

The model of p-xylene (PX) oxidative side-reaction was proposed in this paper. The Bayesian estimation technique was employed to model the relationship between the main operation conditions (i.e. residence time, reaction temperature and catalyst concentration) and the content of carbon dioxide in the reaction off-gas based on the factors. Furthermore, put forward the combustion expanse model of acetic acid and PX. Impacton of HAc and PX consume was analysed based on the network. Result is that the impacton of reaction temperature is greatest and others are less.

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